General theory of entanglement enhancement by external fields in spin chains

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Abstract

In the present thesis, we report several general properties of the enhancement of the entanglement by external fields.

First, we investigate the thermal entanglement of interacting two qubits. We maximize it by tuning a local Hamiltonian under a given interaction Hamiltonian. We prove that the optimizing local Hamiltonian takes a simple form which dose not depend on the temperature and that the corresponding optimized thermal entanglement decays as $1/(T \log T)$ at high temperatures. We also find that at low temperatures the thermal entanglement is maximum without any local Hamiltonians and that the second derivative of the maximized thermal entanglement changes discontinuously at the boundary between the high- and low-temperature phases.

Second, we investigate the maximized entanglement of indirectly interacting two spins, that is, through other spins. We present a necessary condition for the indirect interaction to give a non-zero maximized entanglement between the focused spins. We also prove that if the focused spins are separated by two spins, there is a critical temperature above which the maximized entanglement between the focused spins vanishes. Then, we numerically calculate the maximized entanglement between the end spins of three-spin chains and four-spin chains. We discover that the maximizing local fields on the end spins have asymmetric forms. In the three-spin chains, we attribute the entanglement enhancement to the asymmetry of the local fields qualitatively and quantitatively in terms of the magnons. In XX and XY four-spin chains, we find that the critical temperature shows qualitatively different behavior depending on the conservation of the angular momentum in the z direction.

Contents

Chapter 1

Introduction

In the present chapter, we introduce the theoretical motivation of the present research and the connection with the past works. We also give the fundamental knowledge for understanding the present thesis.

1.1 Motivation

The present thesis is devoted to the study of the enhancement of the quantum entanglement due to external fields. Over the last two decades, the quantum entanglement have attracted much attention. Numerous studies are devoted to understanding its properties and the discovery of the application to new technologies [1–5]. We especially focus on the problem of what cause the entanglement enhancement. In the following, we show the research background in more detail.

First, the quantum entanglement is one of the most essential properties which characterize the degree of the non-classicality. This was discovered by Schrödinger in early times [6] when the quantum physics was established. They also played an essential role in the solution of the EPR paradox [7], which had been one of the most puzzling problems for a long time. The entanglement reflects the non-locality; it makes it possible to create a correlation which cannot be explained by the classical theory [8]. Its existence are observed experimentally in 1981 by A. Aspect *et al.* [9–11]. Until the 1990s, however, specific properties of the entanglement had not been studied because it was not clear then how the quantum entanglement could be put to practical use. Since the quantum information thereby was developed [12], the research of the entanglement have been an essential task in order to understand its properties. Representative examples are the quantum computation [13], the quantum code [14] and so on. As another theoretical application, the entanglement is a useful physical quantity in phenomena in which the quantumness plays a crucial role. Indeed, many researches have suggested the relationship between the quantum phase transition and the entanglement [15,16]. These researches are under development, but they have much potential to bring essential principles. So far, many properties of the entanglement have been clarified and the corresponding experiments are following them. Because of the mathematical difficulties of the entanglement, however, there are still quite a few problems to be solved. From the theoretically and practically aspects, in particular, the generation and the enhancement of the entanglement are very important problem. Their properties often depend on individual systems and it is a tough problem to obtain general properties. We tackle this problem from the direction of using external fields. In the present research, we focus on the entanglement in thermal equilibrium states, including the ground states.

Second, we show the motivation of the present research of the entanglement enhancement by external fields. The first reason for that is that the strong entanglement is required in terms of practical applications [3]. Indeed, there are many researches about the generation of the strong entanglement [4]. Moreover, external fields are easily controllable and the entanglement by external fields is realizable. The second reason is that we can know from this research why the quantumness of the system can be increased by the change of external parameters. There are many examples on the quantum phenomena which occur after changes in external fields [4]. Until now, many studies have been reported on the entanglement in various quantum systems. However, little is known about the reasons why external fields can enhance the entanglement and the general properties of the entanglement enhancement.

Third, we show our final purposes of the present study:

1. To establish the general principles on the enhancement of the (bipartite and multipartite) entanglement.

2. To reveal the general properties of the thermal entanglement at high temperatures, especially the possibilities of the protection of the entanglement.

3. To clarify the general properties of the entanglement over a long distance, which is generated between a pair of spins separated far apart [17–19], as well as the method of generating it, if possible.

These problems are closely related to practical applications of the entanglement and the comprehension of the foundation of quantum physics. However, we have encountered several difficulties in working on these problems. The first one is the difficulty of the calculation of the entanglement [20]. As is shown in the following sections, the calculation of the entanglement needs the density matrix of the total system, which contains all degrees of freedom of the system. Because of this, it is very difficult to calculate the entanglement in complicated systems. The method of the calculation of the multipartite entanglement even has not been established; it goes without saying that we have to know the density matrix of the total system. At present, there are several measures [21–23] in the ground states which distill the essential properties of the multipartite entanglement. In the present thesis, we consider only the bipartite entanglement, for which we can use well-defined entanglement measures such as the concurrence and the negativity.

In order to work on the above problems, we discuss the entanglement maximized by local fields. This is the maximum value of the entanglement between a specific spin pair when we can arbitrarily modulate the local fields on the focused spins (Fig. 1.1). The research of the maximized entanglement is very suitable for the above three problems because of the following reasons:

1. Properties of the local fields which maximize the entanglement reflect the essence of the entanglement enhancement by the local fields.

2. It tells us the limit of the entanglement enhancement due to the local fields. If the maximized entanglement is equal to zero, it means that we cannot generate the entanglement with any local fields under the given condition.

In the present thesis, we mainly show the general properties of the maximized entanglement.

Now, we show the main achievement of the present thesis. We mainly work on the following two targets:

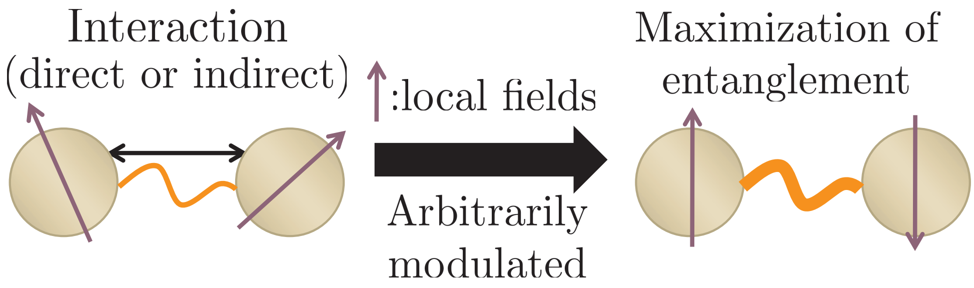


Figure 1.1: A schematic picture of the entanglement maximization. A specific two spins interact with each other directly or indirectly. The word "indirect" means that two spins interact via other spins. We can modulate the local fields on the focused two spins arbitrarily. We thereby define the maximum value of the entanglement as the maximized entanglement.

1. Maximization of the thermal entanglement of arbitrarily interacting two qubits (Chapter 2).

2. General properties of the maximized entanglement of indirectly interacting two spins (Chapter 3)

In Chapter 2, we consider the entanglement maximization problems of two spins which directly interact with each other. In this case, we can calculate the entanglement analytically to some extent. Therefore, we succeeded in obtaining the general properties for the arbitrary interactions. In Chapter 3, we consider the entanglement maximization problem of two spins indirectly interacting through other spins. In this case, unlike the case of the two spins, it is much more difficult to calculate the entanglement in arbitrary cases. Therefore, we focus on the possibilities of the generation of the entanglement and the properties of the entanglement enhancement in short spin chains.

1.2 List of publication

- T. Kuwahara and N. Hatano. Maximization of thermal entanglement of arbitrarily interacting two qubits. Physical Review A **83**, 062311 (2011)

(contains results presented in Chapter 2)

- T. Kuwahara. General properties of the maximized entanglement of indirectly interacting two spins. arXiv:quant-ph/12042337.

(contains results presented in Chapter 3)

1.3 Entanglement theory

In the present section, we overview basic knowledge on the quantum entanglement necessary to understand the present thesis.

1.3.1 Definition of the entanglement

First, we show the definition of the entanglement [24]. We discuss the pure state and the mixed state separately. Let us consider only the bipartite entanglement and assume that the quantum system consists of the spins 1 and 2. The general form of the pure state $|\psi\rangle$ is then given by

$$|\psi\rangle = s|\uparrow_1\uparrow_2\rangle + t|\uparrow_1\downarrow_2\rangle + u|\downarrow_1\uparrow_2\rangle + w|\downarrow_1\downarrow_2\rangle, \tag{1.1}$$

where $\{|\uparrow\rangle, |\downarrow\rangle\}$ are the base of each spin states. It is defined that the spins 1 and 2 are not entangled if and only if the following condition is satisfied:

$$|\psi\rangle = |\psi_1\rangle \otimes |\psi_2\rangle, \tag{1.2}$$

where $|\psi_1\rangle$ and $|\psi_2\rangle$ denote arbitrary states of the spins 1 and 2. In other words, if the states of the spins 1 and 2 cannot be decomposed into the direct product of states of each spin. we say that the state is entangled.

The above definition is applied only to pure states. Let us then discuss the mixed states. In order to discuss the mixed states, we consider the density matrix ρ_{12}, where elements are given by $\langle\uparrow_1\uparrow_2|\rho_{12}|\downarrow_1\downarrow_2\rangle$, for example. By diagonalizing the density matrix, we can denotes ρ_{12} as

$$\rho_{12} = \sum_{i=1}^{4} p_i |\psi_i\rangle\langle\psi_i|, \tag{1.3}$$

which consists of the four eigenstates $\{|\psi_i\rangle\}_{i=1}^4$ with the stochastic weights $\{p_i\}_{i=1}^4$. Generally speaking, we cannot decide the mixedness of the states uniquely. For example, let us define the states $\{|\tilde{\psi}_i\rangle\}_{i=1}^4$ as follows:

$$|\tilde{\psi}_i\rangle = \sum_{i=1}^{4} U_{ij}|\psi_j\rangle, \text{ for } j = 1, 2, 3, 4, \tag{1.4}$$

where U_{ij} is an arbitrary unitary matrix. The states $\{|\tilde{\psi}_i\rangle\}_{i=1}^4$ also give the expansion (1.3). We then define that the spins 1 and 2 are not entangled if and only if there is a unitary matrix U_{ij} which satisfies

$$\rho_{12} = \sum_{i=1}^{4} p_i \big(|\psi_{i1}\rangle \otimes |\psi_{i2}\rangle\big)\big(\langle\psi_{i1}| \otimes \langle\psi_{i2}|\big). \tag{1.5}$$

In other words, if the states of the spins 1 and 2 cannot be decomposed into a mixture of the non-entangled states, we say that the mixed state is entangled. The definition can be extended to any bipartite systems. However, it is usually difficult to judge whether the appropriate unitary matrix U_{ij} which reduces the density matrix to the form (1.5) exists or not in general cases. In the present thesis, we can completely determine the entanglement of the bipartite systems of 2×2 and 2×3.

1.3.2 Entanglement measure

Next, we show the quantification of the entanglement. An entanglement measure $E(\rho_{12})$ has to satisfy the following conditions [25]:

1. An entanglement measure $E(\rho_{12})$ projects the density matrix into a positive number.
2. If $E(\rho_{12})$ is equal to zero, the density matrix ρ_{12} is not entangled. In other words, it can be decomposed as Eq. (1.5).
3. An entanglement measure $E(\rho_{12})$ cannot be increased by LOCC, namely

$$E(\rho_{12}) \geq \sum_i p_i E\left(\frac{(A_i \otimes B_i)\rho_{12}(A_i^\dagger \otimes B_i^\dagger)}{\mathrm{tr}(A_i \otimes B_i)\rho_{12}(A_i^\dagger \otimes B_i^\dagger)} \right), \tag{1.6}$$

   where A_i and B_i are local operators that satisfy $\sum_{i=1} A_i A_i^\dagger \otimes B_i B_i^\dagger = I_{12}$ with I_{12} being the identity operator.

In addition, the condition of the convexity is sometimes demanded; namely,

$$E\left(\sum_i p_i \rho_i\right) \leq \sum_i p_i E(\rho_i). \tag{1.7}$$

In the bipartite system of 2×2, there are several measures which satisfy these conditions. In the following, we use three measures, namely, the concurrence, the negativity, and the determinant measure. The concurrence and the negativity satisfy the above conditions.

Concurrence

The concurrence $C(\rho_{12})$ is the most popular entanglement measure [26] . The concurrence is defined as follows;

$$C(\rho_{12}) \equiv \max(\lambda_1 - \lambda_2 - \lambda_3 - \lambda_4, 0), \tag{1.8}$$

where $\{\lambda_i\}_{i=1}^4$ are the eigenvalues of

$$\sqrt{\rho_{12}(\sigma_1^y \otimes \sigma_2^y)\rho_{12}^*(\sigma_1^y \otimes \sigma_2^y)} \tag{1.9}$$

in the non-ascending order $\lambda_1 \geq \lambda_2 \geq \lambda_3 \geq \lambda_4$.

It is often difficult to obtain the analytical forms of the concurrence, but it is easy in the case in which the density matrix has the form of the X-state

$$\rho_{12} = \begin{pmatrix} p_{\uparrow\uparrow} & 0 & 0 & F_2 \\ 0 & p_{\uparrow\downarrow} & F_1 & 0 \\ 0 & F_1^* & p_{\downarrow\uparrow} & 0 \\ F_2^* & 0 & 0 & p_{\downarrow\downarrow} \end{pmatrix}, \tag{1.10}$$

where the parameters $\{p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ are real numbers and the parameters and $\{F_1, F_2\}$ are complex numbers. The concurrence of this state is simply calculated as

$$C(\rho_{12}) = \max(|F_1| - \sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}, |F_2| - \sqrt{p_{\downarrow\uparrow}p_{\uparrow\downarrow}}, 0). \tag{1.11}$$

Negativity

Next, we introduce the negativity $N(\rho_{12})$ [27]. The negativity is defined as follows:

$$\begin{aligned} N(\rho_{12}) &\equiv ||\rho_{12}^{T_1}||_1 - 1 \\ &= \max(-2\lambda_-, 0), \end{aligned} \tag{1.12}$$

where $||\cdot||_1$ is the trace norm, λ_- is the minimum eigenvalue of $\rho_{12}^{T_1}$, which can be negative, and T_1 denotes the transpose operation with respect to only the spin 1; for example,

$$\begin{pmatrix} p_{\uparrow\uparrow} & F_3 & F_4 & F_2 \\ F_3^* & p_{\uparrow\downarrow} & F_1 & F_5 \\ F_4^* & F_1^* & p_{\downarrow\uparrow} & I_6 \\ F_2^* & F_5^* & F_6^* & p_{\downarrow\downarrow} \end{pmatrix}^{T_1} = \begin{pmatrix} p_{\uparrow\uparrow} & F_3 & F_4^* & F_2^* \\ F_3^* & p_{\uparrow\downarrow} & F_1^* & F_5^* \\ F_4 & F_1 & p_{\downarrow\uparrow} & F_6 \\ F_2 & F_5 & F_6^* & p_{\downarrow\downarrow} \end{pmatrix}. \tag{1.13}$$

The second equation of (1.12) comes from the fact that $\rho_{12}^{T_1}$ can have only one negative eigenvalue [28]. The advantage of this measure is that because it can be described as the minimum eigenvalue, it is easier to calculate than the concurrence; we can simply calculate the perturbation form of the negativity as

$$N(\rho_0 + \delta\rho) \simeq N(\rho_0) - 2\langle\phi_-|\delta\rho^{T_1}|\phi_-\rangle, \tag{1.14}$$

where we refer to the eigenstate corresponding to the negative eigenvalue of $\rho_0^{T_1}$ as $|\phi_-\rangle$. If the eigenstate of λ_- has the form of the Bell state, the negativity and the concurrence have an equal value [29] . In some cases, the logarithmic negativity $\log_2 ||\rho_{12}^{T_1}||_1$ is used instead of $N(\rho_{12})$, but it is known that $\log_2 ||\rho_{12}^{T_1}||_1$ does not satisfy the convexity [30] . We uses the negativity $N(\rho_{12})$ in the present thesis.

Determinant measure

We finally introduce the determinant measure $\pi(\rho_{12})$ [28]. It does not satisfy all the conditions for the entanglement measure, but it is easier to calculate than the concurrence and the negativity. The determinant measure is defined as

$$\pi(\rho_{12}) \equiv \begin{cases} 0, & \text{for } \rho_{12}^{T_1} \geq 0, \\ 2(|\det \rho_{12}^{T_1}|)^{1/4}, & \text{for } \rho_{12}^{T_1} < 0. \end{cases} \tag{1.15}$$

This entanglement measure is not a full entanglement monotone. However, it provides tight lower and upper bounds for other entanglement measures including the negativity and the concurrence. In addition, $\det \rho_{12}^{T_1}$ is expressed in the form of a polynomial and hence is much easier to maximize numerically than the concurrence and the negativity. Utilizing this measure, we tested Hypothesis 1 below by numerical optimization for various kinds of interaction at various temperatures and found it always satisfied.

1.3.3 Thermal entanglement

Next, we introduce the thermal entanglement. The thermal entanglement is defined as the entanglement in thermal equilibrium systems. The density matrix in thermal equilibrium is given by

$$\rho = \frac{e^{-\beta H_{\rm tot}}}{Z}, \tag{1.16}$$

where $Z = \mathrm{tr}(e^{-\beta H_{\mathrm{tot}}})$ is the partition function and $\beta = 1/kT$ with k the Boltzmann constant. The thermal fluctuation generally destroys the entanglement because it decreases the purity of the system; the purity is defined as $\mathrm{tr}\rho^2$. In some systems, however, it is discovered that the entanglement is enhanced by the thermal fluctuation [31, 32].

Chapter 2

Maximization of thermal entanglement of arbitrarily interacting two qubits

In the present chapter, we investigate the thermal entanglement of interacting two qubits. We maximize it by tuning a local Hamiltonian under a given interaction Hamiltonian. We prove that the optimizing local Hamiltonian takes a simple form which dose not depend on the temperature and that the corresponding optimized thermal entanglement decays as $1/(T \log T)$ at high temperatures. We also find that at low temperatures the thermal entanglement is maximum without any local Hamiltonians and that the second derivative of the maximized thermal entanglement changes discontinuously at the boundary between the high- and low-temperature phases.

2.1 Introduction

Quantum entanglement plays an essential role in quantum information processing [24]. Various kinds of investigation have been carried out to understand properties of entanglement for the last two decades [3,4]. The thermal entanglement [31], which is entanglement of thermal equilibrium states, is one of the important concepts because it shows us the effect of thermal fluctuations on entanglement. Thermal disturbances generally cause disentanglement and have serious effects on quantum information processing. Therefore, many schemes have been proposed to protect entanglement from thermal disturbances [33–42]. As one of these schemes, a lot of attention has been paid to methods based on manipulation of local Hamiltonians [33, 36, 38–42]; for example, in quantum spin systems, bipartite thermal entanglement can be enhanced by modulating external magnetic fields. In the present chapter, we focus on a simple question as to how much entanglement can be generated by optimizing the local Hamiltonian. We give a theoretical limit of entanglement enhancement by manipulation of the local Hamiltonians.

Relationships between the thermal entanglement and local parameters have been investigated especially in bipartite quantum spin systems [31, 38–46]. From these researches, behavior of the thermal entanglement under external magnetic fields may be understood in the cases of almost all interactions. However, little has been reported on the *maximization* problem of the thermal entanglement; in the case of the bipartite XY spin model, this problem has been solved only numerically [40]. Until now, there are no analytical approaches to optimizing the thermal entanglement of arbitrarily interacting two qubits.

In the present chapter, we will answer the following question: given a system of two qubits which interact via an arbitrary interaction Hamiltonian, how can we maximize the thermal entanglement between these two qubits by changing only the local Hamiltonian? A naive approach to this problem may be to solve the optimization problem numerically. However, this problem has six local parameters in total and the functional forms of entanglement measures such as the concurrence [26] and the negativity [27] are very complicated. Thus, for an arbitrary interaction, it is difficult to solve this optimization problem numerically. Therefore, we employ perturbation techniques and utilize symmetric properties in order to determine the optimizing local Hamiltonian analytically. In this way, for all kinds of interaction, we give general properties of the optimized entanglement.

Our main results are the following:

1. We find that at low temperatures the thermal entanglement is maximum without any local Hamiltonians, whereas at high temperatures it is maximized by non-zero local fields. We refer to the former temperature range as the low-temperature phase and the latter temperature range as the high-temperature phase. The secondary differentiation of the maximized entanglement is discontinuous at the phase boundary.

2. In the high-temperature phase, the functional form of the optimizing local Hamiltonian is independent of the temperature; only the coefficients depend on the temperature.

3. The optimized entanglement, enhanced by a local Hamiltonian in the high-temperature phase, decreases with increasing temperature as $1/(T \log T)$.

4. If the interaction Hamiltonian has no degeneracy of its eigenvalues, the entanglement is maximized without local Hamiltonians over a finite range of the low-temperature phase.

5. If the interaction Hamiltonian has degeneracy, the low-temperature phase shrinks to the zero-temperature point. The optimizing local Hamiltonian becomes infinitesimal and the optimized entanglement becomes full in the low-temperature limit.

The present chapter is organized as follows. In Section II, we state the main problem after symmetry consideration. In Section III, we give the main theorems on the entanglement optimization. In Section IV, we show numerical results of the optimizing local parameters, the boundary temperatures and the singularity at the phase boundary. We also argue that the two phases appear because of competition between the purifying effect and the decoupling effect both of the local Hamiltonian. Finally, in Section V, a discussion concludes the chapter.

2.2 Entanglement optimization problem

First, we set the fundamental framework of the present problem. We consider a $2 \otimes 2$ system of σ_1 and σ_2. The most general form of the Hamiltonian of this system is given as follows:

$$\begin{aligned} H_{\mathrm{tot}} &\equiv H_{\mathrm{int}} + H_{\mathrm{LO}}, \\ H_{\mathrm{int}} &\equiv \sum_{i,j=x,y,z} J_{ij}\sigma_1^i \otimes \sigma_2^j, \\ H_{\mathrm{LO}} &\equiv \sum_{i=x,y,z} (h_1^i \sigma_1^i \otimes I + h_2^i I \otimes \sigma_2^i), \end{aligned} \tag{2.1}$$

where $\{\sigma_1^i\}_{i=x,y,z}$ and $\{\sigma_2^i\}_{i=x,y,z}$ are the Pauli matrices, H_{int} is an interaction Hamiltonian, and H_{LO} is a local Hamiltonian. We assume that $\{J_{ij}\}_{i,j=x,y,z}$ are fixed and independent of the temperature, whereas we can change the parameters $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\}$ arbitrarily.

We parametrize the local fields in the polar coordinates as

$$\{h_1^i\}_{i=x,y,z} = \{h_1 \sin\theta_1 \cos\phi_1, h_1 \sin\theta_1 \sin\phi_1, h_1 \cos\theta_1\},$$
$$\{h_2^i\}_{i=x,y,z} = \{h_2 \sin\theta_2 \cos\phi_2, h_2 \sin\theta_2 \sin\phi_2, h_2 \cos\theta_2\}. \tag{2.2}$$

Hereafter, we use the parametrization

$$h \equiv \frac{h_1 + h_2}{2}, \quad \zeta \equiv \frac{h_1 - h_2}{h_1 + h_2}, \tag{2.3}$$

where $-1 \leq \zeta \leq 1$ and $h \geq 0$; in other words,

$$h_1 = (1 + \zeta)h, \quad h_2 = (1 - \zeta)h. \tag{2.4}$$

Then, the four eigenvalues of H_{LO} are

$$\{-2h, -2\zeta h, 2\zeta h, 2h\}, \tag{2.5}$$

where we define the corresponding eigenstates as $\{|--\rangle, |-+\rangle, |+-\rangle, |++\rangle\}$.

The density matrix in thermal equilibrium is

$$\rho = \frac{e^{-\beta H_{\mathrm{tot}}}}{Z}, \tag{2.6}$$

where $Z = \mathrm{tr}(e^{-\beta H_{\mathrm{tot}}})$ is the partition function and $\beta = 1/(kT)$ with k the Boltzmann constant. In order to quantify entanglement, we adopt the negativity [27] as an entanglement measure. The negativity is defined as the trace norm of a partially transposed density matrix:

$$N(\rho) \equiv ||\rho^{T_1}||_1 - 1$$
$$= \max(-2\lambda_-, 0), \tag{2.7}$$

where $||\ ||_1$ is the trace norm, T_1 denotes the transpose with respect to only σ_1, and λ_- is the minimum, possibly negative eigenvalue of ρ^{T_1}. The second equation of (2.7) comes from the fact that ρ^{T_1} can have only one negative eigenvalue, if any [28]. Thus, the present entanglement optimization problem is equivalent to finding the values of $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\}$ which maximize $N(\rho)$ for an arbitrary fixed interaction H_{int}.

Note that the maximizing local fields $H_{\mathrm{LO}}^{\mathrm{op}}$ generally depend on the temperature T, or on the inverse temperature $\beta = 1/(kT)$. This is because we tune the local fields at a fixed temperature β. Let us then define the high-temperature limit, in which we mostly develop the argument. In our high-temperature limit, we make β tend to zero with the parameters in H_{int} fixed. In other words, we have $\beta||H_{\mathrm{int}}|| \to 0$ in the high-temperature limit, where $||H_{\mathrm{int}}||$ is the norm of H_{int}. On the other hand, we let the maximizing local fields depend on β as we take the limit $\beta \to 0$. Hence, $\beta||H_{\mathrm{LO}}^{\mathrm{op}}||$ can even diverge in our high-temperature limit.

Before presenting our main results on the entanglement optimization, we prove the following Lemma 1 to simplify the present entanglement optimization problem.

Lemma 1. By local unitary transformations of H_{int}, we can eliminate the interaction parameters $\{J_{ij}\}_{i\neq j}$ and reduce it to the form

$$H_{\mathrm{int}} = \sum_{i=x,y,z} J_i \sigma_1^i \otimes \sigma_2^i. \tag{2.8}$$

We can also choose the parameters $\{J_x, J_y, J_z\}$ such that $\{J_x, J_y\} \geq J_z \geq 0$ or $0 \geq J_z \geq \{J_x, J_y\}$.

In spin-1/2 systems, this means that we can transform any interactions including the Dzyaloshinskii-Moriya (DM) [42,47,48] interaction into a ferromagnetic or an anti-ferromagnetic Heisenberg exchange interaction.

Proof. We can prove this Lemma by applying a singular value decomposition [?] to the matrix $(\hat{J})_{ij} \equiv J_{ij}$. In this case, the singular value decomposition $\hat{U}\hat{J}\hat{W}$ is performed by 3×3 real orthogonal transformations $\hat{U}$ and $\hat{W}$ of the three-dimensional spin spaces of the spins 1 and 2, respectively. A real orthogonal transformation is composed of rotation and inversion operations, but inversion operations cannot be performed by unitary transformations. Therefore, we remove the inversion operations from the real orthogonal transformation of the singular value decomposition and restrict ourselves only to the rotation operations, which means $\det \hat{U} = \det \hat{W} = 1$. In other words, we rotate $\vec{\sigma}_1 = \{\sigma_1^x, \sigma_1^y, \sigma_1^z\}$ with $\hat{U}$ and $\vec{\sigma}_2 = \{\sigma_2^x, \sigma_2^y, \sigma_2^z\}$ with $\hat{W}$. Then we can transform $\{J_{ij}\}_{i,j=x,y,z}$ into the antiferromagnetic cases $\{J_x, J_y\} \geq J_z \geq 0$ or the ferromagnetic cases $0 \geq J_z \geq \{J_x, J_y\}$, with the other elements $\{J_{ij}\}_{i\neq j}$ put to zero. Here, we choose the z-axis so that $|J_z|$ is the least of $\{|J_i|\}_{i=x,y,z}$. Thus, Lemma 1 is proved.

Let us show an example in the case of the XXZ model with the z-component of the DM interaction. The Hamiltonian of such a system is given by

$$
\begin{aligned}
H_{\text{int}} \equiv & J\sigma_1^x \otimes \sigma_2^x + J\sigma_1^y \otimes \sigma_2^y + J_z\sigma_1^z \otimes \sigma_2^z \\
& + D_z(\sigma_1^x \otimes \sigma_2^y - \sigma_1^y \otimes \sigma_2^x),
\end{aligned}
\tag{2.9}
$$

where J and J_z are the real coupling coefficients and D_z is the z-component of the DM interaction. In the case of $J = 1$, $J_z = -2$ and $D_z = 1$, we can transform $\{J_x, J_y, J_z, D_z\}$ into $\{-\sqrt{2}, -\sqrt{2}, -2, 0\}$ by rotating the spin 1 by 135 degrees around the z-axis, namely into

$$
H_{\text{int}} = -\sqrt{2}\sigma_1^x \otimes \sigma_2^x - \sqrt{2}\sigma_1^y \otimes \sigma_2^y - 2\sigma_1^z \otimes \sigma_2^z. \tag{2.10}
$$

This is an antiferromagnetic Heisenberg interaction. To attain this result, first, the singular value decomposition transforms $\{J_x, J_y, J_z, D_z\}$ into $\{\sqrt{2}, \sqrt{2}, 2, 0\}$ by rotating the spin 1 by -45 degrees around the z-axis and inverting the z-axis of the spin. Next, we remove the inversion of the z-axis because it cannot be performed by unitary operations, and thereby transform $\{J_x, J_y, J_z, D_z\}$ into $\{\sqrt{2}, \sqrt{2}, -2, 0\}$. By changing the rotation angle from -45 to 135, we can invert the signs of J_x and J_y and arrive at $\{J_x, J_y, J_z, D_z\} = \{-\sqrt{2}, -\sqrt{2}, -2, 0\}$.

In the following, based on Lemma 1, we always use the diagonalized form (2.8) of the interaction parameters with $\{J_x, J_y\} \geq J_z \geq 0$ or $0 \geq J_z \geq \{J_x, J_y\}$. We now have all the necessary ingredients to state the main theorems.

2.3 Main analytical results

In the present section, we analytically discuss the optimization problem. The main conclusion of the present section is that the negativity is maximized by the parameters $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\text{op}}, 0, 0, -h_{\text{op}}\}$. The optimizing parameter h_{op} must be very large at high temperatures, whereas it may be 0 at low temperatures.

2.3.1 Optimization in the case the high-temperature limit $\beta \to 0$

Let us first discuss the optimization problem in the high-temperature limit.

Theorem 1. In the high-temperature limit $\beta \to 0$, the local parameters which maximize the entanglement $N(\rho)$ are given in the form of $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$. The optimizing value $h_{\rm op}$ is given by the solution of the following equation:

$$e^{2h'_{\rm op}} \simeq \frac{8h'^2_{\rm op}}{\beta|J_x + J_y|} \quad \text{as} \quad \beta \to 0, \tag{2.11}$$

where

$$h'_{\rm op} \equiv \beta h_{\rm op}, \tag{2.12}$$

and the optimized entanglement $N_{\rm op}$ asymptotically behaves as

$$\begin{aligned} N_{\rm op}(\rho) &\simeq \beta \frac{|J_x + J_y|}{2h'_{\rm op}} - 2e^{-2h'_{\rm op}} \\ &\simeq \beta \frac{|J_x + J_y|}{2h'_{\rm op}} \left(1 - \frac{1}{2h'_{\rm op}}\right) \quad \text{as} \quad \beta \to 0, \end{aligned} \tag{2.13}$$

where we used Eq. (2.11) upon moving from the first line to the second line.

The leading order of the solution of Eq. (2.11) is given by

$$h'_{\rm op} \simeq \frac{1}{2} \log \frac{1}{\beta} + \frac{1}{2} \log \frac{8}{|J_x + J_y|}. \tag{2.14}$$

We can thereby obtain the following simpler asymptotes:

$$h_{\rm op} \simeq \frac{\log 1/\beta}{2\beta} \quad \text{as} \quad \beta \to 0, \tag{2.15}$$

$$N_{\rm op}(\rho) \simeq \beta \frac{|J_x + J_y|}{\log 1/\beta} \quad \text{as} \quad \beta \to 0. \tag{2.16}$$

That is, the optimizing value $h_{\rm op}$ depends only on the temperature and the optimized negativity decays in the form $1/(T \log T)$ in the limit $\beta \to 0$. In Appendix A, we compare the asymptotes of Eqs. (2.11) and (2.13) with those of Eqs. (2.15) and (2.16)

Proof. We prove Theorem 1 in the following steps. First, we prove in Lemma 2 that the optimizing local parameter $h_{\rm op}$ is greater than or equal to $(\log 1/\beta)/(2\beta)$ in the high-temperature limit and the optimized thermal state is nearly a pure state. The entanglement of the state comes from perturbations to the pure state. Then, we calculate the negativity approximately by perturbation method in Lemma 4. Using this expression, we finally solve the maximization problem for each local parameter.

First, we determine a lower bound of the optimizing value $h_{\rm op}$ and prove that the optimized thermal state is a nearly pure state. For this purpose, we prove the following Lemma 2.

Lemma 2. A necessary condition for the existence of the entanglement in the high-temperature limit under a fixed interaction Hamiltonian $H_{\rm int}$ is given by

$$\beta h > \frac{\log 1/\beta}{2} \quad \text{as} \quad \beta \to 0. \tag{2.17}$$

This Lemma 2 shows that $(\log 1/\beta)/(2\beta)$ is a lower bound of the optimizing value of $h_{\rm op}$.

Proof. We firstly prove that we need a non-zero value of βh for the existence of the entanglement in the high-temperature limit $\beta \to 0$. In other words, we need h at least of order

$1/\beta$. In order to show this, we consider a general necessary condition for the existence of the entanglement given by [50]

$$\lambda_1 \geq \lambda_3 + 2\sqrt{\lambda_2\lambda_4} \geq 3\lambda_4, \tag{2.18}$$

where $\{\lambda_\mu\}_{\mu=1}^4$ are the eigenvalues of the density matrix ρ in the non-ascending order ($\lambda_1 \geq \lambda_2 \geq \lambda_3 \geq \lambda_4$). Let us define the eigenvalues of $H_{\text{tot}} = H_{\text{LO}} + H_{\text{int}}$ as $\{E_\mu\}_{\mu=1}^4$ in the non-descending order ($E_1 \leq E_2 \leq E_3 \leq E_4$). Equation (3.3) gives the eigenvalues of ρ as $\{e^{-\beta E_\mu}/Z\}_{\mu=1}^4$, and therefore the inequality (2.18), or $e^{-\beta E_1} \geq 3e^{-\beta E_4}$, gives

$$\beta(E_4 - E_1) \geq \log 3. \tag{2.19}$$

Here, H_{int} is a constant matrix and hence $\beta H_{\text{int}} \to 0$ as $\beta \to 0$. If we let H_{LO} be of the same order as H_{int}, the left-hand side of (2.19) would vanish in the limit $\beta \to 0$ and (2.18) would not be satisfied. Therefore, we have to make H_{LO} much greater than H_{int}, and then the eigenvalues of H_{tot} should converge to those of H_{LO}, $\{-2h, -2\zeta h, 2\zeta h, 2h\}$ in the limit $\beta \to 0$. With $E_1 \to -2h$ and $E_4 \to 2h$, the inequality (2.19) reduces to the following inequality:

$$\beta h \geq \frac{\log 3}{4}. \tag{2.20}$$

This inequality means that we need a non-zero value of βh in the high-temperature limit $\beta \to 0$. In other words, we need to make h grow as $1/\beta$ at least, in order for the entanglement to exist in the limit $\beta \to 0$.

Next, we derive an approximation of the density matrix, and then obtain Eq. (2.17) by utilizing the Peres-Horodecki criterion [51, 52], which is a necessary and sufficient condition for the existence of the entanglement. In the present optimization problem, we fix H_{int} to a constant matrix, and therefore we have $\beta H_{\text{int}} \to 0$ in the high-temperature limit. We thereby work in the first-order approximation with respect to βH_{int}:

$$\begin{aligned} Z\rho &= e^{-H'_{\text{LO}} - \beta H_{\text{int}}} \\ &\simeq e^{-H'_{\text{LO}}} - \beta \int_0^1 e^{-(1-x)H'_{\text{LO}}} H_{\text{int}} e^{-xH'_{\text{LO}}} dx \\ &= e^{-H'_{\text{LO}}} - \beta \sum_{\mu,\nu} f_{\mu\nu} \langle\mu|H_{\text{int}}|\nu\rangle |\mu\rangle\langle\nu|, \end{aligned} \tag{2.21}$$

where $Z = \text{tr}(e^{-H'_{\text{LO}} - \beta H_{\text{int}}})$, and we let $H'_{\text{LO}} = \beta H_{\text{LO}}$ with $h' = \beta h$ as well as

$$f_{\mu\nu} = \begin{cases} \frac{e^{-E'_\nu} - e^{-E'_\mu}}{E'_\mu - E'_\nu}, & \text{for } E'_\mu \neq E'_\nu, \\ e^{-E'_\mu}, & \text{for } E'_\mu = E'_\nu. \end{cases} \tag{2.22}$$

Here, $\{E'_\mu\}_{\mu=1}^4$ are the eigenvalues of $H'_{\text{LO}} = \beta H_{\text{LO}}$, $\{-2h', -2\zeta h', 2\zeta h', 2h'\}$, and $\{|\mu\rangle\}_{\mu=1}^4$ are the corresponding eigenstates, $\{|{-}{-}\rangle, |{-}{+}\rangle, |{+}{-}\rangle, |{+}{+}\rangle\}$.

We then utilize the necessary and sufficient condition for the existence of the entanglement, $\det \rho^{T_1} < 0$. This has been proved [28] to be equivalent to the Peres-Horodecki criterion [51,52]. In the following discussion, among the various terms of the expansion of $\det \rho^{T_1}$, we compare the values of the products including off-diagonal elements (POD) with that of the product of the diagonal elements (PD), which has a positive value. Then a necessary condition for $\det \rho^{T_1} < 0$ is that POD is greater than or of the same order as the PD.

To analyze the order of the PD and the PODs, we express ρ^{T_1} in the basis $\{|\mu\rangle\}_{\mu=1}^4$ and focus on the main terms for $\zeta \neq 0$ and $\zeta \neq \pm 1$:

$$
\begin{aligned}
&Z\rho^{T_1} \\
&\xrightarrow{\beta\to 0} \begin{pmatrix} e^{2h'} & a_{12}\frac{\beta e^{2h'}}{h'} & a_{31}\frac{\beta e^{2h'}}{h'} & a_{32}\frac{\beta e^{2\zeta h'}}{h'} \\ a_{21}\frac{\beta e^{2h'}}{h'} & e^{2\zeta h'} & a_{41}\frac{\beta e^{2h'}}{h'} & a_{42}\frac{\beta e^{2\zeta h'}}{h'} \\ a_{13}\frac{\beta e^{2h'}}{h'} & a_{14}\frac{\beta e^{2h'}}{h'} & e^{-2\zeta h'} & a_{34}\frac{\beta e^{-2\zeta h'}}{h'} \\ a_{23}\frac{\beta e^{2\zeta h'}}{h'} & a_{24}\frac{\beta e^{2\zeta h'}}{h'} & a_{43}\frac{\beta e^{-2\zeta h'}}{h'} & e^{-2h'} \end{pmatrix},
\end{aligned}
\tag{2.23}
$$

where $\{a_{ij}\}$ are determined from Eqs. (2.21) and (2.22) and are constants of order 1. Note that on the diagonal of Eq. (2.23), the second term in Eq. (2.21) is neglected in comparison to the first term. Then we compare the orders of the PODs with that of the PD. The PD is given by $e^{2h'}e^{2\zeta h'}e^{-2\zeta h'}e^{-2h'} = 1$, whereas each POD includes at least two off-diagonal elements. The maximum of the absolute value of the PODs is of order $e^{4h'}\beta^2/h'^2$, which comes from the product $-e^{2h'} \times a_{41}\frac{\beta e^{2h'}}{h'} \times a_{14}\frac{\beta e^{2h'}}{h'} \times e^{-2h'}$. Therefore, it is necessary for $\det \rho^{T_1} < 0$ that $e^{4h'}\beta^2/h'^2$ is greater or of order 1, which is the order of PD. By taking the logarithm of $e^{4h'}\beta^2/h'^2$, we can obtain the following inequality as a necessary condition:

$$
\begin{aligned}
\beta h = h' \geq& \frac{\log 1/\beta}{2} + \frac{\log h'}{2} \\
\geq& \frac{\log 1/\beta}{2} + \frac{1}{2}\log\left(\frac{\log 1/\beta}{2} + \frac{\log h'}{2}\right) \\
\geq& \frac{\log 1/\beta}{2} + \frac{1}{2}\log\left(\frac{\log 1/\beta}{2} + \frac{1}{2}\log\left(\frac{\log 3}{4}\right)\right) \\
>& \frac{\log 1/\beta}{2},
\end{aligned}
\tag{2.24}
$$

where we utilized (2.20) in deriving the third inequality and used the fact $\beta \to 0$ in deriving the last inequality. Thus, Lemma 2 is proved for $\zeta \neq 0$ and $\zeta \neq \pm 1$. For $\zeta = 0$ or $\zeta = \pm 1$, some of the eigenvalues of H'_{LO} are degenerate, which means that E'_μ can be equal to E'_ν in Eq. (2.22), and $Z\rho^{T_1}$ is not of the same form as that of Eq. (2.23). However, the inequality (2.24) still holds as is proved in Appendix B.

We now consider the negativity (2.7) in the range given by (2.17). We first show in the following Lemma 3 that the optimized negativity in the cases of $\zeta = \pm 1$ is not large enough.

Lemma 3. In the cases of $\zeta = \pm 1$, the optimized negativity satisfies the following:

$$
\frac{N_{\mathrm{op}}(\rho, \zeta = \pm 1)}{\beta} \xrightarrow{\beta \to 0} 0. \tag{2.25}
$$

This lemma shows that the optimized negativity in the cases of $\zeta = \pm 1$ is of a higher order of β. Indeed, we numerically confirmed in the cases of $\zeta = \pm 1$ that the entanglement exists, but its amplitude is of order β^2.

Proof. Let us prove Eq. (2.25) in the case of $\zeta = 1$. The proof for $\zeta = -1$ is almost the same.

We start from the main term of $Z\rho^{T_1}$ for $\zeta = 1$ in the representation in the basis $\{|\mu\rangle\}_{\mu=1}^4$:

$$
\begin{aligned}
&Z\rho^{T_1}\\
&\xrightarrow{\beta\to 0}\begin{pmatrix} e^{2h'} & a_{12}\beta e^{2h'} & a_{31}\frac{\beta e^{2h'}}{h'} & a_{32}\frac{\beta e^{2h'}}{h'} \\ a_{21}\beta e^{2h'} & e^{2h'} & a_{41}\frac{\beta e^{2h'}}{h'} & a_{42}\frac{\beta e^{2h'}}{h'} \\ a_{13}\frac{\beta e^{2h'}}{h'} & a_{14}\frac{\beta e^{2h'}}{h'} & e^{-2h'} & a_{34}\beta e^{-2h'} \\ a_{23}\frac{\beta e^{2h'}}{h'} & a_{24}\frac{\beta e^{2h'}}{h'} & a_{43}\beta e^{-2h'} & e^{-2h'} \end{pmatrix},
\end{aligned}
\tag{2.26}
$$

where we used the fact that at $\zeta = 1$ the eigenvalues of H'_{LO} in Eq. (2.22) are degenerate as $\{E'_1, E'_2, E'_3, E'_4\} = \{2h', 2h', -2h', -2h'\}$.

In order to optimize the negativity, we necessarily consider the region $h' = \beta h > (\log 1/\beta)/2$ as is given in Lemma 2. Therefore, we can use the fact $e^{2h'} > \beta^{-1}$ in (2.26). Of the elements of the matrix (2.26), the $(1, 1)$ and $(2, 2)$ elements are of order β^{-1} or greater, whereas the $(3, 3)$, $(4, 4)$, $(3, 4)$ and $(4, 3)$ elements are of order β^1 or less. The other elements are approximately of order 1. We therefore break up the matrix (2.26) in the form

$$
\begin{aligned}
\rho^{T_1} = &\frac{1}{Z}\begin{pmatrix} e^{2h'} & 0 & 0 & 0 \\ 0 & e^{2h'} & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}\\
&+\frac{1}{Z}\begin{pmatrix} 0 & a_{12}\beta e^{2h'} & a_{31}\frac{\beta e^{2h'}}{h'} & a_{32}\frac{\beta e^{2h'}}{h'} \\ a_{21}\beta e^{2h'} & 0 & a_{41}\frac{\beta e^{2h'}}{h'} & a_{42}\frac{\beta e^{2h'}}{h'} \\ a_{13}\frac{\beta e^{2h'}}{h'} & a_{14}\frac{\beta e^{2h'}}{h'} & 0 & 0 \\ a_{23}\frac{\beta e^{2h'}}{h'} & a_{24}\frac{\beta e^{2h'}}{h'} & 0 & 0 \end{pmatrix} + O(\beta^2),
\end{aligned}
\tag{2.27}
$$

where $Z \simeq 2e^{2h'} + 2e^{-2h'} \simeq 2/\beta$, and therefore the first term is the dominant term of order 1, whereas the second term is of order β^1. The eigenvalues of the dominant term are $\{e^{2h'}/Z, e^{2h'}/Z, 0, 0\}$ and the corresponding eigenstates are $\{|--\rangle, |-+\rangle, |+-\rangle, |++\rangle\}$. A negative eigenvalue can appear when the degeneracy of the two zero eigenvalues of the states $|+-\rangle$ and $|++\rangle$ is resolved by perturbation. Then, the level repulsion between them makes one of them positive and the other negative. However, the first-order perturbation of the second term of Eq. (2.27) dose not resolve the degeneracy of the zero eigenvalues. Therefore, the negative eigenvalue must be produced in a higher order of β in the case of $\zeta = 1$. Thus, Lemma 3 is proved. We focus on the cases $\zeta \neq \pm 1$ hereafter.

Using the lower bound (2.17) of the optimizing parameter h_{op}, we next prove that the optimized thermal state is a nearly pure state in the cases of $\zeta \neq \pm 1$. For this purpose, we consider the eigenstates of the perturbed density matrix. We define the perturbed eigenstates of $\beta H_{\mathrm{tot}} = H'_{\mathrm{LO}} + \beta H_{\mathrm{int}}$ as $\{|--'\rangle, |-+'\rangle, |+-'\rangle, |++'\rangle\}$ corresponding to the eigenstates $\{|--\rangle, |-+\rangle, |+-\rangle, |++\rangle\}$ of H'_{LO}, respectively, and their eigenvalues as $\{2h' - \beta\delta\epsilon_1, 2\zeta h' - \beta\delta\epsilon_2, -2\zeta h' - \beta\delta\epsilon_3, -2h' - \beta\delta\epsilon_4\}$, where $\{\delta\epsilon_i\}_{i=1}^4$ are the perturbative changes due to H_{int}, which are of order 1. Then the density matrix is given by the summation over these four states. In the high-temperature limit $\beta \to 0$, the mixing ratio $\{\lambda_{--'}, \lambda_{-+'}, \lambda_{+-'}, \lambda_{++'}\}$ of the states $\{|--'\rangle, |-+'\rangle, |+-'\rangle, |++'\rangle\}$ are

$$
\begin{aligned}
&\{\lambda_{--'}, \lambda_{-+'}, \lambda_{+-'}, \lambda_{++'}\}\\
=&\frac{1}{Z}\{e^{2h'-\beta\delta\epsilon_1}, e^{2\zeta h'-\beta\delta\epsilon_2}, e^{-2\zeta h'-\beta\delta\epsilon_3}, e^{-2h'-\beta\delta\epsilon_4}\},
\end{aligned}
\tag{2.28}
$$

where

$$Z = e^{2h' - \beta\delta\epsilon_1} + e^{2\zeta h' - \beta\delta\epsilon_2} + e^{-2\zeta h' - \beta\delta\epsilon_3} + e^{-2h' - \beta\delta\epsilon_4}. \tag{2.29}$$

In the region $h' > (\log 1/\beta)/2$, which is the lower bound of $h'_{\rm op}$, we have

$$\begin{aligned}
\frac{\lambda_{-+'}}{\lambda_{--'}} &= e^{-2(1-\zeta)h' + \beta\delta\epsilon_1 - \beta\delta\epsilon_2} < \beta^{1-\zeta} e^{-\beta\delta\epsilon_1 + \beta\delta\epsilon_2}, \\
\frac{\lambda_{+-'}}{\lambda_{--'}} &= e^{-2(1+\zeta)h' + \beta\delta\epsilon_1 - \beta\delta\epsilon_3} < \beta^{1+\zeta} e^{-\beta\delta\epsilon_1 + \beta\delta\epsilon_3}, \\
\frac{\lambda_{++'}}{\lambda_{--'}} &= e^{-2h' + \beta\delta\epsilon_1 - \beta\delta\epsilon_4} < \beta^{2} e^{-\beta\delta\epsilon_1 + \beta\delta\epsilon_4}.
\end{aligned} \tag{2.30}$$

Since the right-hand sides of the inequalities vanish in the limit $\beta \to 0$, we deduce that the optimized thermal state is a nearly pure state of $|--'\rangle$ in the high-temperature limit $\beta \to 0$ when $\zeta \neq \pm 1$.

Next, we perturbatively calculate the negativity in the cases of $\zeta \neq \pm 1$. Since the optimized state is a nearly pure state of $|--'\rangle$, we regard the other contributions $\{|-+'\rangle, |+-'\rangle, |++'\rangle\}$ as perturbation:

$$\begin{aligned}
\rho_0 &= |--'\rangle\langle --'|, \\
\delta\rho &= \sum_{\{i=-+',+-',++'\}} \lambda_i \rho_i,
\end{aligned} \tag{2.31}$$

where $\{|--'\rangle, |-+'\rangle, |+-'\rangle, |++'\rangle\}$ are the eigenstates of $\beta H_{\rm tot} = H'_{\rm LO} + \beta H_{\rm int}$ as has been stated. In order to calculate the negativity approximately, we derive the expression for the perturbation of the negativity caused by an infinitesimal variation of the density matrix.

Lemma 4. When the negativity has a non-zero value, the first-order perturbation of the negativity is given by

$$N(\rho_0 + \delta\rho) \simeq N(\rho_0) - 2\langle \phi_- | \delta\rho^{T_1} | \phi_- \rangle, \tag{2.32}$$

where we refer to the eigenstate corresponding to the negative eigenvalue of $\rho_0^{T_1}$ as $|\phi_-\rangle$.

Proof. The non-zero negativity is given by the negative eigenvalue λ_- of the partial transpose of the density matrix, $\rho_0^{T_1}$, as is defined in (2.7). Because of the linearity of the partial transpose, if ρ_0 changes into $\rho_0 + \delta\rho$, $\rho_0^{T_1}$ also changes into $\rho_0^{T_1} + \delta\rho^{T_1}$. Moreover, the eigenstate of $\rho_0^{T_1}$ corresponding to λ_- is not degenerate because λ_- is the only possible negative eigenvalue [28]. Then, from the general perturbation theory for λ_-, we have Eq. (2.32) in the first order.

From Eqs. (2.31) and (2.32), we can calculate the negativity in the present case of $\zeta \neq \pm 1$ as

$$\begin{aligned}
&N\left(\sum_{i=1}^{4} \lambda_i \rho_i\right) \\
=&N(|--'\rangle) - \sum_{\{i=-+',+-',++'\}} \left(2\lambda_i \langle \phi_- | \rho_i^{T_1} | \phi_- \rangle + O(\lambda_i^2)\right).
\end{aligned} \tag{2.33}$$

The state $|--'\rangle$ and its negativity $N(|--'\rangle)$ are calculated in the first order of the perturbation $H'_{\rm LO} \to H'_{\rm LO} + \beta H_{\rm int}$. The zeroth-order eigenstates and eigenvalues are $\{|--\rangle, |-+\rangle, |+-\rangle, |++\rangle\}$

and $\{-2h' - 2\zeta h', 2\zeta h', 2h'\}$, respectively. The first-order eigenstate for the state $|--\rangle$ is then given by

$$|--'\rangle = |--\rangle + \beta n_1|-+\rangle + \beta n_2|+-\rangle + \beta n_3|++\rangle + O(\beta^2), \tag{2.34}$$

where

$$\begin{aligned} n_1 &= \frac{\langle -+|H_{\mathrm{int}}|--\rangle}{-2(1-\zeta)h'}, \\ n_2 &= \frac{\langle +-|H_{\mathrm{int}}|--\rangle}{-2(\zeta+1)h'}, \\ n_3 &= \frac{\langle ++|H_{\mathrm{int}}|--\rangle}{-4h'}. \end{aligned} \tag{2.35}$$

Note that the normalization factor of the state $|--'\rangle$ is $1 + O(\beta^2)$. The matrix representation of $\rho_0^{T_1} = \left(|--'\rangle\langle --'|\right)^{T_1}$ is therefore given in the basis of $\{|--\rangle, |-+\rangle, |+-\rangle, |++\rangle\}$ as follows by ignoring the terms of $O(\beta^2)$:

$$\left(|--'\rangle\langle --'|\right)^{T_1} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} + \beta \begin{pmatrix} 0 & n_1^* & n_2 & 0 \\ n_1 & 0 & n_3 & 0 \\ n_2^* & n_3^* & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \tag{2.36}$$

The zeroth-order eigenvalues of $\rho_0^{T_1}$ are $\{1, 0, 0, 0\}$. The negative eigenvalue emerges when the degeneracy of the first and second zero eigenvalues resolve in the first order of β. The third zero eigenvalue remains to be zero. The eigenvalues are then given by $\{1, \beta|n_3|, -\beta|n_3|, 0\}$ in the first order and hence the negative eigenvalue $-\beta|n_3|$ gives the negativity

$$\begin{aligned} N(|--'\rangle) &= 2\beta|n_3| \\ &= \beta\frac{|\langle ++|H_{\mathrm{int}}|--\rangle|}{2h'} + O(\beta^2), \end{aligned} \tag{2.37}$$

The corresponding eigenstate $|\phi_-\rangle$ is given by

$$|\phi_-\rangle = \frac{1}{\sqrt{2}}\Big(|-+\rangle - \frac{n_3}{|n_3|}|+-\rangle\Big) + O(\beta)|--\rangle. \tag{2.38}$$

Similarly, we have

$$\begin{aligned} \rho_{-+'}^{T_1} &= \left(|-+'\rangle\langle -+'|\right)^{T_1} = |-+\rangle\langle -+| + O(\beta), \\ \rho_{+-'}^{T_1} &= \left(|+-'\rangle\langle +-'|\right)^{T_1} = |+-\rangle\langle +-| + O(\beta), \\ \rho_{++'}^{T_1} &= \left(|++'\rangle\langle ++'|\right)^{T_1} = |++\rangle\langle ++| + O(\beta), \end{aligned} \tag{2.39}$$

as well as

$$\begin{aligned} \lambda_{-+'} &= \frac{e^{2\zeta h' - \beta\delta\epsilon_2}}{Z} \\ &\simeq e^{-2(1-\zeta)h'}(1+\beta\delta\epsilon_1)(1-\beta\delta\epsilon_2)(1-e^{-2(1-|\zeta|)h'}) \\ &= e^{-2(1-\zeta)h'} + O(\beta^{2-\zeta-|\zeta|}), \\ \lambda_{+-'} &= \frac{e^{-2\zeta h' - \beta\delta\epsilon_3}}{Z} \simeq e^{-2(1+\zeta)h'} + O(\beta^{2+\zeta-|\zeta|}), \\ \lambda_{++'} &= \frac{e^{-2h' - \beta\delta\epsilon_4}}{Z} \simeq e^{-4h'} + O(\beta^{3-|\zeta|}), \end{aligned} \tag{2.40}$$

where we used Eq. (2.29) for Z. Note that the first term of each of $\lambda_{-+'}$, $\lambda_{+-'}$ and $\lambda_{++'}$ is of order $\beta^{1-\zeta}$, $\beta^{1+\zeta}$ and β^2 or less, respectively, in the range of (2.17), $h' > (\log 1/\beta)/2$. By substituting Eqs. (2.37)–(2.40) in Eq. (2.33), we have

$$N\left(\sum_{i=1}^{4}\lambda_i\rho_i\right) \simeq \beta\frac{|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|}{2h'} - e^{-2(1-\zeta)h'} - e^{-2(1+\zeta)h'} + O(\beta^{2(1-|\zeta|)}) \tag{2.41}$$

for $\zeta \neq \pm 1$.

Because the matrix element $|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|$ is independent of h' and ζ, we can solve the maximization problem of Eq. (2.41) as follows. First, to maximize the negative terms in Eq. (2.41), we must put $\zeta = 0$. Then, by differentiating Eq. (2.41) with h', we have the optimizing parameter h'_{op} as a solution of

$$e^{2h'_{\mathrm{op}}} \simeq \frac{8h'^2_{\mathrm{op}}}{\beta|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|}. \tag{2.42}$$

The optimized negativity is then given by

$$\begin{aligned} N_{\mathrm{op}}(\rho) &\simeq \beta\frac{|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|}{2h'_{\mathrm{op}}} - 2e^{-2h'_{\mathrm{op}}} \\ &\simeq \beta\frac{|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|}{2h'_{\mathrm{op}}}\left(1 - \frac{1}{2h'_{\mathrm{op}}}\right), \end{aligned} \tag{2.43}$$

where we used Eq. (2.42) upon moving from the first line to the second line. This is the result for $\zeta \neq \pm 1$. From Lemma 3, we see that the optimized negativity (2.43) in the case of $\zeta = 0$ is larger in the limit $\beta \to 0$ than the one (2.25) in the cases of $\zeta = \pm 1$.

The other optimizing parameters to be fixed are $\{\theta_1, \phi_1, \theta_2, \phi_2\}$. Let us see how these parameters affect the value of (2.43). These parameters affect the matrix element $|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|$ and hence the value of (2.43) directly as well as indirectly through h'_{op} given by Eq. (2.42). We can write down the solution of Eq. (2.42) in terms of Lambert's W function [53], which is defined as a solution of

$$x = W(x)e^{W(x)}, \tag{2.44}$$

because we can cast Eq. (2.42) into the form

$$(-h'_{\mathrm{op}})e^{-h'_{\mathrm{op}}} = -\sqrt{\frac{\beta|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|}{8}}. \tag{2.45}$$

The appropriate solution of Eq. (2.42) is given by

$$h'_{\mathrm{op}} \simeq -W_{-1}\left(-\sqrt{\frac{\beta|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|}{8}}\right), \tag{2.46}$$

where $W_{-1}(x)$ is the branch of $W(x)$ satisfying $W_{-1}(x) \leq -1$ in the domain $-1/e < x < 0$ [53]. The function $-W_{-1}(-x)$ is a monotonically decreasing function of x in the domain $0 < x < 1/e$. Therefore, maximizing the element $|\langle{+}{+}|H_{\mathrm{int}}|{-}{-}\rangle|$ with respect to the parameters $\{\theta_1, \phi_1, \theta_2, \phi_2\}$ brings h'_{op} to its minimum within the range $h'_{\mathrm{op}} > 1$. Since the factor

$$\frac{1}{2h'_{\mathrm{op}}}\left(1 - \frac{1}{2h'_{\mathrm{op}}}\right) \tag{2.47}$$

in Eq. (2.43) is a decreasing function of h'_{op} for $h'_{\text{op}} > 1$, minimizing h'_{op} within the range $h'_{\text{op}} > 1$ brings the factor (2.47) to its maximum. To summarize, the element $|\langle{++}|H_{\text{int}}|{--}\rangle|$ increases the value of (2.43) not only directly but also through h'_{op} indirectly.

The next task is then to find the parameters $\{\theta_1, \phi_1, \theta_2, \phi_2\}$ that maximize the matrix element $|\langle{++}|H_{\text{int}}|{--}\rangle|$ in Eq. (2.43). The eigenstates of the one-qubit part $\sum_{i=x,y,z} h^i \sigma^i$ of the local Hamiltonian H_{LO} are given by

$$
\begin{aligned}
|+\rangle &= \cos\frac{\theta}{2}|0\rangle + e^{i\phi}\sin\frac{\theta}{2}|1\rangle, \\
|-\rangle &= -\sin\frac{\theta}{2}|0\rangle + e^{i\phi}\cos\frac{\theta}{2}|1\rangle,
\end{aligned} \tag{2.48}
$$

where we define $|0\rangle$ and $|1\rangle$ as the eigenstates of σ^z and represent $\{h^i\}_{i=x,y,z}$ as

$$
\{h\sin\theta\cos\phi, h\sin\theta\sin\phi, h\cos\theta\} \tag{2.49}
$$

in the polar coordinate. We can thereby express the eigenstates $|{++}\rangle$ and $|{--}\rangle$ of H_{LO} in the forms

$$
\begin{aligned}
|{++}\rangle = &\cos\frac{\theta_1}{2}\cos\frac{\theta_2}{2}|00\rangle + \cos\frac{\theta_1}{2}\sin\frac{\theta_2}{2}e^{i\phi_2}|01\rangle \\
&+ \sin\frac{\theta_1}{2}\cos\frac{\theta_2}{2}e^{i\phi_1}|10\rangle + \sin\frac{\theta_1}{2}\sin\frac{\theta_2}{2}e^{i(\phi_1+\phi_2)}|11\rangle, \\
|{--}\rangle = &\sin\frac{\theta_1}{2}\sin\frac{\theta_2}{2}|00\rangle - \sin\frac{\theta_1}{2}\cos\frac{\theta_2}{2}e^{i\phi_2}|01\rangle \\
&- \cos\frac{\theta_1}{2}\sin\frac{\theta_2}{2}e^{i\phi_1}|10\rangle + \cos\frac{\theta_1}{2}\cos\frac{\theta_2}{2}e^{i(\phi_1+\phi_2)}|11\rangle.
\end{aligned} \tag{2.50}
$$

We therefore have the matrix element $\langle{++}|H_{\text{int}}|{--}\rangle$ in the following form:

$$
\begin{aligned}
&\langle{++}|H_{\text{int}}|{--}\rangle \\
=&J_z\sin\theta_1\sin\theta_2 + (J_x - J_y)\Big[\sin^2\frac{\theta_1}{2}\sin^2\frac{\theta_2}{2}e^{-i(\phi_1+\phi_2)} \\
&+\cos^2\frac{\theta_1}{2}\cos^2\frac{\theta_2}{2}e^{i(\phi_1+\phi_2)}\Big] \\
&-(J_x + J_y)\Big[\cos^2\frac{\theta_1}{2}\sin^2\frac{\theta_2}{2}e^{i(\phi_1-\phi_2)} \\
&+\sin^2\frac{\theta_1}{2}\cos^2\frac{\theta_2}{2}e^{i(-\phi_1+\phi_2)}\Big].
\end{aligned} \tag{2.51}
$$

In the cases of $\{J_x, J_y\} \geq J_z \geq 0$ and $0 \geq J_z \geq \{J_x, J_y\}$, the upper bound of $|\langle{++}|H_{\text{int}}|{--}\rangle|$ is given by

$$
|\langle{++}|H_{\text{int}}|{--}\rangle| \leq |J_x + J_y|. \tag{2.52}
$$

We prove this inequality in the cases of $J_x \geq J_y \geq J_z \geq 0$; we can prove the other cases in the same way. First, $|\langle ++|H_{\rm int}|--\rangle|$ satisfies the following inequality:

$$\begin{aligned}
&|\langle ++|H_{\rm int}|--\rangle| \\
\leq &|J_z| \sin\theta_1 \sin\theta_2 \\
&+ |J_x - J_y| \left(\sin^2 \frac{\theta_1}{2} \sin^2 \frac{\theta_2}{2} + \cos^2 \frac{\theta_1}{2} \cos^2 \frac{\theta_2}{2} \right) \\
&+ |J_x + J_y| \left(\cos^2 \frac{\theta_1}{2} \sin^2 \frac{\theta_2}{2} + \sin^2 \frac{\theta_1}{2} \cos^2 \frac{\theta_2}{2} \right) \\
= &|J_z| \sin\theta_1 \sin\theta_2 + |J_x - J_y| \frac{1 + \cos\theta_1 \cos\theta_2}{2} \\
&\qquad + |J_x + J_y| \frac{1 - \cos\theta_1 \cos\theta_2}{2}.
\end{aligned} \tag{2.53}$$

By utilizing the fact that $J_x \geq J_y \geq J_z \geq 0$, the inequality (2.53) reduces to

$$\begin{aligned}
|\langle ++|H_{\rm int}|--\rangle| \leq &J_x - J_y \cos\theta_1 \cos\theta_2 + J_z \sin\theta_1 \sin\theta_2 \\
\leq &|J_x + J_y|.
\end{aligned} \tag{2.54}$$

The inequality (2.52) becomes an equality when we choose $\{\theta_1, \theta_2, \phi_1, \phi_2\}$ as $\{0, \pi, 0, 0\}$ for example, or in the Cartesian coordinate $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h, 0, 0, -h\}$. Then, the optimizing local parameters are given in the form of $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$. Moreover, Eqs. (2.11) and (2.13) can be given by substituting $|\langle ++|H_{\rm int}|--\rangle|$ with $|J_x + J_y|$ in Eqs. (2.42) and (2.43).

Finally, the leading order of Lambert's W function $-W_{-1}(-x)$ is $\log x$ [53]. Therefore, the leading order of Eq. (2.46) gives Eq. (2.14), which then results in Eqs. (2.15) and (2.16). This completes the proof of Theorem 1.

2.3.2 Optimization at arbitrary temperatures

It is difficult to generalize Theorem 1 to arbitrary temperatures. However, we can present the following Theorem 2. Let us now parametrize the local fields as follows:

$$\begin{aligned}
&\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} \\
=&\{h_1^x, h_1^y, h^z(1+\xi), h_2^x, h_2^y, -h^z(1-\xi)\},
\end{aligned} \tag{2.55}$$

or

$$h^z \equiv \frac{h_1^z - h_2^z}{2}, \quad \xi \equiv \frac{h_1^z + h_2^z}{h_1^z - h_2^z}. \tag{2.56}$$

Theorem 2. When we express the negativity as a function of the local parameters $\{h_1^x, h_1^y, h^z(1+\xi), h_2^x, h_2^y, -h^z(1-\xi)\}$, the following equation holds at arbitrary temperatures:

$$\begin{aligned}
&\frac{\partial N}{\partial h_1^x} = \frac{\partial N}{\partial h_2^x} = \frac{\partial N}{\partial h_1^y} = \frac{\partial N}{\partial h_2^y} = \frac{\partial N}{\partial \xi} = 0 \\
&\text{at } \{h_1^x, h_1^y, h_2^x, h_2^y, \xi, h^z\} = \{0, 0, 0, 0, 0, h\}.
\end{aligned} \tag{2.57}$$

This theorem means that the form of the optimizing local parameters in the high-temperature limit, $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$, also gives an extremal value of the negativity at arbitrary temperatures.

Proof. To prove this theorem, we firstly calculate the perturbation of the negativity due to an infinitesimal variation of the local parameters at arbitrary temperatures. If it always vanishes, Eq. (2.57) is proved. We first derive the perturbation of the density matrix due to an infinitesimal variation of the local parameters, from $\{0, 0, h, 0, 0, -h\}$ to $\{\delta h_1^x, \delta h_1^y, h(1 + \delta\xi), \delta h_2^x, \delta h_2^y, -h(1 - \delta\xi)\}$. This means the perturbation of the form

$$H_{\text{tot}} = H_{\text{tot}}^{\text{op}} + \delta\mathcal{H}_{\text{LO}}, \tag{2.58}$$

where

$$H_{\text{tot}}^{\text{op}} \equiv \sum_{i=x,y,z} J_i \sigma_1^i \otimes \sigma_2^i + h(\sigma_1^z \otimes I - I \otimes \sigma_2^z) \tag{2.59}$$

is the total Hamiltonian with the local parameters $\{0, 0, h, 0, 0, -h\}$ and

$$\begin{aligned}\delta\mathcal{H}_{\text{LO}} \equiv \sum_{i=x,y} &(\delta h_1^i \sigma_1^i \otimes I + \delta h_2^i I \otimes \sigma_2^i) \\ &+ h\delta\xi(\sigma_1^z \otimes I + I \otimes \sigma_2^z)\end{aligned} \tag{2.60}$$

is the infinitesimal variation of the local Hamiltonian. Equation (2.21) gives the perturbation of the density matrix $\delta\rho$ as

$$\begin{aligned}\delta\rho &= \frac{e^{-\beta(H_{\text{tot}}^{\text{op}} + \delta\mathcal{H}_{\text{LO}})}}{Z + \delta Z} - \frac{e^{-\beta H_{\text{tot}}^{\text{op}}}}{Z} \\ &= -\frac{\delta Z}{Z}\rho_{\text{op}} - \frac{\beta}{Z}\int_0^1 e^{-\beta(1-x)H_{\text{tot}}^{\text{op}}} \delta\mathcal{H}_{\text{LO}} e^{-\beta x H_{\text{tot}}^{\text{op}}} dx,\end{aligned} \tag{2.61}$$

where $\rho_{\text{op}} = e^{-\beta H_{\text{tot}}^{\text{op}}}/Z$ and

$$\delta Z = \text{tr}\left(-\beta \int_0^1 e^{-\beta(1-x)H_{\text{tot}}^{\text{op}}} \delta\mathcal{H}_{\text{LO}} e^{-\beta x H_{\text{tot}}^{\text{op}}} dx\right). \tag{2.62}$$

Then, the perturbation of the negativity in Eq. (2.32), $\delta N = -2\langle\phi_-|\delta\rho^{T_1}|\phi_-\rangle$, is given as

$$\begin{aligned}\delta N &= -\frac{\delta Z}{Z} N(\rho_{\text{op}}) \\ &+ \frac{2\beta}{Z}\int_0^1 \text{tr}\left[|\phi_-\rangle\langle\phi_-|\left(e^{-\beta(1-x)H_{\text{tot}}^{\text{op}}} \delta\mathcal{H}_{\text{LO}} e^{-\beta x H_{\text{tot}}^{\text{op}}}\right)^{T_1}\right] dx \\ &= \frac{\beta N(\rho_{\text{op}})}{Z} \text{tr}\left(\int_0^1 e^{-\beta(1-x)H_{\text{tot}}^{\text{op}}} \delta\mathcal{H}_{\text{LO}} e^{-\beta x H_{\text{tot}}^{\text{op}}} dx\right) \\ &+ \frac{2\beta}{Z}\int_0^1 \text{tr}\left[\big(|\phi_-\rangle\langle\phi_-|\big)^{T_1} e^{-\beta(1-x)H_{\text{tot}}^{\text{op}}} \delta\mathcal{H}_{\text{LO}} e^{-\beta x H_{\text{tot}}^{\text{op}}}\right] dx \\ &= \int_0^1 \text{tr}\left[e^{-\beta x H_{\text{tot}}^{\text{op}}} \hat{n} e^{-\beta(1-x)H_{\text{tot}}^{\text{op}}} \delta\mathcal{H}_{\text{LO}}\right] dx\end{aligned} \tag{2.63}$$

where

$$\hat{n} \equiv \frac{\beta}{Z}\left[N(\rho_{\text{op}})\big(I \otimes I\big) + 2\big(|\phi_-\rangle\langle\phi_-|\big)^{T_1}\right] \tag{2.64}$$

and we used the identity $N(\rho_{\rm op}) = -2\langle\phi_-|\rho_{\rm op}^{T_1}|\phi_-\rangle$ as well as $\mathrm{tr}(A^{T_1}B) = \mathrm{tr}(AB^{T_1})$. We will prove that the integrand of Eq. (2.63),

$$\mathrm{tr}\left[e^{-\beta x H_{\rm tot}^{\rm op}}\hat{n}e^{-\beta(1-x)H_{\rm tot}^{\rm op}}\delta\mathcal{H}_{\rm LO}\right], \tag{2.65}$$

always vanishes for $\{h_1^x, h_1^y, h_2^x, h_2^y, \xi, h_z\} = \{0, 0, 0, 0, 0, h\}$.

We prove in Appendix C that the operator

$$e^{-\beta x H_{\rm tot}^{\rm op}}\hat{n}e^{-\beta(1-x)H_{\rm tot}^{\rm op}} \tag{2.66}$$

has the same symmetry as the Hamiltonian $H_{\rm tot}^{\rm op}$ in Eq. (2.59), and thereby must be expanded in terms of the Pauli matrices in the form

$$\frac{1}{4}\left(q_{00}I\otimes I + q_{z0}(\sigma_1^z\otimes I - I\otimes\sigma_2^z) + \sum_{i=x,y,z} q_{ii}\sigma_1^i\otimes\sigma_2^i\right), \tag{2.67}$$

where q_{00}, q_{z0} and q_{ii} are appropriate coefficients. Therefore, we can calculate Eq. (2.65) to have the following equation:

$$\begin{aligned}
&\mathrm{tr}\left[e^{-\beta x H_{\rm tot}^{\rm op}}\hat{n}e^{-\beta(1-x)H_{\rm tot}^{\rm op}}\delta\mathcal{H}_{\rm LO}\right] \\
=&\mathrm{tr}\left\{\frac{1}{4}\left[q_{00}I\otimes I + q_{z0}(\sigma_1^z\otimes I - I\otimes\sigma_2^z)\right.\right. \\
&\left. + \sum_{i=x,y,z} q_{ii}\sigma_1^i\otimes\sigma_2^i\right]\times \\
&\left.\left[\sum_{i=x,y}(\delta h_1^i\sigma_1^i\otimes I + \delta h_2^i I\otimes\sigma_2^i) + h\delta\xi(\sigma_1^z\otimes I + I\otimes\sigma_2^z)\right]\right\}.
\end{aligned} \tag{2.68}$$

A straightforward algebra, such as $\mathrm{tr}(\sigma_1^z\otimes\sigma_2^x) = 0$, yields that Eq. (2.68) vanishes. This means that the perturbation of the negativity due to the infinitesimal variation of the local parameters $\{\delta h_x^1, \delta h_y^1, \delta h_x^2, \delta h_y^2, \delta\xi\}$ always vanishes at $\{h_1^x, h_1^y, h_2^x, h_2^y, \xi, h_z\} = \{0, 0, 0, 0, 0, h\}$. This completes the proof of Theorem 2.

To extend Theorem 1 to arbitrary temperatures, we assume the following hypothesis:
Hypothesis 1. The local parameters of the form $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{op}, 0, 0, -h_{op}\}$ *give not only an extremal value but also the maximum value of entanglement at arbitrary temperatures.*
We numerically tested this hypothesis using determinant-based entanglement measure $\pi(\rho)$ [28], which is given as

$$\pi(\rho) \equiv \begin{cases} 0, & \text{for } \rho^{T_1} \geq 0, \\ 2(|\det\rho^{T_1}|)^{1/4}, & \text{for } \rho^{T_1} < 0. \end{cases} \tag{2.69}$$

Though this entanglement measure is not a full entanglement monotone, it provides tight lower and upper bounds for other entanglement measures including the negativity and the concurrence. In addition, $\det\rho^{T_1}$ is expressed in the form of a polynomial and is much easier to maximize numerically than the concurrence and the negativity. Utilizing this measure, we tested Hypothesis 1 by numerical optimization for various kinds of interaction at various temperatures and found it always satisfied. In the following, we will assume Hypothesis 1 and conclude that $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$ is the globally optimizing solution at any temperatures.

For the local parameters $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h, 0, 0, -h\}$, the density matrix $Z\rho^{T_1}$ is given at arbitrary temperatures in the basis of the eigenstates of $\sigma_1^z \otimes \sigma_2^z$, $\{|00\rangle, |01\rangle, |10\rangle, |11\rangle\}$, as

$$Z\rho^{T_1} = \begin{pmatrix} a_1 & 0 & 0 & a_2 \\ 0 & b_1 - b_2 & b_3 & 0 \\ 0 & b_3 & b_1 + b_2 & 0 \\ a_2 & 0 & 0 & a_1 \end{pmatrix}, \tag{2.70}$$

where

$$\begin{aligned} a_1 &= e^{-\beta J_z} \cosh \beta J_1, \quad a_2 = -\frac{e^{\beta J_z}(J_x + J_y) \sinh \beta J_2}{J_2}, \\ b_1 &= e^{\beta J_z} \cosh \beta J_2, \quad b_2 = \frac{2h e^{\beta J_z} \sinh \beta J_2}{J_2}, \\ b_3 &= -e^{-\beta J_z} \sinh \beta J_1, \\ J_1 &\equiv |J_x - J_y|, \quad J_2 \equiv \sqrt{4h^2 + (J_x + J_y)^2}. \end{aligned} \tag{2.71}$$

Its eigenvalues are

$$\left\{a_1 - |a_2|, a_1 + |a_2|, b_1 + \sqrt{b_2^2 + b_3^2}, b_1 - \sqrt{b_2^2 + b_3^2}\right\}. \tag{2.72}$$

In Appendix D, we will prove that only $a_1 - |a_2|$ can have a negative value for $\{J_x, J_y\} \geq J_z \geq 0$ and $0 \geq J_z \geq \{J_x, J_y\}$. Therefore, the optimized negativity is given by

$$N(J_x, J_y, J_z, h, \beta) = \max(\tilde{N}, 0), \tag{2.73}$$

where

$$\begin{aligned} \tilde{N} &= -2\frac{a_1 - |a_2|}{Z} \\ &= -\frac{e^{-\beta J_z} \cosh \beta J_1 - \left(e^{\beta J_z} |J_x + J_y| \sinh \beta J_2\right)/J_2}{e^{-\beta J_z} \cosh \beta J_1 + e^{\beta J_z} \cosh \beta J_2}, \\ Z &= 2e^{-\beta J_z} \cosh \beta J_1 + 2e^{\beta J_z} \cosh \beta J_2. \end{aligned} \tag{2.74}$$

We find from this expression that we can always make the negativity positive by choosing an appropriate value of h.

The remaining task is to find the value of the optimizing field $h_{\rm op}$ at each temperature. We will do it analytically in the low-temperature limit $\beta \to \infty$ in Sec. III.C as well as do it numerically rigorously for a wide range of the temperature in Sec. IV.

2.3.3 Optimization in the low-temperature limit

We now discuss the optimization problem in the low-temperature limit.

Theorem 3. In the low-temperature limit $\beta \to \infty$, the optimized entanglement approaches to 1. The optimizing parameter $h_{\rm op}$ approaches to 0 when we choose the optimizing parameters as $\{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$.

Proof. We need to consider the three cases, namely the cases where the ground state of H_{int} is non-degenerate, doubly degenerate and triply degenerate. The eigenvalues $\{\epsilon_i\}_{i=1}^{4}$ and the corresponding eigenstates $\{|\psi_i\rangle\}_{i=1}^{4}$ of H_{int} are given by the following:

$$\begin{aligned} H_{\text{int}} &= \sum_{i=x,y,z} J_i \sigma_1^i \otimes \sigma_2^i, \\ \epsilon_1 &= -J_x - J_y - J_z, \quad |\psi_1\rangle = \frac{1}{\sqrt{2}}\Big(|01\rangle - |10\rangle\Big), \\ \epsilon_2 &= J_x + J_y - J_z, \quad |\psi_2\rangle = \frac{1}{\sqrt{2}}\Big(|01\rangle + |10\rangle\Big), \\ \epsilon_3 &= J_x - J_y + J_z, \quad |\psi_3\rangle = \frac{1}{\sqrt{2}}\Big(|00\rangle + |11\rangle\Big), \\ \epsilon_4 &= -J_x + J_y + J_z, \quad |\psi_4\rangle = \frac{1}{\sqrt{2}}\Big(|00\rangle - |11\rangle\Big). \end{aligned} \tag{2.75}$$

As has been described in Sec. II, we consider only the cases of $\{J_x, J_y\} \geq J_z \geq 0$ and $0 \geq J_z \geq \{J_x, J_y\}$.

In each case of $\{J_x, J_y\} \geq J_z \geq 0$ or $0 \geq J_z > \{J_x, J_y\}$, the ground state of H_{int} is non-degenerate, and ϵ_1 or ϵ_2 is the ground-state eigenvalue, respectively. In these cases, the ground state is a Bell state and it is clear that its entanglement is maximum. In other words, there is no need to optimize it further and $H_{\text{LO}}^{\text{op}} = 0$. We will see in Sec. IV that, in this non-degenerate case, there is indeed a finite range of the temperature where the negativity is maximized for $H_{\text{LO}}^{\text{op}} = 0$.

In each case of $0 \geq J_z = J_x > J_y$ and $0 \geq J_z = J_y > J_x$, the ground state of H_{int} is doubly degenerate and $\epsilon_2 = \epsilon_4$ or $\epsilon_2 = \epsilon_3$ is the ground-state eigenvalue, respectively. In the case $0 \geq J_z = J_x = J_y$, the ground state of H_{int} is triply degenerate and $\epsilon_2 = \epsilon_3 = \epsilon_4$ is the ground-state eigenvalue. In these degenerate cases, the ground states are mixed states and their entanglement always vanish. However, we can resolve the degeneracy of the ground states by an infinitesimal local Hamiltonian.

We hence employ Hypothesis 1 and put $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\text{op}}, 0, 0, -h_{\text{op}}\}$. We then calculate the asymptotic behavior of the optimized entanglement in the low-temperature limit $\beta \to \infty$. Below we will derive

$$\begin{aligned} h_{\text{op}} &\simeq \sqrt{\frac{\tilde{J}}{2\beta} \log 2\beta\tilde{J}} \quad \text{as} \quad \beta \to \infty, \\ N_{\text{op}} &\simeq 1 - \frac{1 + \log 2\beta\tilde{J}}{\beta\tilde{J}} \quad \text{as} \quad \beta \to \infty. \end{aligned} \tag{2.76}$$

in the doubly degenerate cases, where we defined $\tilde{J} \equiv |J_x + J_y|$, and

$$\begin{aligned} h_{\text{op}} &\simeq \sqrt{\frac{\tilde{J}}{2\beta} \log 4\beta\tilde{J}} \quad \text{as} \quad \beta \to \infty \\ N_{\text{op}} &\simeq 1 - \frac{1 + \log 4\beta\tilde{J}}{\beta\tilde{J}} \quad \text{as} \quad \beta \to \infty. \end{aligned} \tag{2.77}$$

in the triply degenerate case. In both cases the optimizing parameter h_{op} is infinitesimal and the optimized negativity N_{op} approaches to 1 in the low-temperature limit $\beta \to \infty$, although the forms of h_{op} and N_{op} are slightly different in the two cases. We will see in Sec. IV that, in these degenerate cases, there is indeed *no* finite range of the temperature where the negativity is maximized without local fields. In other words, we need a non-zero value of h_{op} at any non-zero temperatures.

Now we derive Eqs. (2.76) and (2.77). We start from Eq. (2.74) under Hypothesis 1. In the doubly degenerate cases $0 \geq J_z = J_x > J_y$ and $0 \geq J_z = J_y > J_x$, we can approximate Eq. (2.74) as

$$\begin{aligned}\tilde{N} &\simeq -\frac{e^{-\beta(J_z-J_1)} - \left(e^{\beta(J_z+J_2)}|J_x+J_y|\right)/J_2}{e^{-\beta(J_z-J_1)} + e^{\beta(J_z+J_2)}} \\ &= \frac{-1 + \left(e^{\beta(2J_z+J_2-J_1)}|J_x+J_y|\right)/J_2}{1+e^{\beta(2J_z+J_2-J_1)}} \end{aligned} \tag{2.78}$$

in the low-temperature limit $\beta \to \infty$, where we used the facts that $2\cosh\beta J_1 \simeq e^{\beta J_1}$, $2\sinh\beta J_2 \simeq e^{\beta J_2}$ and $2\cosh\beta J_2 \simeq e^{\beta J_2}$. Moreover, in these doubly degenerate cases, $2J_z + J_2 - J_1$ is either $2J_z + J_2 - J_x + J_y$ or $2J_z + J_2 + J_x - J_y$, which are summarized to $J_2 - |J_x + J_y|$. Then Eq. (2.78) reduces to

$$\tilde{N} \simeq \frac{-1 + e^{\beta X}\tilde{J}/(X+\tilde{J})}{1+e^{\beta X}}, \tag{2.79}$$

where

$$\begin{aligned}\tilde{J} &= |J_x + J_y|, \\ X \equiv J_2 - \tilde{J} &= \sqrt{4h^2 + (J_x+J_y)^2} - |J_x+J_y|. \end{aligned} \tag{2.80}$$

We first prove that $X \to 0$ and $\beta X \to \infty$ is a necessary and sufficient condition for $\tilde{N} \to 1$ in the low-temperature limit $\beta \to \infty$. In order to prove this, we calculate the value of $1 - \tilde{N}$ as follows:

$$\begin{aligned}1 - \tilde{N} &= 1 - \frac{-1 + e^{\beta X}\tilde{J}/(X+\tilde{J})}{1+e^{\beta X}} \\ &= \frac{2e^{-\beta X} + X/(X+\tilde{J})}{1+e^{-\beta X}}, \end{aligned} \tag{2.81}$$

Because $X \geq 0$ and $0 < e^{-\beta X} \leq 1$, we have $X/(X+\tilde{J}) \geq 0$ and $1 < 1 + e^{-\beta X} \leq 2$. Therefore, the necessary and sufficient condition for $1 - \tilde{N} \to 0$ in the low-temperature limit is

$$\beta X \to \infty \quad \text{and} \quad X \to 0 \quad \text{as} \quad \beta \to \infty. \tag{2.82}$$

In such cases, the negativity can be maximized to 1 in the low temperature limit $\beta \to \infty$.

Let us now calculate the optimizing parameter X_{op}. From the extremal condition for Eq. (2.79),

$$\frac{d\tilde{N}}{dX} = \frac{e^{\beta X}(\beta X^2 + 3\beta\tilde{J}X + 2\beta\tilde{J}^2 - \tilde{J} - e^{\beta X})}{(1+e^{\beta X})^2(\tilde{J}+X)^2} = 0, \tag{2.83}$$

we obtain

$$\beta X_{\text{op}} = \log\Big(\frac{\beta X_{\text{op}}^2}{\tilde{J}} + 3\beta X_{\text{op}} + 2\beta\tilde{J} - 1\Big). \tag{2.84}$$

Because of the condition (2.82), Eq. (2.84) reduces to

$$\begin{aligned}\beta X_{\rm op} &= \log 2\beta\tilde{J} + \log\Big(1 + \frac{X_{\rm op}^2}{2\tilde{J}^2} + \frac{3X_{\rm op}}{2\tilde{J}} - \frac{1}{2\beta\tilde{J}}\Big)\\ &\simeq \log 2\beta\tilde{J} \end{aligned}\tag{2.85}$$

in the limit $\beta \to \infty$. We thus have

$$X_{\rm op} \simeq \frac{\log 2\beta\tilde{J}}{\beta},\tag{2.86}$$

which indeed satisfies (2.82). The optimizing parameter $h_{\rm op}$ is thereby obtained in the form

$$\begin{aligned}h_{\rm op} &= \frac{1}{2}\sqrt{X_{\rm op}^2 + 2\tilde{J}X_{\rm op}}\\ &\simeq \sqrt{\frac{\tilde{J}X_{\rm op}}{2}}\\ &\simeq \sqrt{\frac{\tilde{J}}{2\beta}\log 2\beta\tilde{J}},\end{aligned}\tag{2.87}$$

where we utilized Eq. (2.80) to derive the first equality. Moreover, the optimized negativity is given by

$$\begin{aligned}N_{\rm op} &\simeq \frac{-e^{-\beta X_{\rm op}} + 1/(X_{\rm op}/\tilde{J} + 1)}{e^{-\beta X_{\rm op}} + 1}\\ &\simeq (1 - e^{-\beta X_{\rm op}})\Big(-e^{-\beta X_{\rm op}} + 1 - \frac{X_{\rm op}}{\tilde{J}}\Big)\\ &\simeq 1 - \frac{X_{\rm op}}{\tilde{J}} - 2e^{-\beta X_{\rm op}}\\ &\simeq 1 - \frac{1 + \log 2\beta\tilde{J}}{\beta\tilde{J}},\end{aligned}\tag{2.88}$$

where we used Eq. (2.82) upon moving from the first line to the second line. Thus Eq. (2.76) is proved.

In the triply degenerate case $0 \geq J_z = J_x = J_y$, we have $J_1 = 0$, and thereby we can approximate Eq. (2.74) as

$$\begin{aligned}\tilde{N} &\simeq -\frac{2e^{-\beta J_z} - \big(e^{\beta(J_z+J_2)}|J_x + J_y|\big)/J_2}{2e^{-\beta J_z} + e^{\beta(J_z+J_2)}}\\ &= \frac{2 + \big(e^{\beta(2J_z+J_2)}|J_x + J_y|\big)/J_2}{2 + e^{\beta(2J_z+J_2)}}\end{aligned}\tag{2.89}$$

in the low-temperature limit $\beta \to \infty$, where we used the facts that $\cosh\beta J_1 \simeq 1$, $2\sinh\beta J_2 \simeq e^{\beta J_2}$ and $2\cosh\beta J_2 \simeq e^{\beta J_2}$. Moreover, in this case, $2J_z + J_2$ is equal to $J_2 - |J_x + J_y|$, and therefore Eq. (2.89) reduces to

$$\tilde{N} \equiv \frac{-2 + e^{\beta X}\tilde{J}/(X + \tilde{J})}{2 + e^{\beta X}},\tag{2.90}$$

where X and $\tilde{J}$ are defined in Eq. (2.80). From the extremal condition $d\tilde{N}/dX = 0$, we obtain

$$\begin{aligned} X_{\rm op}\beta &= \log\Big(\frac{2\beta X_{\rm op}^2}{\tilde{J}} + 6\beta X_{\rm op} + 4\beta\tilde{J} - 2\Big) \\ &\simeq \log 4\beta\tilde{J}, \end{aligned} \tag{2.91}$$

where we used the same logic as the one with which we derived Eq. (2.85) in the doubly degenerate case. In this way, the optimizing parameter $h_{\rm op}$ and the optimized negativity $N_{\rm op}$ are given as

$$h_{\rm op} \simeq \sqrt{\frac{\tilde{J}}{2\beta}\log 4\beta\tilde{J}}, \tag{2.92}$$

and

$$\begin{aligned} N_{\rm op} &\simeq 1 - \frac{X_{\rm op}}{\tilde{J}} - 4e^{-\beta X_{\rm op}} \\ &\simeq 1 - \frac{1 + \log 4\beta\tilde{J}}{\beta\tilde{J}}. \end{aligned} \tag{2.93}$$

Thus Eq. (2.77) is proved. This completes the proof of Theorem 3.

2.3.4 Negativity and concurrence

We here mention the relationship between the negativity and the concurrence [26]. The concurrence is also an important entanglement measure. Concerning the optimization problem of the concurrence, we can only prove that the negativity N and the concurrence C have the same value for the local parameters $\{0, 0, h, 0, 0, -h\}$ with an arbitrary value of h; namely,

$$N(0, 0, h, 0, 0, -h, \beta) = C(0, 0, h, 0, 0, -h, \beta). \tag{2.94}$$

This equation is proven by the theorem in Ref. [29], which says that the concurrence is equal to the negativity iff the eigenvector of ρ^{T_1} corresponding to its negative eigenvalue is a Bell state up to local unitary transformations.

For the local parameters $\{0, 0, h, 0, 0, -h\}$, the density matrix $Z\rho^{T_1}$ is given in Eq. (2.70) and only the eigenvalue $\tilde{N} = a_1 - |a_2|$ can be negative. For $\{J_x, J_y\} \geq J_z \geq 0$ and $0 \geq J_z \geq \{J_x, J_y\}$ the eigenvectors of $Z\rho^{T_1}$ corresponding to the eigenvalue $\tilde{N} = a_1 - |a_2|$ is $(|00\rangle + |11\rangle)/\sqrt{2}$ and $(|00\rangle - |11\rangle)/\sqrt{2}$, respectively, both being a Bell state. In the case of $\tilde{N} > 0$, the concurrence must be equal to the negativity because the eigenvector of ρ^{T_1} corresponding to its negative eigenvalue is a Bell state. In the case of $\tilde{N} \leq 0$, the negativity $N = \max(\tilde{N}, 0)$ is equal to 0 and the entanglement does not exist. Therefore, the concurrence and the negativity are both equal to 0. This completes the proof of Eq. (2.94)

2.4 High- and low-temperature phases

In the present section, we calculate the optimizing local Hamiltonian and the optimized entanglement numerically rigorously. After the analysis in Sec. III, we here set $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$. In the calculations below, we will see that there are two kinds of temperature range, which we refer to as the high- and low-temperature phases. We will find

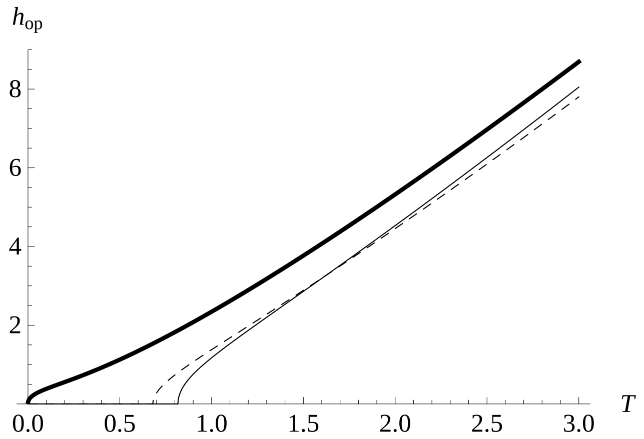


Figure 2.1: Numerically rigorous solution of the optimizing local parameter $h_{\rm op}$: the solid line for $\{J_x, J_y, J_z\} = \{1/3, 1/3, 1/3\}$, the dashed line for $\{J_x, J_y, J_z\} = \{1/2, 1/3, 1/6\}$ and the thick line for $\{J_x, J_y, J_z\} = \{-1/2, -1/4, -1/4\}$. The number of data points is 3000 for each case. The boundary temperatures $T_{\rm c}$ between the high- and low-temperature phases are $0.8168\cdots$, $0.6803\cdots$ and 0 for $\{1/3, 1/3, 1/3\}$, $\{1/2, 1/3, 1/6\}$ and $\{-1/2, -1/4, -1/4\}$, respectively.

that in the low-temperature phase the optimizing local parameter $h_{\rm op}$ vanishes, whereas in the high-temperature phase it dose not. We start from Eq. (2.74) with the optimizing parameters $\{h_1^x, h_1^y, h_1^z, h_2^x, h_2^y, h_2^z\} = \{0, 0, h_{\rm op}, 0, 0, -h_{\rm op}\}$. The parameter $h_{\rm op}$ which maximizes the negativity can be calculated from

$$\begin{aligned} \frac{1}{h}\frac{\partial \tilde{N}}{\partial h}\bigg|_{h=h_{\rm op}} &\propto |J_x + J_y|(\beta J_2 \cosh \beta J_2 - \sinh \beta J_2) \\ &+ J_2^2 \beta \sinh \beta J_2 - \frac{e^{2\beta J_z}|J_x + J_y|}{\cosh \beta J_1}\left(-J_2\beta + \frac{\sinh 2\beta J_2}{2}\right) \\ =&0, \end{aligned} \tag{2.95}$$

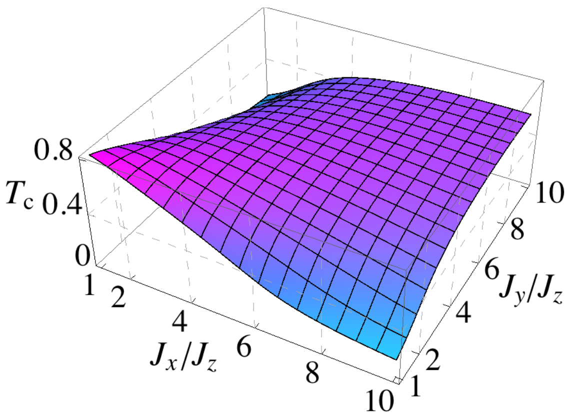
where the factor $1/h$ is added to remove the trivial solution of $h = 0$. In Fig. 2.1, we show the optimizing local parameter $h_{\rm op}$ in the cases of $\{J_x, J_y, J_z\} = \{1/3, 1/3, 1/3\}$, $\{1/2, 1/3, 1/6\}$ and $\{-1/2, -1/4, -1/4\}$. See Appendix A for the convergence of $h_{\rm op}$ to the asymptotes (2.11) and (2.15).

In the high-temperature phase, Eq. (2.95) has a non-trivial solution of $h_{\rm op} > 0$, while in the low-temperature phase, Eq. (2.95) has no solutions and the optimizing value $h_{\rm op}$ is zero, which is the trivial solution of $\partial \tilde{N}/\partial h = 0$. Therefore, the boundary temperature $T_{\rm c}$ between the high- and low-temperature phases is a solution of

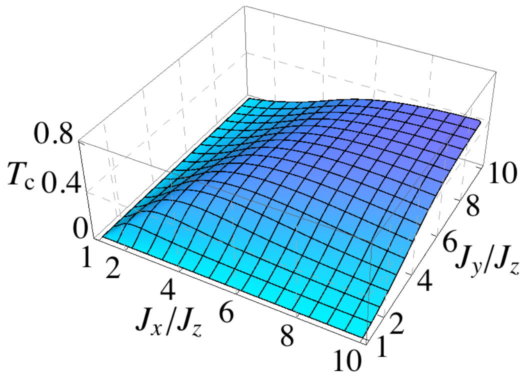
$$\lim_{h\to 0} \frac{1}{h}\frac{\partial \tilde{N}(J_x, J_y, J_z, h, \beta)}{\partial h} = 0. \tag{2.96}$$

The boundary temperature $T_{\rm c}$ is defined for each interaction Hamiltonian $H_{\rm int}$.

In Fig. 2.2, we show the boundary temperature $T_{\rm c}$ in the cases of $\{J_x, J_y\} \geq J_z \geq 0$ and $0 \geq J_z \geq \{J_x, J_y\}$, which correspond to all kinds of interaction thanks to Lemma 1. We calculated the data in Fig. 2.2 from (2.96), normalizing the interaction parameters so that $||H_{\rm int}||_2 = 1$, where $||\ ||_2$ is the spectral norm. From Fig. 2.2, we see the following properties. First, the boundary temperatures $T_{\rm c}$ are higher in the antiferromagnetic cases $\{J_x, J_y\} \geq J_z \geq 0$ than in the ferromagnetic cases $0 \geq J_z \geq \{J_x, J_y\}$. Second, in the antiferromagnetic systems,



(a)



(b)

Figure 2.2: (color online) The boundary temperature T_c between the high- and low-temperature phases, (a) for the antiferromagnetic case $\{J_x, J_y\} \geq J_z \geq 0$ and (b) for the ferromagnetic case $0 \geq J_z \geq \{J_x, J_y\}$. The point of origin is $(1, 1)$, which corresponds to the isotropic Heisenberg interaction. In (a), the boundary temperatures are $0.8168\cdots$, $0.1292\cdots$, $0.1292\cdots$, $0.5735\cdots$ and $0.6208\cdots$ at $(1, 1)$, $(1, 10)$, $(10, 1)$, $(10, 10)$ and $(5, 5)$, respectively. The maximum temperature is $0.8168\cdots$ at $(1, 1)$, which is the XXX point. In (b), the boundary temperatures are 0, 0, 0, $0.4126\cdots$ and $0.3188\cdots$ at $(1, 1)$, $(1, 10)$, $(10, 1)$, $(10, 10)$ and $(5, 5)$, respectively. The maximum temperature is $0.5184\cdots$ at $\lim_{x\to\infty}(x, x)$, which is the XX point.

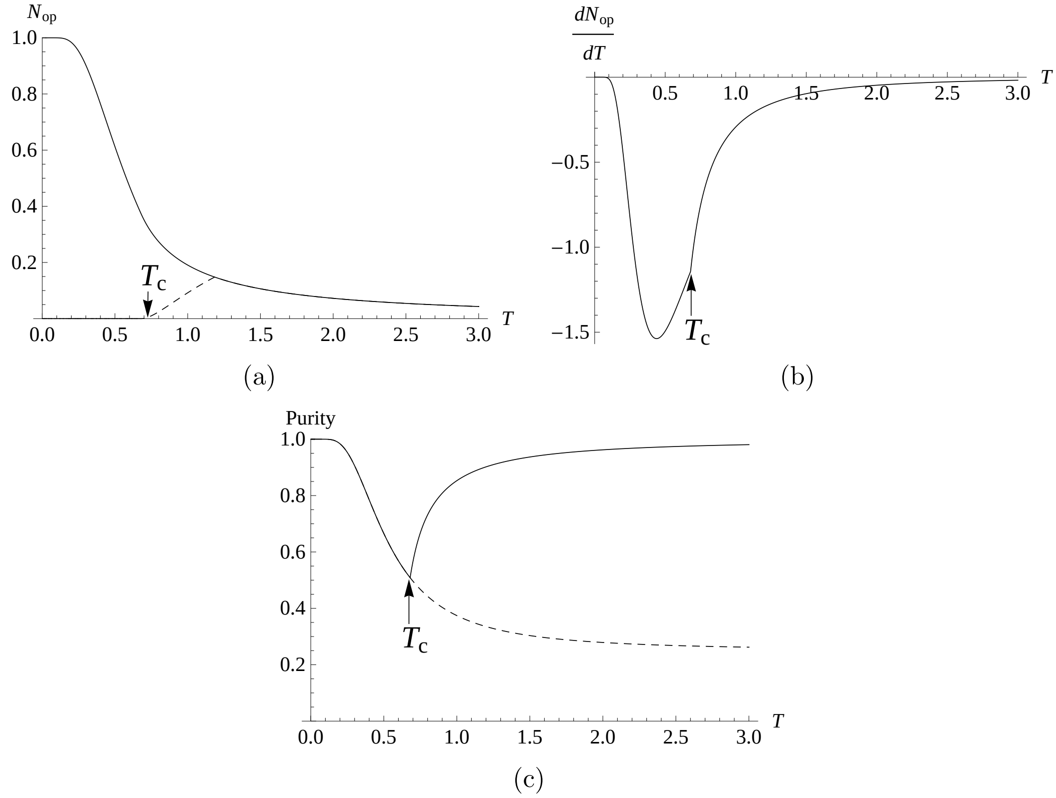


Figure 2.3: Plots of (a) the negativity; (b) its first derivatives; (c) the purity, for $J_x = 1/2$, $J_y = 1/3$, $J_z = 1/6$. The solid line is the optimized entanglement and the dashed line is the entanglement enhancement defined in the text. The boundary temperature is $T_c = 0.6803\cdots$. At $T = 1.185\cdots$, the entanglement enhancement is maximum, where the value is $0.1480\cdots$. In (b), we obtained the data points by the finite-difference method. In (c), the solid line is the purity of the optimized state and the dashed line is the one under no local Hamiltonian. The minimum value of the purity is $0.5087\cdots$ at T_c.

the boundary temperature T_c is maximum of $0.8168\cdots$ for the isotropic Heisenberg interaction (the XXX model). Next, the boundary temperature T_c is zero in the cases of $0 \geq J_z = J_x \geq J_y$ and $0 \geq J_z = J_y \geq J_x$ as well as the case of the ferromagnetic isotropic Heisenberg model, which means that the low-temperature phase shrinks to the zero temperature in these doubly and triply degenerate cases analyzed in Sec. III.C. We have revealed in Sec III.C that in the low-temperature limit $\beta \to \infty$ the negativity is strictly 1 with no local Hamiltonian in the non-degenerate cases. The present calculation indeed shows that the low-temperature phase extends to a finite temperature in the non-degenerate cases. In the antiferromagnetic system, on the other hand, the boundary temperature is zero only in the case of the Ising model, $J_x = J_z = 0$ or $J_y = J_z = 0$.

Next, we consider the singularity at the boundary between the high- and low-temperature phases. In Fig. 2.3, we show the optimized negativity, its first derivative and the purity $\mathrm{tr}(\rho^2)$ in the case of $\{J_x, J_y, J_z\} = \{1/2, 1/3, 1/6\}$. We also consider the entanglement enhancement, which is defined as the difference of the entanglement between the optimized entanglement and the entanglement under no local Hamiltonian, namely $N(H_{\mathrm{LO}}^{\mathrm{op}}) - N(H_{\mathrm{LO}} = 0)$. We numerically rigorously calculated the data in Fig. 2.3(a) using (2.95), and the derivatives by the finite-difference method. Figure 2.3(b) shows that the second derivative of the negativity is not continuous at the boundary and Fig. 2.3(c) shows that the first derivative of the purity is not continuous at the boundary. On the other hand, there is no singularity at the point of $T = 1.185\cdots$, where the derivative of the entanglement enhancement is not continuous.

The emergence of the high- and low-temperature phases is due to the following reason. First, the entanglement enhancement by addition of the local Hamiltonian comes from the fact that a local Hamiltonian increases the purity and suppresses the entanglement loss caused by thermal mixing, which is demonstrated in Fig. 2.3(c). On the other hand, too strong magnetic fields bring the quantum system close to a direct product states and hence destroy the entanglement. These two effects compete to give rise to the two phases. In the low-temperature phase, we do not need a magnetic field because the purity is already high. In the high-temperature phase, on the other hand, we need a magnetic field because the thermal fluctuation decreases the purity. The transition from the low-temperature phase to the high-temperature phase means that the enhancement of the entanglement due to the increase of the purity becomes predominant compared with the entanglement decay caused by the magnetic decoupling.

2.5 Summary and conclusion

We have analytically and numerically rigorously studied thermal states of quantum systems where two qubits interact under a local Hamiltonian H_{LO} and have determined the local Hamiltonian H_{LO} which maximizes the thermal entanglement under a fixed interaction. As a result, we have found that the interaction Hamiltonian can be transformed into the XYZ-exchange interactions whose parameters are either antiferromagnetic as $\{J_x, J_y\} \geq J_z \geq 0$ or ferromagnetic as $0 \geq J_z \geq \{J_x, J_y\}$ and that the optimizing local Hamiltonian always takes the form of $h_{\mathrm{op}}(\sigma_1^z \otimes I - I \otimes \sigma_2^z)$, where h_{op} depends on the temperature. In addition, we have proved that the optimized entanglement does not vanish at any temperatures and decays slowly according to $1/(T \log T)$ at high temperatures. We have also found that in the low-temperature phase the entanglement is maximum without any local Hamiltonian and have investigated the interaction dependence of the boundary temperature of this range. Indeed, the low-temperature phase shrinks to the zero temperature point if the interaction Hamiltonian has degeneracy. At the same time, we have discovered a singularity of the optimized entanglement at the boundary temperature, where the second derivative is discontinuous.

In conclusion, our work has revealed general properties of the thermal entanglement of interacting two qubits, though we have assumed a numerically confirmed hypothesis. The concept of high- and low-temperature phases is an interesting property in that it is based on the response to external manipulation of local Hamiltonians. It is likely that we can find more interesting properties of entanglement in this regard. In the next chapter, we investigate two qubits which interact indirectly or general bipartite systems.

Chapter 3

General properties of the maximized entanglement of indirectly interacting two spins

In the present chapter, we investigate the thermal entanglement of indirectly interacting two spins through other spins, that is, two spins at the ends of a spin chain. We maximize it by tuning the local fields on the two spins to obtain the maximized entanglement. We present a necessary condition for the indirect interaction to give a non-zero maximized entanglement. We also prove that if the two spins are separated by two sites or more, there is a critical temperature above which the maximized entanglement vanishes. We numerically calculate the maximized entanglement in three-spin chains and four-spin chains. We discover that the maximizing local fields on the spins 1 and 2 have asymmetric forms, which implies that the asymmetry of the two spins essentially contributes to the entanglement enhancement. In the three-spin chains, we explain this enhancement due to the asymmetry qualitatively and quantitatively in terms of the magnons. In XX and XY four-spin chains, we find that the critical temperature shows qualitatively different behavior depending on the conservation of the angular momentum in the z direction.

3.1 Introduction

In the previous chapter, we showed how much entanglement we can enhance with the local fields in the two-spin system. In the present chapter, we will extend the result to multipartite spin systems; we study the enhancement of the bipartite entanglement due to the local fields in the system where the two spins indirectly interact with each other via other spins. The main research targets are the following three problems:

- What is the condition for the indirect interaction to generate the entanglement?
- Is it possible to generate the entanglement at high temperatures in the indirectly interacting spins?
- What are the main factors which make it possible for the local fields to enhance the entanglement?

For the directly interacting two spins, we obtained the answer for these questions in the previous chapter [55]: any direct interaction can generate the entanglement for appropriate local fields;

the maximized entanglement decays as $1/T \ln T$ as the temperature increases; the entanglement enhancement by the local fields is brought about by the suppression of the thermal fluctuation, or in other words, the increase of the purity. Our purpose in the present chapter is to answer these questions qualitatively and quantitatively for indirectly interacting spins.

The possibility of the enhancement of the entanglement with the external fields has been shown in many papers [4,31,54]. However, there have been few reports about the entanglement enhancement of specific two spins. In particular, little is known about the mechanism of the entanglement enhancement due to the external fields or the condition for the indirect interaction to generate the entanglement. By making these problems clear, we may be able to control the entanglement efficiently with the external fields.

In our research, we define the maximized entanglement as the maximum value of the entanglement between the particular two spins under the condition that we can arbitrarily tune the local fields only on these two spins. If the maximized entanglement is equal to zero, the entanglement generation is impossible for any values of the local fields. The forms of the maximizing local fields reflect basic properties of the entanglement enhancement, and thereby help us solve the above three problems.

We study general properties of the maximized entanglement between the focused two spins which are connected by a spin chain. Our main results are the following three:

- We obtain a necessary condition for the indirect interaction to generate the non-zero entanglement by optimizing the local fields.
- At high temperatures, we prove that the maximized entanglement is always equal to zero between the two spins which are separated by two or more spins. In other words, above a critical temperature $\tilde{T}_{\mathrm{c}}$, we can never generate the entanglement between spins far apart for any values of the local fields.
- The main factors of the entanglement enhancement due to the local fields are not only the increase of the purity but also their effect on the indirect interaction. To be more specific, the external fields affect the magnons which mediate the indirect interaction. The form of the maximizing local fields depends on this effect and has asymmetric forms in a particular parameter region.

We show these results analytically and numerically. This chapter consists of the following sections: in Section 2, we state the problem specifically and give some definitions; in Section 3, we give general theorems on the entanglement enhancement which can be applied to any spin chains; in Section 4, we show the numerical and analytical results on the maximization of the entanglement in three-spin chains; in Section 5, we show the numerical and analytical results on the maximization of the entanglement in four-spin chains; and in Section 6, discussion concludes the chapter.

3.2 Statement of the problem

First, we formulate the framework of the entanglement maximization problem and describe conditions. We consider a general XYZ N-spin chain with external fields in the z direction. The most general form of the Hamiltonian of this system is given as follows:

$$H_{\mathrm{tot}} = \sum_{i=1}^{N-1} (J_i^x \sigma_i^x \sigma_{i+1}^x + J_i^y \sigma_i^y \sigma_{i+1}^y + J_i^z \sigma_i^z \sigma_{i+1}^z) + \sum_{i=1}^{N} h_i^z \sigma_i^z, \tag{3.1}$$

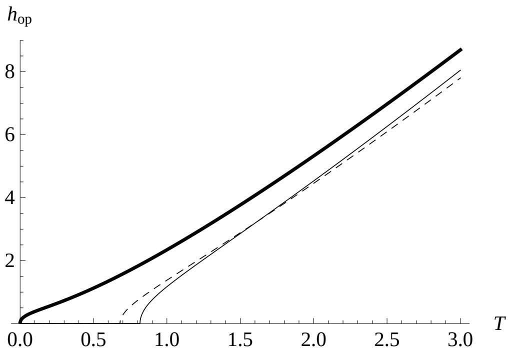


Figure 3.1: Schematic picture of the spin chains. We define the spins 1 and N as the focused spins and the spins which mediate the indirect interaction between the spins 1 and N as the media spins. We assume that we can tune the magnetic fields of the spins 1 and N, while the other fields $\{h_i^z\}_{i=2}^{N-1}$ are arbitrary but fixed.

where $\{\sigma_i^i\}_{i=x,y,z}$ are the Pauli matrices and we adopt the free boundary conditions.

We hereafter consider the entanglement between the spins 1 and N at the ends of the chain. We define these two spins as the focused spins and the extremal fields h_1^z and h_N^z on the two spins as the local fields. We refer to the other spins $2 \leq i \leq N-1$ as the media spins (Fig. 3.1). We then define the interaction Hamiltonian as the total Hamiltonian (3.1) minus the terms of the local fields:

$$H_{\text{int}} = \sum_{i=1}^{N-1} (J_i^x \sigma_i^x \sigma_{i+1}^x + J_i^y \sigma_i^y \sigma_{i+1}^y + J_i^z \sigma_i^z \sigma_{i+1}^z) + \sum_{i=2}^{N-1} h_i^z \sigma_i^z, \tag{3.2}$$

The basic problem that we are going to solve is to maximize the thermal entanglement between the focused spins 1 and N by tuning the local fields h_1^z and h_N^z at a fixed temperature. We also fix all the parameters in H_{int}, namely, $\{J_i^x, J_i^y, J_i^z\}$ for $1 \leq i \leq N-1$ and $\{h_i^z\}$ for $2 \leq i \leq N-1$. We refer to the maximizing values of the local fields h_1^z and h_N^z as $h_{1\text{op}}$ and $h_{N\text{op}}$.

Note that the maximizing local fields $h_{1\text{op}}$ and $h_{N\text{op}}$ generally depend on the temperature T, or on the inverse temperature $\beta = 1/(kT)$ with k the Boltzmann constant. This is because we tune the local fields at a fixed temperature β. Let us then define the high-temperature limit, in which we mostly develop the argument. In our high-temperature limit, we make β tend to zero with the parameters in H_{int} fixed. In other words, we have $\beta||H_{\text{int}}|| \to 0$ in the high-temperature limit, where $||H_{\text{int}}||$ is the norm of H_{int}. On the other hand, we let the maximizing local fields depend on β as we take the limit $\beta \to 0$. Hence, $\beta h_{1\text{op}}$ and $\beta h_{N\text{op}}$ can even diverge in our high-temperature limit.

The density matrix of the total system in thermal equilibrium is

$$\rho_{\text{tot}} = \frac{e^{-\beta H_{\text{tot}}}}{Z}, \tag{3.3}$$

where $Z = \text{tr}(e^{-\beta H_{\text{tot}}})$ is the partition function. The density matrix of the focused spins 1 and N is

$$\rho_{1N} = \text{tr}_{1N} \rho_{\text{tot}}, \tag{3.4}$$

where tr_{1N} denotes the trace operation on the spins *except* the focused spins 1 and N. For the present system (3.1), the general form of the density matrix ρ_{1N} is given by:

$$\rho_{1N} = \begin{pmatrix} p_{\uparrow\uparrow} & 0 & 0 & F_2 \\ 0 & p_{\uparrow\downarrow} & F_1 & 0 \\ 0 & F_1 & p_{\downarrow\uparrow} & 0 \\ F_2 & 0 & 0 & p_{\downarrow\downarrow} \end{pmatrix} \tag{3.5}$$

in the basis of the eigenstates of $\sigma_1^z \otimes \sigma_N^z$, where $p_{\uparrow\uparrow}$, $p_{\uparrow\downarrow}$, $p_{\downarrow\uparrow}$, $p_{\downarrow\downarrow}$, F_1 and F_2 are real numbers. We have $F_2 = 0$ when $J_i^x = J_i^y$, for $1 \le i \le N-1$, in particular.

In order to quantify the entanglement, we here adopt the concurrence [26] as an entanglement measure. The concurrence $C(\rho_{1N})$ is defined as follows:

$$C(\rho_{1N}) \equiv \max(\lambda_1 - \lambda_2 - \lambda_3 - \lambda_4, 0), \tag{3.6}$$

where $\{\lambda_n\}_{n=1}^4$ are the eigenvalues of the matrix

$$\sqrt{\rho_{1N}(\sigma_1^y \otimes \sigma_N^y)\rho_{1N}^*(\sigma_1^y \otimes \sigma_N^y)} \tag{3.7}$$

in the non-ascending order $\lambda_1 \ge \lambda_2 \ge \lambda_3 \ge \lambda_4$. For density matrices of the form (3.5), the concurrence $C(\rho_{1N})$ is reduced to the simpler form

$$C(\rho_{1N}) = 2\max(|F_1| - \sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}, |F_2| - \sqrt{p_{\uparrow\downarrow}p_{\downarrow\uparrow}}, 0). \tag{3.8}$$

Then, the necessary and sufficient condition for the existence of the entanglement is given by

$$\max(|F_1| - \sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}, |F_2| - \sqrt{p_{\uparrow\downarrow}p_{\downarrow\uparrow}}) > 0. \tag{3.9}$$

Thus, the present entanglement optimization problem for the spin pair $(1, N)$ is equivalent to finding the values of $\{h_1^z, h_N^z\}$ which maximize $C(\rho_{1N})$ for the fixed parameters $\{J_i^x, J_i^y, J_i^z\}$ $(1 \le i \le N-1)$, $\{h_i^z\}$ $(2 \le i \le N-1)$, and β.

3.3 General theorems on entanglement generation

In the previous section, we formulated the entanglement maximization problem. In fact, there are cases in which we cannot generate the entanglement for any values of the local fields at all. In such cases, it is necessary to distinguish whether the entanglement is exactly equal to zero or rapidly approaches to zero. In the present section, we introduce general theorems on a necessary condition to generate the entanglement by optimizing the local fields. In other words, we give a sufficient condition for the entanglement to be zero exactly for any values of the local fields.

Theorem 1. If there exists the following separation of the interaction Hamiltonian H_{int}, we cannot generate the entanglement between the focused spins σ_1 and σ_N for any values of the local fields at arbitrarily temperatures:

$$\begin{aligned} &H_{\text{int}} = H_A(\sigma_1) + H_B(\sigma_N) \\ &\text{with} \quad [H_A(\sigma_1), H_B(\sigma_N)] = 0, \end{aligned} \tag{3.10}$$

where $[\cdots]$ is the commutator. Note that H_A does not contain σ_N nor H_B contains σ_1.

Comments. For example, we can separate the following Hamiltonian in the form (3.10):

$$H_{\text{int}} = J_1^x\sigma_1^x\sigma_2^x + J_1^y\sigma_1^y\sigma_2^y + J_1^z\sigma_1^z\sigma_2^z + J_2^x\sigma_2^x\sigma_3^x + J_3^x\sigma_3^x\sigma_4^x, \tag{3.11}$$

where the spin pair $(1, 2)$ interact with each other through the XYZ interaction, while the spin pairs $(2, 3)$ and $(3, 4)$ interact with each other through the Ising interaction. We can separate this Hamiltonian into $H_A(\sigma_1)$ and $H_B(\sigma_4)$ as

$$\begin{aligned} H_A(\sigma_1) &= J_1^x \sigma_1^x \sigma_2^x + J_1^y \sigma_1^y \sigma_2^y + J_1^z \sigma_1^x \sigma_2^x + J_2^x \sigma_2^x \sigma_3^x, \\ H_B(\sigma_4) &= J_3^x \sigma_3^x \sigma_4^x. \end{aligned} \tag{3.12}$$

These Hamiltonians satisfy the condition $[H_A(\sigma_1), H_B(\sigma_4)] = 0$, and hence we cannot generate the entanglement between the focused spins σ_1 and σ_4 in this system for any values of the local fields h_1^z and h_4^z. However, the spins 1 and 4 are classically correlated with each other.

If we add $h_3^z \sigma_3^z$ to H_{int} as

$$H_{\text{int}} = J_1^x \sigma_1^x \sigma_2^x + J_1^y \sigma_1^y \sigma_2^y + J_1^z \sigma_1^z \sigma_2^z + J_2^x \sigma_2^x \sigma_3^x + h_3^z \sigma_3^z + J_3^x \sigma_3^x \sigma_4^x, \tag{3.13}$$

we cannot separate the Hamiltonian H_{int} into the forms of $H_A(\sigma_1)$ and $H_B(\sigma_4)$ which satisfy $[H_A(\sigma_1), H_B(\sigma_4)] = 0$, and hence the spins 1 and 4 can entangle with each other. It is worth noting that the field in the z direction on the media spin 3 makes the classical interaction (3.12) into the non-classical one.

Eigenstates can have the entanglement even if the condition (3.10) is satisfied. For example, the Hamiltonian for the focused spins 1 and 3,

$$\begin{aligned} H_{\text{tot}} &= H_{\text{int}} + h_1^z \sigma_1^z + h_3^z \sigma_3^z, \\ H_{\text{int}} &= J_1^z \sigma_1^z \sigma_2^z + J_2^z \sigma_2^z \sigma_3^z \end{aligned} \tag{3.14}$$

with $h_1^z = h_3^z = J_1^z = J_2^z$ satisfies the condition (3.10), but it has an eigenstate $(|\uparrow_1 \uparrow_3 \uparrow_2\rangle + |\downarrow_1 \uparrow_3 \downarrow_2\rangle)/\sqrt{2}$, which is highly entangled. Mixing of all the eigenstates with the Boltzmann weight always destroys the entanglement between the focused spins.

Finally, under this condition, for the appropriate values of the local fields we can generate the quantum discord, which is one of the non-classical correlations. We discuss the quantum discord in Appendix B.1. We also note that the present theorem applies to the general case in which there are couplings in all possible pairs of N spins.

Proof. In order to prove the present theorem, we prove that the density matrix can be decomposed into the mixture of the product states as

$$\rho_{1N} = \sum_{\tilde{n}} \rho_1^{\tilde{n}} \otimes \rho_N^{\tilde{n}}, \tag{3.15}$$

where the density matrices $\rho_1^{\tilde{n}}$ and $\rho_N^{\tilde{n}}$ of the focused spins 1 and N, respectively, are physical states, or, in other words, positive matrices. Then, the spins 1 and N are not entangled with each other by definition.

First, if the condition (3.10) is satisfied, we can decompose the density matrix as follows:

$$Z\rho_{\text{tot}} = e^{-\beta H_A(\sigma_1)} e^{-\beta H_B(\sigma_N)}. \tag{3.16}$$

We can express $e^{-\beta H_A(\sigma_1)}$ and $e^{-\beta H_B(\sigma_N)}$ as

$$\begin{aligned} e^{-\beta H_A(\sigma_1)} &= \sum_{\mu=0,x,y,z} \sigma_1^\mu \otimes \rho_{\text{media}}^{1\mu} \otimes I_N, \\ e^{-\beta H_B(\sigma_N)} &= \sum_{\nu=0,x,y,z} I_1 \otimes \rho_{\text{media}}^{N\nu} \otimes \sigma_N^\nu, \end{aligned} \tag{3.17}$$

where I_1 and I_N are the unit matrices in the spaces of the spins 1 and N, respectively, and we define $\sigma_1^0 = I_1$ and $\sigma_N^0 = I_N$. We also define that $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{N\nu}$ are Hermitian operators in the media-spin space. Because $H_A(\sigma_1)$ and $H_B(\sigma_N)$ commute with each other, $e^{-\beta H_A(\sigma_1)}$ and $e^{-\beta H_B(\sigma_N)}$ also commute with each other. Therefore, we obtain the following equation:

$$\mathrm{tr}^{1N}\big[\sigma_1^\mu \otimes \sigma_N^\nu e^{-\beta H_A(\sigma_1)} e^{-\beta H_B(\sigma_N)}\big] = \mathrm{tr}^{1N}\big[\sigma_1^\mu \otimes \sigma_N^\nu e^{-\beta H_B(\sigma_N)} e^{-\beta H_A(\sigma_1)}\big], \tag{3.18}$$

where tr^{1N} denotes the trace operation only on the spins 1 and N. From this equation we can obtain

$$\rho_{\text{media}}^{1\mu}\rho_{\text{media}}^{N\nu} = \rho_{\text{media}}^{N\nu}\rho_{\text{media}}^{1\mu} \tag{3.19}$$

for $\mu, \nu = 0, x, y, z$. Therefore, $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{N\nu}$ have simultaneous eigenstates. Then, we can express $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{N\nu}$ as

$$\rho_{\text{media}}^{1\mu} = \sum_{n=1}^{2^{N-2}} \lambda_\mu^n |n, \mu_1, \nu_N\rangle\langle n, \mu_1, \nu_N| \tag{3.20}$$

and

$$\rho_{\text{media}}^{N\nu} = \sum_{n=1}^{2^{N-2}} \tau_\nu^n |n, \mu_1, \nu_N\rangle\langle n, \mu_1, \nu_N|. \tag{3.21}$$

where $\{|n, \mu_1, \nu_N\rangle\}$ are 2^{N-2} pieces of the simultaneous eigenstates of $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{N\nu}$. Note, however, that $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{1\mu'}$ may not have simultaneous eigenvalues when they have different degeneracies, nor $\rho_{\text{media}}^{N\nu}$ and $\rho_{\text{media}}^{N\nu'}$. As a result, we obtain

$$\begin{aligned} &e^{-\beta H_A(\sigma_1)} e^{-\beta H_B(\sigma_N)} \\ =&\Big(\sum_{n,\mu} \lambda_\mu^n \sigma_1^\mu \otimes |n, \mu_1, \nu_N'\rangle\langle n, \mu_1, \nu_N'| \otimes I_N\Big)\Big(\sum_{n',\nu} \tau_\nu^n I_1 \otimes |n', \mu_1', \nu_N\rangle\langle n', \mu_1', \nu_N| \otimes \sigma_N^\nu\Big) \\ =&\sum_{n,\mu,\nu} \lambda_\mu^n \tau_\nu^n \sigma_1^\mu \otimes |n, \mu_1, \nu_N\rangle\langle n, \mu_1, \nu_N| \otimes \sigma_N^\nu, \end{aligned} \tag{3.22}$$

where the indices μ' and ν' in the first line can be arbitrarily chosen ($\nu', \mu' = 0, x, y, z$), and hence we choose μ' and ν' in accordance with μ and ν. By tracing out the media spins, we have

$$\begin{aligned} \mathrm{tr}_{1N} e^{-\beta H_A(\sigma_1)} e^{-\beta H_B(\sigma_N)} &= \sum_{n,\mu,\nu} \lambda_\mu^n \tau_\nu^n \sigma_1^\mu \otimes \sigma_N^\nu \\ &= \sum_N \Big(\sum_\mu \lambda_\mu^n \sigma_1^\mu\Big) \otimes \Big(\sum_\nu \tau_\nu^n \sigma_N^\nu\Big). \end{aligned} \tag{3.23}$$

At this moment, we cannot say that $\sum_\mu \lambda_\mu^n \sigma_1^\mu$ and $\sum_\tau \nu_\nu^n \sigma_N^\nu$ are necessarily physical states. In the following, we prove that Eq. (3.23) can be reduced to the mixture of the product states as in the form (3.15).

For the purpose, we should pay attention to the degeneracies of the matrices $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{N\nu}$. In fact, if there are no degeneracies in the eigenspaces of all these matrices for $\mu, \nu = 0, x, y, z$, we can easily prove that each of $\sum_\mu \lambda_\mu^n \sigma_1^\mu$ and $\sum_\nu \tau_\nu^n \sigma_N^\nu$ ($n = 1, 2, \cdots, 2^{N-2}$) in Eq. (3.23) is a positive matrix. Since $\rho_{\text{media}}^{1\mu}$ and $\rho_{\text{media}}^{N\nu}$ commute with each other as well as

$\rho_{\rm media}^{1\mu'}$ and $\rho_{\rm media}^{N\nu}$ do, $\rho_{\rm media}^{1\mu}$ and $\rho_{\rm media}^{1\mu'}$ should also have simultaneous eigenstates if there are no degeneracies. If there are absolutely no degeneracies in all eigenspaces of $\rho_{\rm media}^{1\mu}$ and $\rho_{\rm media}^{N\nu}$ ($\mu, \nu = 0, x, y, z$), we have an orthonormal set of 2^{N-2} pieces of states $|n\rangle$, each of which is the simultaneous eigenstate $|n, \mu_1, \nu_N\rangle$ for all of $\mu, \nu = 0, x, y, z$. Then, we have from Eq. (3.17)

$$\sum_{\mu=0,x,y,z} \lambda_\mu^n \sigma_1^\mu = {\rm tr}_{1N}(e^{-\beta h_1^z(\sigma_1)}|n\rangle\langle n|), \tag{3.24}$$

$$\sum_{\nu=0,x,y,z} \tau_\nu^n \sigma_N^\nu = {\rm tr}_{1N}(e^{-\beta H_N(\sigma_N)}|n\rangle\langle n|), \tag{3.25}$$

for $n = 1, 2, \cdots 2^{N-2}$. This means that each of $\sum_\mu \lambda_\mu^n \sigma_1^\mu$ and $\sum_\nu \tau_\nu^n \sigma_N^\nu$ ($n = 1, 2, \cdots, 2^{N-2}$) is a positive matrix, and hence Eq. (3.23) indeed takes the form (3.15).

If there are degeneracies in some of the eigenspaces of the matrices $\rho_{\rm media}^{1\mu}$ and $\rho_{\rm media}^{N\nu}$, there is a possibility that we cannot choose a common state $|n\rangle$ that represents the simultaneous eigenstate $|n, \mu_1, \nu_N\rangle$ for $\mu, \nu = 0, x, y, z$. Let us then inspect the degeneracies in more detail.

Suppose that $\rho_{\rm media}^{1\mu}$ and $\rho_{\rm media}^{N\nu}$ share the eigenstate $|n_0, \mu_1, \nu_N\rangle$ with the respective eigenvalues $\lambda_\mu^{n_0}$ and $\tau_\nu^{n_0}$. We can choose the state $|n_0, \mu_1, \nu_N\rangle$ even when each of the eigenvalues $\lambda_\mu^{n_0}$ and $\tau_\nu^{n_0}$ has degeneracies in its own eigenspace. Suppose also that $\rho_{\rm media}^{1\mu'}$ and $\rho_{\rm media}^{N\nu}$ share the eigenstate $|n_1, \mu'_1, \nu_N\rangle$ with the respective eigenvalues $\lambda_{\mu'}^{n_1}$ and $\tau_\nu^{n_1}$. After close inspection, we can state the following: if $\tau_\nu^{n_0}$ and $\tau_\nu^{n_1}$ are not degenerate, the states $|n_0, \mu_1, \nu_N\rangle$ and $|n_1, \mu'_1, \nu_N\rangle$ are orthogonal to each other.

The only possibility that we cannot choose a common state $|n\rangle$ then occurs when all the matrices $\rho_{\rm media}^{1\mu}$ ($\mu = 0, x, y, z$) have degeneracies in the corresponding eigenspaces and/or all the matrices $\rho_{\rm media}^{N\nu}$ ($\nu = 0, x, y, z$) have degeneracies in the corresponding eigenspaces. If one of $\rho_{\rm media}^{1\mu}$ ($\mu = 0, x, y, z$) does not have degeneracies, we can rotate the states in the degenerate eigenspaces of the other matrices to obtain common eigenstates.

We can thereby break down the whole eigenspace into blocks. We form a block of eigenspace in which all the matrices $\rho_{\rm media}^{1\mu}$ ($\mu = 0, x, y, z$) and/or all the matrices $\rho_{\rm media}^{N\nu}$ ($\nu = 0, x, y, z$) have degeneracies. Let us denote each block as $\mathcal{H}_{\tilde{n}}$ with the dimensionality $D_{\tilde{n}}$. Let us choose an arbitrary orthonormal set of states $|n\rangle_{\tilde{n}}$ ($n = 1, 2, \cdots, D_{\tilde{n}}$) in the block $\mathcal{H}_{\tilde{n}}$. Then we sum the terms $\sum_\mu \lambda_\mu^n \sigma_1^\mu$ and $\sum_\nu \tau_\nu^n \sigma_N^\nu$ inside each block $H_{\tilde{n}}$ to have

$$\sum_{n:|n\rangle_{\tilde{n}} \in \mathcal{H}_{\tilde{n}}} \sum_{\mu=0,x,y,z} \lambda_\mu^n \sigma_1^\mu = \sum_{n=1}^{D_{\tilde{n}}} {\rm tr}_{1N}\left(e^{-\beta H_A(\sigma_1)}|n\rangle_{\tilde{n}}\langle n|_{\tilde{n}}\right), \tag{3.26}$$

$$\sum_{n:|n\rangle_{\tilde{n}} \in \mathcal{H}_{\tilde{n}}} \sum_{\nu=0,x,y,z} \tau_\nu^n \sigma_N^\nu = \sum_{n=1}^{D_{\tilde{n}}} {\rm tr}_{1N}\left(e^{-\beta H_B(\sigma_N)}|n\rangle_{\tilde{n}}\langle n|_{\tilde{n}}\right), \tag{3.27}$$

which proves that each left-hand side is a positive matrix. This in turn shows that Eq. (3.23) can be summarized into the form Eq. (3.15), where the summation in the right-hand side of Eq. (3.15) is taken over the blocks $\tilde{n}$. Thus, Theorem 1 is proved.

Theorem 1 gives us a necessary condition that we can generate the entanglement between the focused spins by optimizing the local fields. However, this is not a sufficient condition. For example, we can prove the following theorem for spin chains.

Theorem 2. Let us consider the XYZ chain (3.1) with $\{J_i^x, J_i^y, J_i^z\} = \{J^x, J^y, J^z\}$ ($1 \le i \le N-1$). We tune the local fields h_1^z and h_N^z, while the media fields $\{h_i^z\}_{i=2}^{N-1}$ are arbitrary but fixed. There is a critical temperature above which the maximized entanglement between the focused spins 1 and N is exactly zero if they are separated by two or more spins ($N \ge 4$). In other words, we cannot generate the entanglement for any values of the local fields above this temperature for $N \geq 4$.

Comments. This critical temperature yields a stronger restriction than the known ones [31], which discuss the entanglement disappearance under a fixed Hamiltonian. Let us denote this critical temperature as $\tilde{T}_c$. We say $\tilde{T}_c = \infty$ if we cannot generate the entanglement at any temperatures, whereas we say $\tilde{T}_c = 0$ if the maximized entanglement is equal to zero even in the ground state.

Proof. In order to prove this theorem, it is enough to show

$$\max(F_1^2 - p_{\uparrow\uparrow}p_{\downarrow\downarrow}, F_2^2 - p_{\uparrow\downarrow}p_{\downarrow\uparrow}) < 0 \tag{3.28}$$

after the maximization of the left-hand side with respect to h_1^z and h_N^z in the high temperature limit $\beta \to 0$, where $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ are the elements of the density matrix defined in Eq. (3.5). Then, Eq. (3.8) yields the exactly zero concurrence in the limit $\beta \to 0$. Since the system (3.1) has a finite number of degrees of freedom, the elements $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ must be analytic as a function of β. Therefore, there can be a finite value of β at which $\max(F_1^2 - p_{\uparrow\uparrow}p_{\downarrow\downarrow}, F_2^2 - p_{\uparrow\downarrow}p_{\downarrow\uparrow}) = 0$ after the maximization. This gives the critical temperature. Note that the elements of ρ_{1N} here are functions of $h_{1\mathrm{op}}(\beta)$, $h_{N\mathrm{op}}(\beta)$ and β.

Now, let us define the exponents κ_1 and κ_N as

$$h_{1\mathrm{op}} = O(\beta^{-\kappa_1}) \text{ and } h_{N\mathrm{op}} = O(\beta^{-\kappa_N}) \tag{3.29}$$

in the limit $\beta \to 0$, where κ_1 and κ_N are real number. We estimate the order of each element of $\{F_1^2, F_2^2\}$ and $\{p_{\uparrow\uparrow}p_{\downarrow\downarrow}, p_{\uparrow\downarrow}p_{\downarrow\uparrow}\}$ in the following three cases:

- Case (a): $\kappa_1 < 1$ and $\kappa_N < 1$.
- Case (b): $\kappa_1 \geq \kappa_N$, $\kappa_1 \geq 1$ and $\kappa_N > 0$; or $\kappa_N \geq \kappa_1$, $\kappa_1 > 0$ and $\kappa_N \geq 1$.
- Case (c): $\kappa_1 \geq 1$ and $\kappa_N \leq 0$; or $\kappa_1 \leq 0$ and $\kappa_N \geq 1$.

Notice that the three cases cover the entire space of κ_1 and κ_N.

Case (a). In this case, we can prove the inequality (3.28) by utilizing a necessary condition for the existence of the entanglement [56], which is

$$\mathrm{tr}\,\rho_{1N}^2 \geq \frac{1}{3}. \tag{3.30}$$

In the case (a), $\beta h_{1\mathrm{op}}$ and $\beta h_{N\mathrm{op}}$ are of order $\beta^{1-\kappa_1}$ and $\beta^{1-\kappa_N}$, respectively, and approach to zero in the high temperature limit $\beta \to 0$. The density matrix ρ_{1N} becomes proportional to the identity matrix because then $\beta||H_{\mathrm{tot}}|| \to 0$; hence we have

$$\lim_{\beta\to 0} \mathrm{tr}\,\rho_{1N}^2 = \frac{1}{4}. \tag{3.31}$$

Therefore, in the case(a), the entanglement between the spins 1 and N is exactly zero in the high-temperature limit.

Case (b). To simplify the problem, we consider the case of $h_{1\mathrm{op}}, h_{N\mathrm{op}} > 0$, $\kappa_1 \geq \kappa_N$, $\kappa_1 \geq 1$ and $\kappa_N > 0$, but we can prove the other cases in the same way. We define the exponent $\tilde{\kappa}$ as

$$h_{1\mathrm{op}} - h_{N\mathrm{op}} = O(\beta^{-\tilde{\kappa}}) \tag{3.32}$$

in the limit $\beta \to 0$ and consider the case

$$\tilde{\kappa} > 0 \tag{3.33}$$

in the following. We discuss the case $\tilde{\kappa} \leq 0$ in Appendix B.2. Here, in order to obtain the inequality (3.28), we prove the following; F_1 and F_2 are of order of

$$O(\beta^{\kappa_1+\kappa_N+\kappa}) \text{ and } O(\beta^{\kappa_1+\kappa_N+\kappa'}), \tag{3.34}$$

or higher, respectively, where

$$\kappa = \min(\kappa_N, 1) > 0 \text{ and } \kappa' = \min(\kappa_N, \tilde{\kappa}, 1) > 0. \tag{3.35}$$

On the other hand, $p_{\uparrow\uparrow}p_{\downarrow\downarrow}$ and $p_{\uparrow\downarrow}p_{\downarrow\uparrow}$ are both of order of

$$O(\beta^{2\kappa_1+2\kappa_N}), \tag{3.36}$$

or lower. Then, the inequality (3.28) is satisfied below a certain value of β.

In order to prove (3.34) and (3.36), we separate the total Hamiltonian as follows:

$$H_{\text{tot}} = H_1 + H_{\text{media}} + H_{\text{couple}} + H_N, \tag{3.37}$$

where

$$\begin{aligned}
&H_1 = h_{1\text{op}}\sigma_1^z, \quad H_N = h_{N\text{op}}\sigma_N^z, \\
&H_{\text{media}} = \sum_{i=2}^{N-2}(J^x\sigma_i^x\sigma_{i+1}^x + J^y\sigma_i^y\sigma_{i+1}^y + J^z\sigma_i^z\sigma_{i+1}^z) + \sum_{i=2}^{N-1} h_i^z\sigma_i^z, \\
&H_{\text{couple}} = J^x\sigma_1^x\sigma_2^x + J^y\sigma_1^y\sigma_2^y + J^z\sigma_1^z\sigma_2^z + J^x\sigma_{N-1}^x\sigma_N^x + J^y\sigma_{N-1}^y\sigma_N^y + J^z\sigma_{N-1}^z\sigma_N^z.
\end{aligned} \tag{3.38}$$

Now, we consider the term H_{couple}, which couples the focused spins and the media spins, as perturbation. The unperturbed density matrix $\rho_{\text{tot}}^{(0)}$ is given by

$$\rho_{\text{tot}}^{(0)} = e^{-\beta H_1 - \beta H_N} e^{-\beta H_{\text{media}}} \tag{3.39}$$

because H_1, H_{media} and H_N commute with each other. Because the external fields are applied in the z direction, the eigenstates of $H_1 + H_N$ are given by $\{|\uparrow_1\uparrow_N\rangle, |\uparrow_1\downarrow_N\rangle, |\downarrow_1\uparrow_N\rangle, |\downarrow_1\downarrow_N\rangle\}$ with the corresponding eigenvalues $\{-h_{1\text{op}} - h_{N\text{op}}, -h_{1\text{op}} + h_{N\text{op}}, h_{1\text{op}} - h_{N\text{op}}, h_{1\text{op}} + h_{N\text{op}}\}$; we denote these eigenvalues as $\{E_{1N}^{\uparrow\uparrow}, E_{1N}^{\uparrow\downarrow}, E_{1N}^{\downarrow\uparrow}, E_{1N}^{\downarrow\downarrow}\}$. We also give the eigenstates of H_{media} as

$$|\psi_{\text{media}}^n\rangle = s_n|\uparrow_2\rangle|\tilde{\psi}_n^{\uparrow\uparrow}\rangle|\uparrow_{N-1}\rangle + t_n|\uparrow_2\rangle|\tilde{\psi}_n^{\uparrow\downarrow}\rangle|\downarrow_{N-1}\rangle + u_n|\downarrow_2\rangle|\tilde{\psi}_n^{\downarrow\uparrow}\rangle|\uparrow_{N-1}\rangle + w_n|\downarrow_2\rangle|\tilde{\psi}_n^{\downarrow\downarrow}\rangle|\downarrow_{N-1}\rangle, \tag{3.40}$$

for $n = 1, 2, \cdots, 2^{N-2}$, where $\{|\tilde{\psi}_n^{\uparrow\uparrow}\rangle, |\tilde{\psi}_n^{\uparrow\downarrow}\rangle, |\tilde{\psi}_n^{\downarrow\uparrow}\rangle, |\tilde{\psi}_n^{\downarrow\downarrow}\rangle\}$ are the states of the spins from 3 to $N-2$. Because the total Hamiltonian H_{tot} is a real matrix, the parameters $\{s_n, t_n, u_n, w_n\}$ are real numbers. We define the corresponding eigenvalues of H_{media} as $\{E_{\text{media}}^n\}$. Then, the unperturbed eigenstates are given by

$$\{|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\text{media}}^n\rangle, |\uparrow_1\downarrow_N\rangle \otimes |\psi_{\text{media}}^n\rangle, |\uparrow_1\downarrow_N\rangle \otimes |\psi_{\text{media}}^n\rangle, |\downarrow_1\downarrow_N\rangle \otimes |\psi_{\text{media}}^n\rangle\}. \tag{3.41}$$

We define the perturbed eigenstates corresponding to each of (3.41) as $\{|\psi_{\text{tot}}^{n,\uparrow\uparrow}\rangle, |\psi_{\text{tot}}^{n,\uparrow\downarrow}\rangle, |\psi_{\text{tot}}^{n,\downarrow\uparrow}\rangle, |\psi_{\text{tot}}^{n,\downarrow\downarrow}\rangle\}$, respectively. We express them as

$$|\psi_{\text{tot}}^{n,\xi}\rangle = |\uparrow_1\uparrow_N\rangle \otimes |\psi_{\text{media},\uparrow\uparrow}^{n,\xi}\rangle + |\uparrow_1\downarrow_N\rangle \otimes |\psi_{\text{media},\uparrow\downarrow}^{n,\xi}\rangle + |\downarrow_1\uparrow_N\rangle \otimes |\psi_{\text{media},\downarrow\uparrow}^{n,\xi}\rangle + |\downarrow_1\downarrow_N\rangle \otimes |\psi_{\text{media},\downarrow\downarrow}^{n,\xi}\rangle, \tag{3.42}$$

for $n = 1, 2, \cdots, 2^{N-2}$ and $\xi =\uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow, \downarrow\downarrow$, where $|\psi_{\text{media},\uparrow\uparrow}^{n,\xi}\rangle$, $|\psi_{\text{media},\uparrow\downarrow}^{n,\xi}\rangle$, $|\psi_{\text{media},\downarrow\uparrow}^{n,\xi}\rangle$ and $|\psi_{\text{media},\downarrow\downarrow}^{n,\xi}\rangle$ are the states of the spins from 2 to $N-1$ and may be not normalized. We also define the corresponding perturbed eigenvalues as $\{E_{\text{tot}}^{n,\uparrow\uparrow}, E_{\text{tot}}^{n,\uparrow\downarrow}, E_{\text{tot}}^{n,\downarrow\uparrow}, E_{\text{tot}}^{n,\downarrow\downarrow}\}$, which we express as

$$E_{\text{tot}}^{n,\xi} = E_{1N}^{\xi} + E_{\text{media}}^{n} + \delta E_{\text{tot}}^{n,\xi} \tag{3.43}$$

for $n = 1, 2, \cdots, 2^{N-2}$ and $\xi =\uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow, \downarrow\downarrow$. Note that $\{E_{\text{media}}^{n}\}$ and $\{\delta E_{\text{tot}}^{n,\xi}\}$ are of order β^0 because H_{media} and H_{couple} do not depend on the temperature β. Then, we have

$$\frac{e^{-\beta E_{\text{tot}}^{n,\uparrow\uparrow}}}{Z_{\text{tot}}} = \frac{e^{\beta(h_{1\text{op}}+h_{N\text{op}})-\beta(E_{\text{media}}^{n}+\delta E_{\text{tot}}^{n,\uparrow\uparrow})}}{Z_{\text{tot}}} = O(\beta^0) \tag{3.44}$$

in the limit $\beta \to 0$ in the case $h_{1\text{op}}, h_{N\text{op}} > 0$, where Z_{tot} is the partition function of the total Hamiltonian.

Now, we show the outline of the proof. First, the elements $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ can be calculated from (3.42) and (3.43) as

$$\begin{aligned} F_1 &= \frac{1}{Z_{\text{tot}}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\text{tot}}^{n,\xi}} \langle \psi_{\text{media},\uparrow\downarrow}^{n,\xi} | \psi_{\text{media},\downarrow\uparrow}^{n,\xi} \rangle, \\ F_2 &= \frac{1}{Z_{\text{tot}}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\text{tot}}^{n,\xi}} \langle \psi_{\text{media},\uparrow\uparrow}^{n,\xi} | \psi_{\text{media},\downarrow\downarrow}^{n,\xi} \rangle \end{aligned} \tag{3.45}$$

and

$$\begin{aligned} p_{\uparrow\uparrow} &= \frac{1}{Z_{\text{tot}}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\text{tot}}^{n,\xi}} \langle \psi_{\text{media},\uparrow\uparrow}^{n,\xi} | \psi_{\text{media},\uparrow\uparrow}^{n,\xi} \rangle, \\ p_{\uparrow\downarrow} &= \frac{1}{Z_{\text{tot}}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\text{tot}}^{n,\xi}} \langle \psi_{\text{media},\uparrow\downarrow}^{n,\xi} | \psi_{\text{media},\uparrow\downarrow}^{n,\xi} \rangle, \\ p_{\downarrow\uparrow} &= \frac{1}{Z_{\text{tot}}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\text{tot}}^{n,\xi}} \langle \psi_{\text{media},\downarrow\uparrow}^{n,\xi} | \psi_{\text{media},\downarrow\uparrow}^{n,\xi} \rangle, \\ p_{\downarrow\downarrow} &= \frac{1}{Z_{\text{tot}}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\text{tot}}^{n,\xi}} \langle \psi_{\text{media},\downarrow\downarrow}^{n,\xi} | \psi_{\text{media},\downarrow\downarrow}^{n,\xi} \rangle. \end{aligned} \tag{3.46}$$

Second, we estimate the leading orders of $\{F_1, F_2\}$ and $\{p_{\uparrow\downarrow}p_{\downarrow\uparrow}, p_{\uparrow\uparrow}p_{\downarrow\downarrow}\}$. From the perturbation theory, we can obtain the approximate forms of $\{|\psi_{\text{tot}}^{n,\xi}\rangle\}$ and expand $\{F_1, F_2\}$ and $\{p_{\uparrow\downarrow}p_{\downarrow\uparrow}, p_{\uparrow\uparrow}p_{\downarrow\downarrow}\}$ with respect to β. The elements $\{F_1, F_2\}$ are additive with respect to the indices n and ξ; we define each contribution to the elements $\{F_1, F_2\}$ as

$$\begin{aligned} F_1^{n,\xi} &= \langle \psi_{\text{media},\uparrow\downarrow}^{n,\xi} | \psi_{\text{media},\downarrow\uparrow}^{n,\xi} \rangle, \\ F_2^{n,\xi} &= \langle \psi_{\text{media},\uparrow\uparrow}^{n,\xi} | \psi_{\text{media},\downarrow\downarrow}^{n,\xi} \rangle. \end{aligned} \tag{3.47}$$

Then, the elements $\{F_1, F_2\}$ are given by

$$F_1 = \frac{1}{Z_{\rm tot}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\rm tot}^{n,\xi}} F_1^{n,\xi},$$
$$F_2 = \frac{1}{Z_{\rm tot}} \sum_{n=1}^{2^{N-2}} \sum_{\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\rm tot}^{n,\xi}} F_2^{n,\xi}, \tag{3.48}$$

respectively. The elements $\{p_{\uparrow\downarrow}p_{\downarrow\uparrow}, p_{\uparrow\uparrow}p_{\downarrow\downarrow}\}$, on the other hand, are given by double summations as

$$p_{\uparrow\downarrow}p_{\downarrow\uparrow} = \frac{1}{Z_{\rm tot}^2} \sum_{n,n'=1}^{2^{N-2}} \sum_{\xi,\xi'=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\rm tot}^{n,\xi}} \big\langle \psi_{{\rm media},\uparrow\downarrow}^{n,\xi} \big| \psi_{{\rm media},\uparrow\downarrow}^{n,\xi} \big\rangle e^{-\beta E_{\rm tot}^{n',\xi'}} \big\langle \psi_{{\rm media},\downarrow\uparrow}^{n',\xi'} \big| \psi_{{\rm media},\downarrow\uparrow}^{n',\xi'} \big\rangle,$$
$$p_{\uparrow\uparrow}p_{\downarrow\downarrow} = \frac{1}{Z_{\rm tot}^2} \sum_{n,n'=1}^{2^{N-2}} \sum_{\xi,\xi'=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow} e^{-\beta E_{\rm tot}^{n,\xi}} \big\langle \psi_{{\rm media},\uparrow\uparrow}^{n,\xi} \big| \psi_{{\rm media},\downarrow\downarrow}^{n,\xi} \big\rangle e^{-\beta E_{\rm tot}^{n',\xi'}} \big\langle \psi_{{\rm media},\downarrow\downarrow}^{n',\xi'} \big| \psi_{{\rm media},\downarrow\downarrow}^{n',\xi'} \big\rangle. \tag{3.49}$$

Because the summands of (3.49) are all positive, we can obtain the following inequalities for $\{p_{\uparrow\downarrow}p_{\downarrow\uparrow}, p_{\uparrow\uparrow}p_{\downarrow\downarrow}\}$:

$$p_{\uparrow\downarrow}p_{\downarrow\uparrow} \geq \frac{1}{Z_{\rm tot}^2} \sum_{n=1}^{2^{N-2}} e^{-\beta E_{\rm tot}^{n,\uparrow\uparrow}} \big\langle \psi_{{\rm media},\uparrow\downarrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\uparrow\downarrow}^{n,\uparrow\uparrow} \big\rangle e^{-\beta E_{\rm tot}^{n,\uparrow\uparrow}} \big\langle \psi_{{\rm media},\downarrow\uparrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\downarrow\uparrow}^{n,\uparrow\uparrow} \big\rangle \equiv P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow},$$
$$p_{\uparrow\uparrow}p_{\downarrow\downarrow} \geq \frac{1}{Z_{\rm tot}^2} \sum_{n=1}^{2^{N-2}} e^{-\beta E_{\rm tot}^{n,\uparrow\uparrow}} \big\langle \psi_{{\rm media},\uparrow\uparrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\uparrow\uparrow}^{n,\uparrow\uparrow} \big\rangle e^{-\beta E_{\rm tot}^{n,\uparrow\uparrow}} \big\langle \psi_{{\rm media},\downarrow\downarrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\downarrow\downarrow}^{n,\uparrow\uparrow} \big\rangle \equiv P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow}, \tag{3.50}$$

where we pick up only the terms of $n = n'$ and $\xi = \xi' =\uparrow\uparrow$ from the summations in (3.49). The elements $P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow}$ and $P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow}$ are additive with respect to n; we define each contribution to the elements $\{P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow}, P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow}\}$ as

$$P_{\uparrow\downarrow,\downarrow\uparrow}^{n,\uparrow\uparrow} = \big\langle \psi_{{\rm media},\uparrow\downarrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\uparrow\downarrow}^{n,\uparrow\uparrow} \big\rangle \big\langle \psi_{{\rm media},\downarrow\uparrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\downarrow\uparrow}^{n,\uparrow\uparrow} \big\rangle,$$
$$P_{\uparrow\uparrow,\downarrow\downarrow}^{n,\uparrow\uparrow} = \big\langle \psi_{{\rm media},\uparrow\uparrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\uparrow\uparrow}^{n,\uparrow\uparrow} \big\rangle \big\langle \psi_{{\rm media},\downarrow\downarrow}^{n,\uparrow\uparrow} \big| \psi_{{\rm media},\downarrow\downarrow}^{n,\uparrow\uparrow} \big\rangle. \tag{3.51}$$

Then, the elements $\{P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow}, P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow}\}$ are given by

$$P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow} = \frac{1}{Z_{\rm tot}^2} \sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} P_{\uparrow\downarrow,\downarrow\uparrow}^{n,\uparrow\uparrow},$$
$$P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow} = \frac{1}{Z_{\rm tot}^2} \sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} P_{\uparrow\uparrow,\downarrow\downarrow}^{n,\uparrow\uparrow}, \tag{3.52}$$

respectively. In this way, we calculate each contribution to the elements $\{F_1, F_2\}$ and $\{P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow}, P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow}\}$ separately.

In accordance with the above outline, we first calculate $|\psi_{\rm tot}^{n_0,\uparrow\uparrow}\rangle$, namely the perturbed state of $|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle$. In order to simplify the calculation, we rewrite the perturbation

Hamiltonian $H_{\rm couple}$ as follows;

$$\begin{aligned}H_{\rm couple} =& \frac{1}{2}\Big\{ J\big[(1+\gamma)\sigma_1^x\sigma_2^x + (1-\gamma)\sigma_1^y\sigma_2^y\big] + J^z\sigma_1^z\sigma_2^z \\ &+ J\big[(1+\gamma)\sigma_{N-1}^x\sigma_N^x + (1-\gamma)\sigma_{N-1}^y\sigma_N^y\big] + J^z\sigma_{N-1}^z\sigma_N^z\Big\} \\ =& \Big\{ J\big[\sigma_1^+\sigma_2^- + \sigma_1^-\sigma_2^+ + \gamma(\sigma_1^+\sigma_2^+ + \sigma_1^-\sigma_2^-)\big] + \frac{1}{2}J^z\sigma_1^z\sigma_2^z \\ &+ J\big[\sigma_{N-1}^+\sigma_N^- + \sigma_{N-1}^-\sigma_N^+ + \gamma(\sigma_{N-1}^+\sigma_N^+ + \sigma_{N-1}^-\sigma_N^-)\big] + \frac{1}{2}J^z\sigma_{N-1}^z\sigma_N^z\Big\}, \end{aligned} \tag{3.53}$$

where

$$J = J^x + J^y, \quad \gamma = \frac{J^x - J^y}{J_x + J_y}. \tag{3.54}$$

From the calculation in Appendix B.3, the leading terms of the elements $\{|\psi^{n_0,\uparrow\uparrow}_{{\rm media},\xi}\rangle\}$ for $\xi = \uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow, \downarrow\downarrow$ in Eq. (3.42) are given by

$$|\psi^{n_0,\uparrow\uparrow}_{{\rm media},\uparrow\uparrow}\rangle = |\psi^{n_0}_{\rm media}\rangle, \tag{3.55}$$

$$\begin{aligned}|\psi^{n_0,\uparrow\uparrow}_{{\rm media},\uparrow\downarrow}\rangle =& \frac{J}{-2h_{N{\rm op}}}\Big(\gamma s_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|\downarrow_{N-1}\rangle + t_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|\uparrow_{N-1}\rangle \\ &+ \gamma u_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|\downarrow_{N-1}\rangle + w_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|\uparrow_{N-1}\rangle + O(\beta^{\kappa_N})\Big),\end{aligned} \tag{3.56}$$

$$\begin{aligned}|\psi^{n_0,\uparrow\uparrow}_{{\rm media},\downarrow\uparrow}\rangle =& \frac{J}{-2h_{1{\rm op}}}\Big(\gamma s_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|\uparrow_{N-1}\rangle + \gamma t_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|\downarrow_{N-1}\rangle \\ &+ u_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|\uparrow_{N-1}\rangle + w_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|\downarrow_{N-1}\rangle + O(\beta^{\kappa_1})\Big)\end{aligned} \tag{3.57}$$

and

$$\begin{aligned}|\psi^{n_0,\uparrow\uparrow}_{{\rm media},\downarrow\downarrow}\rangle =& \frac{J^2}{4h_{1{\rm op}}h_{N{\rm op}}}\Big(\gamma^2 s_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|\downarrow_{N-1}\rangle + \gamma t_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|\uparrow_{N-1}\rangle \\ &+ \gamma u_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|\downarrow_{N-1}\rangle + w_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|\uparrow_{N-1}\rangle + O(\beta^{\kappa_N})\Big).\end{aligned} \tag{3.58}$$

Now, we calculate the contribution of $|\psi^{n_0,\uparrow\uparrow}_{\rm tot}\rangle$ to the elements $\{F_1, F_2\}$ and $\{P^{\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}, P^{\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}\}$, which are defined as $\{F_1^{n_0,\uparrow\uparrow}, F_2^{n_0,\uparrow\uparrow}\}$ and $\{P^{n_0,\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}, P^{n_0,\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}\}$. From Eqs. (3.55)–(3.58), we have the elements $\{F_1^{n_0,\uparrow\uparrow}, F_2^{n_0,\uparrow\uparrow}\}$ as

$$\begin{aligned}F_1^{n_0,\uparrow\uparrow} =& \frac{J^2}{4h_{1{\rm op}}h_{N{\rm op}}}\Big(\gamma s_{n_0}w_{n_0}\langle\tilde{\psi}^{\downarrow\downarrow}_{n_0}|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle + u_{n_0}t_{n_0}\langle\tilde{\psi}^{\uparrow\downarrow}_{n_0}|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle \\ &+ \gamma^2 t_{n_0}u_{n_0}\langle\tilde{\psi}^{\downarrow\uparrow}_{n_0}|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle + \gamma w_{n_0}s_{n_0}\langle\tilde{\psi}^{\uparrow\uparrow}_{n_0}|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle + O(\beta^{\kappa_N})\Big)\end{aligned} \tag{3.59}$$

and

$$\begin{aligned}F_2^{n_0,\uparrow\uparrow} =& \frac{J^2}{4h_{1{\rm op}}h_{N{\rm op}}}\Big(\gamma^2 w_{n_0}s_{n_0}\langle\tilde{\psi}^{\downarrow\downarrow}_{n_0}|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle + \gamma u_{n_0}t_{n_0}\langle\tilde{\psi}^{\downarrow\uparrow}_{n_0}|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle \\ &+ \gamma t_{n_0}u_{n_0}\langle\tilde{\psi}^{\uparrow\downarrow}_{n_0}|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle + s_{n_0}w_{n_0}\langle\tilde{\psi}^{\uparrow\uparrow}_{n_0}|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle + O(\beta^{\kappa_N})\Big)\end{aligned} \tag{3.60}$$

as well as the elements $\{P^{n_0,\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}, P^{n_0,\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}\}$ in the forms

$$\begin{aligned}
P^{n_0,\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow} &= \frac{J^2}{16h_{1\mathrm{op}}^2 h_{N\mathrm{op}}^2}\Big(\gamma^2 s_{n_0}^2 + t_{n_0}^2 + \gamma^2 u_{n_0}^2 + w_{n_0}^2 + O\big(\beta^{\kappa_N}\big)\Big) \\
&\qquad\qquad \times \Big(\gamma^2 s_{n_0}^2 + \gamma^2 t_{n_0}^2 + u_{n_0}^2 + w_{n_0}^2 + O(\beta^{\kappa_N})\Big) \\
&\geq \frac{J^2}{16h_{1\mathrm{op}}^2 h_{N\mathrm{op}}^2}(w_{n_0}^4 + \beta^{\kappa_N}\delta P^{n_0,\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow})
\end{aligned} \tag{3.61}$$

and

$$\begin{aligned}
P^{n_0,\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow} &= \frac{J^2}{16h_{1\mathrm{op}}^2 h_{N\mathrm{op}}^2}\Big(\gamma^4 s_{n_0}^2 + \gamma^2 t_{n_0}^2 + \gamma^2 u_{n_0}^2 + w_{n_0}^2 + O(\beta^{\kappa_N})\Big) \\
&\geq \frac{J^2 w_{n_0}^2}{16h_{1\mathrm{op}}^2 h_{N\mathrm{op}}^2}(w_{n_0}^2 + \beta^{\kappa_N}\delta P^{n_0,\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow})
\end{aligned} \tag{3.62}$$

in the limit $\beta \to 0$, where we define the real numbers of order β^0 as $\delta P^{n_0,\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}$ and $\delta P^{n_0,\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}$ so as to satisfy the above inequalities.

Then, we sum the elements $\{F^{n,\uparrow\uparrow}_1, F^{n,\uparrow\uparrow}_2\}$ and $\{P^{n,\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}, P^{n,\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}\}$ with the Boltzmann weight $e^{-\beta E^{n,\uparrow\uparrow}_{\mathrm{tot}}}$ over the label n. First, we calculate the summation of $\{F^{n,\uparrow\uparrow}_1, F^{n,\uparrow\uparrow}_2\}$. Because the spins 2 and $(N-1)$ are separated by $(N-4)$ spins, the correlation between the spins 2 and $(N-1)$ are generated by the $(N-3)$th-order perturbation of H_{media}. Therefore, we obtain

$$\begin{aligned}
\langle \sigma_2^x \sigma_{N-1}^x \rangle_0 &= O(\beta^{\alpha_1}), \\
\langle \sigma_2^y \sigma_{N-1}^y \rangle_0 &= O(\beta^{\alpha_2}),
\end{aligned} \tag{3.63}$$

where $\langle \cdots \rangle_0$ denotes the thermal average with respect to $\rho^{(0)}_{\mathrm{tot}}$ in (3.39) and $\alpha_1 \geq N-3$, $\alpha_2 \geq N-3$. Since we are considering the case $N \geq 4$, we have $\alpha_1 \geq 1$ and $\alpha_2 \geq 1$. From the equations

$$\begin{aligned}
&\frac{\langle \sigma_2^x \sigma_{N-1}^x + \sigma_2^y \sigma_{N-1}^y \rangle_0}{4} \\
=&\frac{\langle \sigma_2^+ \sigma_{N-1}^- + \sigma_2^- \sigma_{N-1}^+ \rangle_0}{2} \\
=&\mathrm{tr}\bigg(e^{-\beta H_{\mathrm{media}}}\frac{|\downarrow_2\uparrow_{N-1}\rangle\langle\uparrow_2\downarrow_{N-1}| + |\uparrow_2\downarrow_{N-1}\rangle\langle\downarrow_2\uparrow_{N-1}|}{2}\bigg) \\
=&\mathrm{tr}\Big(e^{-\beta H_{\mathrm{media}}}|\downarrow_2\uparrow_{N-1}\rangle\langle\uparrow_2\downarrow_{N-1}|\Big) \\
=&\frac{1}{Z_{\mathrm{media}}}\sum_{n=1}^{2^{N-2}} e^{-\beta E^n_{\mathrm{media}}}\big\langle\uparrow_2\downarrow_{N-1}\,\big|\psi^n_{\mathrm{media}}\big\rangle\big\langle\psi^n_{\mathrm{media}}\big|\downarrow_2\uparrow_{N-1}\big\rangle
\end{aligned} \tag{3.64}$$

and

$$
\begin{aligned}
&\frac{\langle\sigma_2^x\sigma_{N-1}^x-\sigma_2^y\sigma_{N-1}^y\rangle_0}{4}\\
=&\frac{\langle\sigma_2^+\sigma_{N-1}^+ +\sigma_2^-\sigma_{N-1}^-\rangle_0}{2}\\
=&\mathrm{tr}\Big(e^{-\beta H_{\mathrm{media}}}\frac{|{\downarrow_2}{\downarrow_{N-1}}\rangle\langle{\uparrow_2}{\uparrow_{N-1}}|+|{\uparrow_2}{\uparrow_{N-1}}\rangle\langle{\downarrow_2}{\downarrow_{N-1}}|}{2}\Big)\\
=&\mathrm{tr}\Big(e^{-\beta H_{\mathrm{media}}}|{\uparrow_2}{\uparrow_{N-1}}\rangle\langle{\downarrow_2}{\downarrow_{N-1}}|\Big)\\
=&\frac{1}{Z_{\mathrm{media}}}\sum_{n=1}^{2^{N-2}}e^{-\beta E^n_{\mathrm{media}}}\big\langle{\downarrow_2}{\downarrow_{N-1}}\,\big|\psi^n_{\mathrm{media}}\big\rangle\big\langle\psi^n_{\mathrm{media}}\big|{\uparrow_2}{\uparrow_{N-1}}\big\rangle,
\end{aligned}
\tag{3.65}
$$

where $Z_{\mathrm{media}}\equiv\mathrm{tr}(e^{-\beta H_{\mathrm{media}}})$, $\sigma^+\equiv(\sigma^x+i\sigma^y)/2$ and $\sigma^-\equiv(\sigma^x-i\sigma^y)/2$, we also have

$$
\sum_{n=1}^{2^{N-2}}e^{-\beta E^n_{\mathrm{media}}}t_nu_n\langle\tilde\psi_n^{\uparrow\downarrow}|\tilde\psi_n^{\downarrow\uparrow}\rangle=\sum_{n=1}^{2^{N-2}}e^{-\beta E^n_{\mathrm{media}}}t_nu_n\langle\tilde\psi_n^{\downarrow\uparrow}|\tilde\psi_n^{\uparrow\downarrow}\rangle=O(\beta^\alpha)
\tag{3.66}
$$

and

$$
\sum_{n=1}^{2^{N-2}}e^{-\beta E^n_{\mathrm{media}}}s_nw_n\langle\tilde\psi_n^{\uparrow\uparrow}|\tilde\psi_n^{\downarrow\downarrow}\rangle=\sum_{n=1}^{2^{N-2}}e^{-\beta E^n_{\mathrm{media}}}s_nw_n\langle\tilde\psi_n^{\downarrow\downarrow}|\tilde\psi_n^{\uparrow\uparrow}\rangle=O(\beta^\alpha),
\tag{3.67}
$$

where $\alpha=\min(\alpha_1,\alpha_2)$. Moreover, because $\{\delta E^{n,\xi}_{\mathrm{tot}}\}$ in Eq. (3.43) are of order β^0, we have

$$
e^{-\beta E^{n,\xi}_{\mathrm{tot}}}=e^{-\beta(E^\xi_{1N}+E^n_{\mathrm{media}})}\Big(1-\beta\delta E^{n,\xi}_{\mathrm{tot}}+O(\beta^2)\Big),
\tag{3.68}
$$

for $n=1,2,\cdots,2^{N-2}$ and $\xi={\uparrow\uparrow},{\uparrow\downarrow},{\downarrow\uparrow},{\downarrow\downarrow}$. Then, we obtain from Eqs. (3.59), (3.60), and (3.66)–(3.68),

$$
\begin{aligned}
&\sum_{n=1}^{2^{N-2}}e^{-\beta E^{n,\uparrow\uparrow}_{\mathrm{tot}}}F_1^{n,\uparrow\uparrow}\\
=&\sum_{n=1}^{2^{N-2}}e^{-\beta(E^\xi_{1N}+E^n_{\mathrm{media}})}F_1^{n,\uparrow\uparrow}+\sum_{n=1}^{2^{N-2}}e^{-\beta(E^\xi_{1N}+E^n_{\mathrm{media}})}\big(-\beta\delta E^{n,\xi}_{\mathrm{tot}}+O(\beta^2)\big)F_1^{n,\uparrow\uparrow}\\
=&e^{-\beta E^{\uparrow\uparrow}_{1N}}\frac{J^2}{4h_{1\mathrm{op}}h_{N\mathrm{op}}}\Big(O(\beta^\alpha)+O(\beta^{\kappa_N})\Big)+e^{-\beta E^{\uparrow\uparrow}_{1N}}\frac{J^2}{4h_{1\mathrm{op}}h_{N\mathrm{op}}}O(\beta^\alpha)\\
=&e^{-\beta E^{\uparrow\uparrow}_{1N}}\times O(\beta^{\kappa_1+\kappa_N+\kappa})
\end{aligned}
\tag{3.69}
$$

and

$$
\sum_{n=1}^{2^{N-2}}e^{-\beta E^{n,\uparrow\uparrow}_{\mathrm{tot}}}F_2^{n,\uparrow\uparrow}=e^{-\beta E^{\uparrow\uparrow}_{1N}}\frac{J^2}{4h_{1\mathrm{op}}h_{N\mathrm{op}}}\Big(O(\beta^\alpha)+O(\beta^{\kappa_N})+O(\beta)\Big)=e^{-\beta E^{\uparrow\uparrow}_{1N}}\times O(\beta^{\kappa_1+\kappa_N+\kappa}),
\tag{3.70}
$$

where $\alpha=\min(\alpha_1,\alpha_2)\geq 1$ and κ is defined in (3.35).

We can similarly calculate the contributions of the other states $\{|\psi_{\rm tot}^{n,\xi}\rangle\}$ ($\xi=\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow$). From Eqs. (B.39) and (B.42) in Appendix B.3, we have the contributions of the states $\{|\psi_{\rm tot}^{n,\uparrow\downarrow}\rangle\}$ to F_1 and F_2 as

$$\sum_{n=1}^{2^{N-2}} e^{-\beta E_{\rm tot}^{n,\uparrow\downarrow}} F_1^{n,\uparrow\downarrow} = e^{-\beta E_{1N}^{\uparrow\downarrow}} \frac{J^2}{4h_{1\rm op}h_{N\rm op}} \Big(O(\beta^{\alpha})+O(\beta^{\kappa_N})+O(\beta^{\tilde{\kappa}})+O(\beta)\Big) = e^{-\beta E_{1N}^{\uparrow\downarrow}} \times O(\beta^{\kappa_1+\kappa_N+\kappa'}),$$
$$\sum_{n=1}^{2^{N-2}} e^{-\beta E_{\rm tot}^{n,\uparrow\downarrow}} F_2^{n,\uparrow\downarrow} = e^{-\beta E_{1N}^{\uparrow\downarrow}} \frac{J^2}{4h_{1\rm op}h_{N\rm op}} \Big(O(\beta^{\alpha})+O(\beta^{\kappa_N})+O(\beta)\Big) = e^{-\beta E_{1N}^{\uparrow\downarrow}} \times O(\beta^{\kappa_1+\kappa_N+\kappa}), \tag{3.71}$$

where we utilized Eqs. (3.66)–(3.68) and κ' is defined in (3.35). From the inequality

$$\frac{e^{-\beta E_{1N}^{\xi}}}{Z_{\rm tot}} < 1 \tag{3.72}$$

for $\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow$, we finally obtain (3.34) substituting Eqs. (3.69)–(3.71) into Eq. (3.48).

Second, we calculate the summation of $\{P_{\uparrow\downarrow,\downarrow\uparrow}^{n,\uparrow\uparrow}, P_{\uparrow\uparrow,\downarrow\downarrow}^{n,\uparrow\uparrow}\}$. The parameter w_n in Eq. (3.40) cannot vanish for all n. Therefore, we have

$$\frac{1}{Z_{\rm tot}^2}\sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} w_n^4 = O(\beta^0),$$
$$\frac{1}{Z_{\rm tot}^2}\sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} w_n^2 = O(\beta^0), \tag{3.73}$$

where we utilized Eq. (3.44). Hence we obtain

$$\begin{aligned} P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow} &= \frac{1}{Z_{\rm tot}^2}\sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} P_{\uparrow\downarrow,\downarrow\uparrow}^{n,\uparrow\uparrow} \\ &\geq \frac{J^2}{16h_{1\rm op}^2h_{N\rm op}^2}\frac{1}{Z_{\rm tot}^2}\sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} (w_n^4+\beta^{\kappa_N}\delta P_{\uparrow\downarrow,\downarrow\uparrow}^{n,\uparrow\uparrow}) = O(\beta^{2\kappa_1+2\kappa_N}) \end{aligned} \tag{3.74}$$

and

$$\begin{aligned} P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow} &= \frac{1}{Z_{\rm tot}^2}\sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} P_{\uparrow\uparrow,\downarrow\downarrow}^{n,\uparrow\uparrow} \\ &\geq \frac{J^2}{16h_{1\rm op}^2h_{N\rm op}^2}\frac{1}{Z_{\rm tot}^2}\sum_{n=1}^{2^{N-2}} e^{-2\beta E_{\rm tot}^{n,\uparrow\uparrow}} (w_n^2+\beta^{\kappa_N}\delta P_{\uparrow\uparrow,\downarrow\downarrow}^{n,\uparrow\uparrow}) = O(\beta^{2\kappa_1+2\kappa_N}). \end{aligned} \tag{3.75}$$

Because $p_{\uparrow\downarrow}p_{\downarrow\uparrow} \geq P_{\uparrow\downarrow,\downarrow\uparrow}^{\uparrow\uparrow}$ and $p_{\uparrow\uparrow}p_{\downarrow\downarrow} \geq P_{\uparrow\uparrow,\downarrow\downarrow}^{\uparrow\uparrow}$, we obtain (3.36). Thus, the inequality (3.28) is satisfied below a certain value of β in the case (b).

Case (c). To simplify the problem, we consider the case of $h_1>0$, $\kappa_1\geq 1$ and $\kappa_N\leq 0$, but we can prove the other cases in the same way. In this case, we prove the following; F_1^2 and F_2^2 are both of order of

$$O(\beta^{2\kappa_1+2}), \tag{3.76}$$

or higher. On the other hand, $p_{\uparrow\uparrow}p_{\downarrow\downarrow}$ and $p_{\uparrow\downarrow}p_{\downarrow\uparrow}$ are both of order of

$$O(\beta^{2\kappa_1}), \tag{3.77}$$

or lower. Then, the inequality (3.28) is satisfied below a certain value of β.

In a similar manner to the case (b), we separate the total Hamiltonian as follows:

$$H_{\rm tot} = H_1 + H'_{\rm media} + H'_{\rm couple}, \tag{3.78}$$

where

$$\begin{aligned} H_1 &= h_{1\rm op}\sigma_1^z, \\ H'_{\rm media} &= \sum_{i=2}^{N-1}(J^x\sigma_i^x\sigma_{i+1}^x + J^y\sigma_i^y\sigma_{i+1}^y + J^z\sigma_i^z\sigma_{i+1}^z) + \sum_{i=2}^{N-1} h_i^z\sigma_i^z + h_{N\rm op}\sigma_N^z, \\ H'_{\rm couple} &= J^x\sigma_1^x\sigma_2^x + J^y\sigma_1^y\sigma_2^y + J^z\sigma_1^z\sigma_2^z. \end{aligned} \tag{3.79}$$

We consider the interaction term $H'_{\rm couple}$, which couples the spins 1 and the other spins, as perturbation. In the case (c), the norm of the Hamiltonian $H'_{\rm media}$ is of order β^0 because $\kappa_N < 0$. Then, the unperturbed density matrix $\rho_{\rm tot}^{'(0)}$ is expressed as

$$\rho_{\rm tot}^{'(0)} = e^{-\beta H_1}e^{-\beta H'_{\rm media}}. \tag{3.80}$$

The eigenstates of H_1 can be given by $\{|\uparrow_1\rangle, |\downarrow_1\rangle\}$ with the corresponding eigenvalues $\{-h_{1\rm op}, h_{1\rm op}\}$; we denote these eigenvalues as $\{E_1^\uparrow, E_1^\downarrow\}$. We also define the eigenstates of $H'_{\rm media}$ as

$$|\phi^n_{\rm media}\rangle = s'_n|\uparrow_2\rangle|\tilde{\phi}_n^{\uparrow\uparrow}\rangle|\uparrow_N\rangle + t'_n|\uparrow_2\rangle|\tilde{\phi}_n^{\uparrow\downarrow}\rangle|\downarrow_N\rangle + u'_n|\downarrow_2\rangle|\tilde{\phi}_n^{\downarrow\uparrow}\rangle|\uparrow_N\rangle + w'_n|\downarrow_2\rangle|\tilde{\phi}_n^{\downarrow\downarrow}\rangle|\downarrow_N\rangle \tag{3.81}$$

for $n = 1, 2, \cdots 2^{N-1}$, where $\{|\tilde{\phi}_n^{\uparrow\uparrow}\rangle, |\tilde{\phi}_n^{\uparrow\downarrow}\rangle, |\tilde{\phi}_n^{\downarrow\uparrow}\rangle, |\tilde{\phi}_n^{\downarrow\downarrow}\rangle\}$ are the states of the spins from 3 to $N-1$. Because the total Hamiltonian $H_{\rm tot}$ is a real matrix, the parameters $\{s'_n, t'_n, u'_n, w'_n\}$ are real numbers. We define the corresponding eigenvalues of $H'_{\rm media}$ as $\{E'^n_{\rm media}\}$. Then, the unperturbed eigenstates are given by $\{|\uparrow_1\rangle \otimes |\phi^n_{\rm media}\rangle\}$ and $\{|\downarrow_1\rangle \otimes |\phi^n_{\rm media}\rangle\}$. We define the corresponding perturbed eigenstates as $\{|\phi^{n,\uparrow}_{\rm tot}\rangle\}$ and $\{|\phi^{n,\downarrow}_{\rm tot}\rangle\}$ and the corresponding perturbed eigenvalues as $\{E^{n,\uparrow}_{\rm tot}, E^{n,\downarrow}_{\rm tot}\}$. We express them in the forms

$$|\phi^{n,\eta}_{\rm tot}\rangle = |\uparrow_1\rangle \otimes |\phi^{n,\eta}_{{\rm media},\uparrow}\rangle + |\downarrow_1\rangle \otimes |\phi^{n,\eta}_{{\rm media},\downarrow}\rangle \tag{3.82}$$

and

$$E'^{n,\eta}_{\rm tot} = E_1^\eta + E'^n_{\rm media} + \delta E'^{n,\eta}_{\rm tot} \tag{3.83}$$

for $n = 1, 2, \cdots 2^{N-1}$ and $\eta =\uparrow,\downarrow$, where $|\phi^{n,\eta}_{{\rm media},\uparrow}\rangle$ and $|\phi^{n,\eta}_{{\rm media},\downarrow}\rangle$ are the states of the spins from 2 to N and may not be normalized. Note that $\{E^n_{\rm media}\}$ and $\{\delta E'^{n,\xi}_{\rm tot}\}$ are of order β^0 because $||H'_{\rm media}||$ and $||H'_{\rm couple}||$ are of order β^0. Then, in the limit $\beta \to 0$, we have

$$\frac{e^{-\beta E'^{n,\uparrow}_{\rm tot}}}{Z_{\rm tot}} = \frac{e^{\beta h_{1\rm op} - \beta(E'^n_{\rm media} + \delta E'^{n,\uparrow}_{\rm tot})}}{Z_{\rm tot}} = O(\beta^0) \tag{3.84}$$

in the case $h_{1\rm op} > 0$.

Next, we calculate the elements $\{F_1, F_2\}$ and $\{p_{\uparrow\downarrow}p_{\downarrow\uparrow}, p_{\uparrow\uparrow}p_{\downarrow\downarrow}\}$. The calculation in Appendix (B.4) gives the state $|\phi_{\rm tot}^{n_0,\uparrow}\rangle$ in the form

$$|\phi_{\rm tot}^{n_0,\uparrow}\rangle = |\uparrow_1\rangle\otimes|\phi_{\rm media}^{n_0}\rangle + \frac{J}{-2h_{\rm 1op}}|\downarrow_1\rangle\otimes\Big(\gamma s'_{n_0}|\downarrow_2\rangle|\tilde{\phi}_{n_0}^{\uparrow\uparrow}\rangle|\uparrow_N\rangle + \gamma t'_{n_0}|\downarrow_2\rangle|\tilde{\phi}_{n_0}^{\uparrow\downarrow}\rangle|\downarrow_N\rangle$$
$$+ u'_{n_0}|\uparrow_2\rangle|\tilde{\phi}_{n_0}^{\downarrow\uparrow}\rangle|\uparrow_N\rangle + w'_{n_0}|\uparrow_2\rangle|\tilde{\phi}_{n_0}^{\downarrow\downarrow}\rangle|\downarrow_N\rangle + O(\beta^{\kappa_1})\Big). \tag{3.85}$$

Then, we can calculate the contribution of $|\phi_{\rm tot}^{n_0,\uparrow}\rangle$ to the elements $\{F_1, F_2\}$ and $\{p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$; we denote them as $\{F_1^{n_0,\uparrow}, F_2^{n_0,\uparrow}\}$ and $\{p_{\uparrow\uparrow}^{n_0,\uparrow}, p_{\uparrow\downarrow}^{n_0,\uparrow}, p_{\downarrow\uparrow}^{n_0,\uparrow}, p_{\downarrow\downarrow}^{n_0,\uparrow}\}$. First, the elements $\{F_1^{n_0,\uparrow}, F_2^{n_0,\uparrow}\}$ are given by

$$F_1^{n_0,\uparrow} = \frac{J}{-2h_{\rm 1op}}\Big(\gamma s'_{n_0}w'_{n_0}\langle\phi_{n_0}^{\downarrow\downarrow}|\phi_{n_0}^{\uparrow\uparrow}\rangle + u'_{n_0}t'_{n_0}\langle\phi_{n_0}^{\downarrow\uparrow}|\phi_{n_0}^{\uparrow\downarrow}\rangle + O(\beta^{\kappa_1})\Big) \tag{3.86}$$

and

$$F_2^{n_0,\uparrow} = \frac{J}{-2h_{\rm 1op}}\Big(\gamma t'_{n_0}u'_{n_0}\langle\phi_{n_0}^{\downarrow\uparrow}|\phi_{n_0}^{\uparrow\downarrow}\rangle + w'_{n_0}s'_{n_0}\langle\phi_{n_0}^{\uparrow\uparrow}|\phi_{n_0}^{\downarrow\downarrow}\rangle + O(\beta^{\kappa_1})\Big). \tag{3.87}$$

Second, the elements $\{p_{\uparrow\uparrow}^{n_0,\uparrow}, p_{\uparrow\downarrow}^{n_0,\uparrow}, p_{\downarrow\uparrow}^{n_0,\uparrow}, p_{\downarrow\downarrow}^{n_0,\uparrow}\}$ are given by

$$\begin{aligned}
p_{\uparrow\uparrow}^{n_0,\uparrow} &= s'^2_{n_0} + u'^2_{n_0},\\
p_{\uparrow\downarrow}^{n_0,\uparrow} &= t'^2_{n_0} + w'^2_{n_0},\\
p_{\downarrow\uparrow}^{n_0,\uparrow} &= \frac{J^2}{4h_{\rm 1op}^2}\big(\gamma^2 s'^2_{n_0} + u'^2_{n_0} + O(\beta^{\kappa_1})\big),\\
p_{\downarrow\downarrow}^{n_0,\uparrow} &= \frac{J^2}{4h_{\rm 1op}^2}\big(\gamma^2 t'^2_{n_0} + w'^2_{n_0} + O(\beta^{\kappa_1})\big).
\end{aligned} \tag{3.88}$$

Then, we sum $\{F_1^{n,\uparrow}, F_2^{n,\uparrow}\}$ and $\{p_{\uparrow\uparrow}^{n,\uparrow}, p_{\uparrow\downarrow}^{n,\uparrow}, p_{\downarrow\uparrow}^{n,\uparrow}, p_{\downarrow\downarrow}^{n,\uparrow}\}$ with the Boltzmann weight $e^{-\beta E'^{n,\uparrow}_{\rm tot}}$ over the label n. First, we calculate the summation of $\{F_1^{n,\uparrow}, F_2^{n,\uparrow}\}$. From the same discussion as in Eqs. (3.66) and (3.67) in the case (b), we have

$$\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n}_{\rm media}} t'_n u'_n \langle\tilde{\phi}_n^{\uparrow\downarrow}|\tilde{\phi}_n^{\uparrow\downarrow}\rangle = \sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n}_{\rm media}} t'_n u'_n \langle\tilde{\phi}_n^{\downarrow\uparrow}|\tilde{\phi}_n^{\downarrow\uparrow}\rangle = O(\beta^{\alpha'}) \tag{3.89}$$

and

$$\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n}_{\rm media}} s'_n w'_n \langle\tilde{\phi}_n^{\uparrow\uparrow}|\tilde{\phi}_n^{\downarrow\downarrow}\rangle = \sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n}_{\rm media}} s'_n w'_n \langle\tilde{\phi}_n^{\downarrow\downarrow}|\tilde{\phi}_n^{\uparrow\uparrow}\rangle = O(\beta^{\alpha'}), \tag{3.90}$$

where $||H'_{\rm media}||$ is of order β^0, and the exponent α' is defined as follows:

$$\begin{aligned}
&\alpha' = \min(\alpha'_1, \alpha'_2),\\
&\langle\sigma_2^x\sigma_N^x\rangle = O(\beta^{\alpha'_1}), \quad \langle\sigma_2^y\sigma_N^y\rangle = O(\beta^{\alpha'_2}),
\end{aligned} \tag{3.91}$$

where $\alpha'_1 \geq N-2$ and $\alpha'_2 \geq N-2$. Since we are considering the case $N \geq 4$, we have $\alpha'_1 \geq 2$ and $\alpha'_2 \geq 2$. Moreover, because $\{\delta E'^{n,\eta}_{\rm tot}\}$ in Eq. (3.83) are of order β^0, we have

$$e^{-\beta E'^{n,\eta}_{\rm tot}} = e^{-\beta(E_1^\eta + E'^{n}_{\rm media})}\Big(1 - \beta\delta E'^{n,\eta}_{\rm tot} + O(\beta^2)\Big), \tag{3.92}$$

for $n = 1, 2, \cdots, 2^{N-1}$ and $\eta =\uparrow, \downarrow$. Then, we obtain from Eqs. (3.86), (3.87), and (3.89)–(3.92),

$$\begin{aligned}
&\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} F_1^{n_0,\uparrow} \\
&= \sum_{n=1}^{2^{N-1}} e^{-\beta(E_1^\eta + E'^n_{\rm media})} F_1^{n_0,\uparrow} + \sum_{n=1}^{2^{N-1}} e^{-\beta(E_1^\eta + E'^n_{\rm media})} \Big(1 - \beta\delta E'^{n,\eta}_{\rm tot} + O(\beta^2)\Big) F_1^{n_0,\uparrow} \\
&= e^{-\beta E_1^\uparrow} \frac{J}{-2h_{\rm 1op}} \Big(O(\beta^{\alpha'}) + O(\beta^{\kappa_1})\Big) + e^{-\beta E_1^\uparrow} \frac{J}{-2h_{\rm 1op}} O(\beta) = e^{-\beta E_1^\uparrow} \times O(\beta^{\kappa_1+1}) \qquad (3.93)
\end{aligned}$$

and

$$\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} F_2^{n_0,\uparrow} = e^{-\beta E_1^\uparrow} \frac{J}{-2h_{\rm 1op}} \Big(O(\beta^{\alpha'}) + O(\beta^{\kappa_1}) + O(\beta)\Big) = e^{-\beta E_1^\uparrow} \times O(\beta^{\kappa_1+1}). \qquad (3.94)$$

We similarly calculate the contributions of the other states $\{|\psi^{n,\downarrow}_{\rm tot}\rangle\}$; then, we finally arrive at (3.76),

$$\begin{aligned}
F_1 &= \frac{1}{Z_{\rm tot}} \sum_{\eta=\uparrow,\downarrow} e^{-\beta E_1^\eta} \times O(\beta^{\kappa_1+1}) = O(\beta^{\kappa_1+1}), \\
F_2 &= \frac{1}{Z_{\rm tot}} \sum_{\eta=\uparrow,\downarrow} e^{-\beta E_1^\eta} \times O(\beta^{\kappa_1+1}) = O(\beta^{\kappa_1+1}), \qquad (3.95)
\end{aligned}$$

where we utilized the inequality

$$\frac{e^{-\beta E_1^\eta}}{Z_{\rm tot}} < 1 \qquad (3.96)$$

for $\eta =\uparrow, \downarrow$.

Second, we calculate the summation of $\{p^{n,\uparrow}_{\uparrow\uparrow}, p^{n,\uparrow}_{\uparrow\downarrow}, p^{n,\uparrow}_{\downarrow\uparrow}, p^{n,\uparrow}_{\downarrow\downarrow}\}$. From Eq. (3.88), we obtain

$$\begin{aligned}
\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\uparrow\uparrow} &= e^{-\beta E_1^\uparrow} \times O(\beta^0), \\
\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\uparrow\downarrow} &= e^{-\beta E_1^\uparrow} \times O(\beta^0), \\
\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\downarrow\uparrow} &= e^{-\beta E_1^\uparrow} \times O(\beta^{2\kappa_1}), \\
\sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\downarrow\downarrow} &= e^{-\beta E_1^\uparrow} \times O(\beta^{2\kappa_1}), \qquad (3.97)
\end{aligned}$$

because $s'^2_n + u'^2_n$, $t'^2_n + w'^2_n$, $\gamma^2 s'^2_n + u'^2_n$ and $\gamma^2 t'^2_n + w'^2_n$ cannot vanish for all n. From Eq. (3.84),

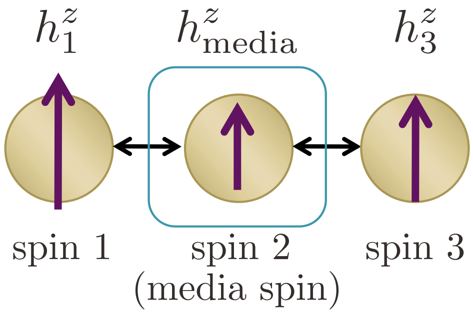


Figure 3.2: Schematic picture of the three-spin chain. We define the spins 1 and 3 as the focused spins and the spin 2 as the media spin. The spins 1 and 3 are symmetric with each other and they indirectly interact to each other through the spin 2.

we have

$$
\begin{aligned}
p_{\uparrow\uparrow} &\geq \frac{1}{Z_{\rm tot}} \sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\uparrow\uparrow} = O(\beta^0), \\
p_{\uparrow\downarrow} &\geq \frac{1}{Z_{\rm tot}} \sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\uparrow\downarrow} = O(\beta^0), \\
p_{\downarrow\uparrow} &\geq \frac{1}{Z_{\rm tot}} \sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\downarrow\uparrow} = O(\beta^{2\kappa_1}), \\
p_{\downarrow\downarrow} &\geq \frac{1}{Z_{\rm tot}} \sum_{n=1}^{2^{N-1}} e^{-\beta E'^{n,\uparrow}_{\rm tot}} p^{n,\uparrow}_{\downarrow\downarrow} = O(\beta^{2\kappa_1}).
\end{aligned} \tag{3.98}
$$

We thereby obtain (3.76). We thus obtain (3.76) and (3.77) and hence the inequality (3.28) is satisfied below a certain value of β in the case (c).

Thus, we prove the inequality (3.28) in the cases (a), (b) and (c). This completes the proof of Theorem 2.

3.4 Entanglement maximization in three-spin chains

In the present section, we discuss the maximization problem in three-spin chains. A significant result of the present section is that the maximizing local fields are asymmetric to each other in a particular parameter region. This effect is nontrivial; the positions of the spins 1 and 3 are geometrically symmetric to each other and the entanglement is invariant with respect to the exchange of the two spins. Nevertheless, we obtain $|h_{1\rm op} \neq |h_{3\rm op}|$ in a region. We show that the asymmetry can be mainly attributed to the behavior of the magnon. This effect indicates that the purity of the focused spins is not the only criterion for the enhancement of the bipartite entanglement. This is different from the case of the entanglement maximization in two-spin systems, which we considered in the previous chapter.

3.4.1 Numerical results

The Hamiltonian which we consider in the present section is

$$H_{\rm tot} = \sum_{i=1}^{2}(J^x\sigma_i^x\sigma_{i+1}^x + J^y\sigma_i^y\sigma_{i+1}^y + J^z\sigma_i^z\sigma_{i+1}^z) + h_1^z\sigma_1^z + h_{\rm media}^z\sigma_2^z + h_3^z\sigma_3^z, \tag{3.99}$$

where the spins 1 and 3 are the focused spins and the spin 2 is the media spin (Fig. 3.2). We assume $J^x \geq J^y \geq J^z$. We solve the entanglement maximization problem about the spins 1 and 3 by fixing the temperature T and the field $h_{\rm media}^z$ on the spin 2. In order to solve this maximization problem numerically, we use the random search method and the Newton method together. According to Theorem 2, if the two spins were separated by two spins, there would always be a critical temperature above which the maximized entanglement is exactly equal to zero. In the present case, the focused two spins are separated by only one spin and therefore Theorem 2 does not apply; the elements $|F_1|$, $\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}$, $|F_2|$ and $\sqrt{p_{\uparrow\downarrow}p_{\downarrow\uparrow}}$ are shown to be of the same order in the same way as in the proof of Theorem 2. Therefore, it generally depends on the interaction Hamiltonian and the positions of the focused spins whether the critical temperature exists or not. As for the Hamiltonian (3.99), we can prove that the entanglement between the spins 1 and 3 can exist at any temperatures by letting $h_1^z = h_3^z \to \infty$ (Appendix B.5).

In Fig. 3.3, we show the numerical results about the entanglement maximization. The main feature is that in a parameter region the maximizing local fields $h_{\rm 1op}$ and $h_{\rm 3op}$ are asymmetric to each other, namely $|h_{\rm 1op}| \neq |h_{\rm 3op}|$. In this region, the asymmetry must be essential to the enhancement of the entanglement. The asymmetry appears continuously (solid line in Fig. 3.3) or discontinuously (broken line in Fig. 3.3). Note that in these case there is no critical temperature above which the maximized entanglement would be zero.

3.4.2 Analytical argument

Here, we argue the origin of the asymmetry for the XX model; the phase diagram of the other models are not different from the XX model qualitatively. The main reason of the asymmetry is the strong dependence of the indirect interaction on the local fields. Then, we focus on the magnons which mediate the indirect interaction and discuss the effect of the local fields on the magnons. We show that the following three points affect the asymmetry:

1. The Boltzmann weights of the states with the magnons.

2. Localization of the magnons.

3. Suppression of the off-diagonal elements $\{F_1, F_2\}$ due to mixing of different magnon states.

We define a magnon as a spin flip; a down spin in the background of up spins or an up spin in the background of down spins. For example, the magnon number is two for both of the states $|\uparrow\uparrow\uparrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\downarrow\downarrow\downarrow\rangle$. In the XX three-spin chain, the number of the magnons can be either zero or one according to the above definition. The magnon does not exist in the states $|\uparrow_1\uparrow_2\uparrow_3\rangle$ and $|\downarrow_1\downarrow_2\downarrow_3\rangle$, nor the entanglement. This suggests that the magnons mediate the indirect interaction and is essential to the entanglement between the focused spins. Even if the magnons exist, however, the entanglement can be very small when the magnon is localized in one site. It is also possible that the off-diagonal elements $\{F_1, F_2\}$ in the density matrix are

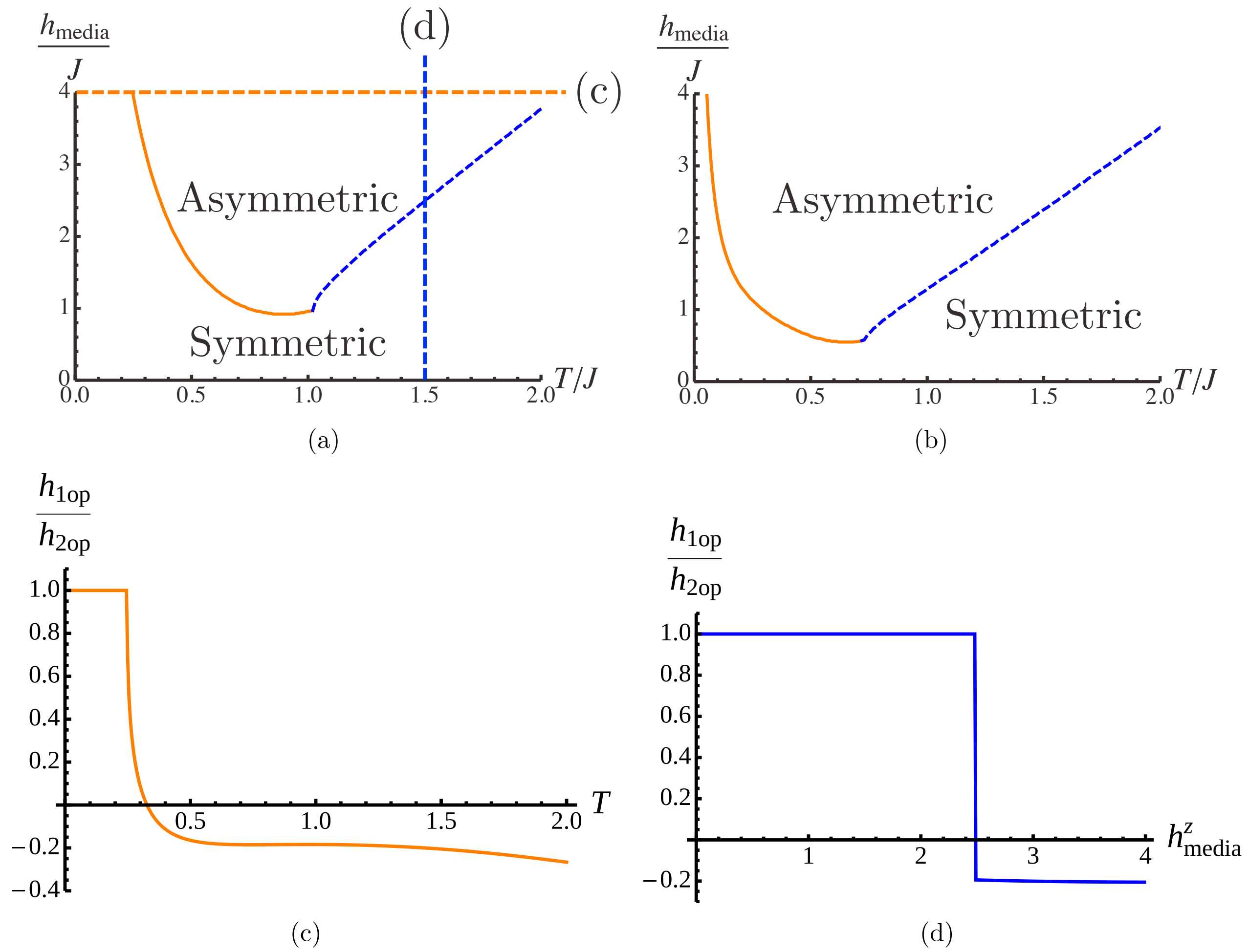


Figure 3.3: The parameter ranges in which the asymmetry appears (a) in the case of $J^x = J^y = 1$ and (b) in the case of $J^x = 1, J^y = 0.5$. In the asymmetric phase, the optimizing fields $h_{1\mathrm{op}}$ and $h_{3\mathrm{op}}$ are asymmetric to each other as $|h_{1\mathrm{op}}| \neq |h_{3\mathrm{op}}|$, while they satisfy $|h_{1\mathrm{op}}| = |h_{3\mathrm{op}}|$ in the symmetric phase. On the solid line, the asymmetry appears continuously, while on the broken line the asymmetry appears discontinuously. The ratio $h_{1\mathrm{op}}/h_{3\mathrm{op}}$ is shown for (c) $h^z_{\mathrm{media}} = 4$ and (d) $T = 1.5$ on the chained lines of (a).

suppressed by the mixture of the two kinds of magnon states. As shown in Eqs. (3.64) and (3.65), these elements are related to the correlation between the focused spins;

$$\begin{aligned} F_1 &= \frac{\langle \sigma_2^x \sigma_{N-1}^x + \sigma_2^y \sigma_{N-1}^y \rangle_0}{4}, \\ F_2 &= \frac{\langle \sigma_2^x \sigma_{N-1}^x - \sigma_2^y \sigma_{N-1}^y \rangle_0}{4}. \end{aligned} \tag{3.100}$$

Let us show an example; each of the two states

$$\frac{1}{\sqrt{2}}(|\uparrow_1\uparrow_2\downarrow_3\rangle + |\downarrow_1\uparrow_2\uparrow_3\rangle),\ \frac{1}{\sqrt{2}}(|\uparrow_1\uparrow_2\downarrow_3\rangle - |\downarrow_1\uparrow_2\uparrow_3\rangle), \tag{3.101}$$

has one magnon and its entanglement is maximum. However, the correlations which their magnons mediate are opposite to each other; the first state has $\langle \sigma_1^x \sigma_3^x \rangle = \langle \sigma_1^y \sigma_3^y \rangle = 1$, whereas the second one has $\langle \sigma_1^x \sigma_3^x \rangle = \langle \sigma_1^y \sigma_3^y \rangle = -1$. Then, the off-diagonal elements $\{F_1, F_2\}$ are also opposite to each other. Therefore, the entanglement is enhanced if we suppress the mixedness of the one state of (3.101).

Let us analytically show how the above three points affect the asymmetry. First, we consider the case of $T \simeq 0$ and $h^z_{\mathrm{media}} \gg J$, that is, the upper left corner of the phase diagram in Fig. 3.3. In this case, we have $h_{1\mathrm{op}} = h_{3\mathrm{op}} > 0$ in the 'symmetric' phase and $h_{1\mathrm{op}}, h_{3\mathrm{op}} > 0$ but $h_{1\mathrm{op}} \neq h_{3\mathrm{op}}$ in the 'asymmetric' phase. Let us consider the interaction J and the local fields $h_{1\mathrm{op}}, h_{3\mathrm{op}}$ on the focused spins as perturbation. Then, the four unperturbed ground states $|\uparrow_1\uparrow_2\uparrow_3\rangle$, $|\uparrow_1\uparrow_2\downarrow_3\rangle$, $|\downarrow_1\uparrow_2\uparrow_3\rangle$ and $|\downarrow_1\uparrow_2\downarrow_3\rangle$ are degenerate; we define these four states as $\{\psi_i^{(0)}\}$. Because we assume $h^z_{\mathrm{media}} \gg J$, we consider the mixing of only these four states. The first-order contribution of the excited states $|\uparrow_1\downarrow_2\uparrow_3\rangle$, $|\uparrow_1\downarrow_2\downarrow_3\rangle$, $|\downarrow_1\downarrow_2\uparrow_3\rangle$ and $|\downarrow_1\downarrow_2\downarrow_3\rangle$ to the above four unperturbed ground states are given in the forms

$$\begin{aligned} &\psi_1^{(1)} = |\uparrow_1\uparrow_2\uparrow_3\rangle,\ \psi_2^{(1)} = |\uparrow_1\uparrow_2\downarrow_3\rangle + \frac{J}{-2h^z_{\mathrm{media}}}|\uparrow_1\downarrow_2\uparrow_3\rangle, \\ &\psi_3^{(1)} = |\downarrow_1\uparrow_2\uparrow_3\rangle + \frac{J}{-2h^z_{\mathrm{media}}}|\uparrow_1\downarrow_2\uparrow_3\rangle, \text{and } \psi_4^{(1)} = |\downarrow_1\uparrow_2\downarrow_3\rangle + \frac{J}{-2h^z_{\mathrm{media}}}|\uparrow_1\downarrow_2\downarrow_3\rangle + \frac{J}{-2h^z_{\mathrm{media}}}|\downarrow_1\downarrow_2\uparrow_3\rangle. \end{aligned} \tag{3.102}$$

Then, we calculate the following matrix:

$$\langle \psi_i^{(0)} | \delta H | \psi_j^{(1)} \rangle \tag{3.103}$$

for $i, j = 1, 2, 3, 4$, where δH is the perturbation Hamiltonian, namely the interaction J and the local fields $h_{1\mathrm{op}}, h_{3\mathrm{op}}$. The matrix (3.103) is given by

$$\begin{pmatrix} -h_{1\mathrm{op}} - h_{3\mathrm{op}} & 0 & 0 & 0 \\ 0 & -h_{1\mathrm{op}} + h_{3\mathrm{op}} - J^2/(2h^z_{\mathrm{media}}) & -J^2/(2h^z_{\mathrm{media}}) & 0 \\ 0 & -J^2/(2h^z_{\mathrm{media}}) & h_{1\mathrm{op}} - h_{3\mathrm{op}} - J^2/(2h^z_{\mathrm{media}}) & 0 \\ 0 & 0 & 0 & h_{1\mathrm{op}} + h_{3\mathrm{op}} - J^2/h^z_{\mathrm{media}} \end{pmatrix}. \tag{3.104}$$

Diagonalizing this matrix, we obtain the eigenstates in the first-order perturbation as

$$
\begin{aligned}
|\psi_1\rangle &= |\uparrow_1\uparrow_2\uparrow_3\rangle,\ \epsilon_1 = -h_{1\text{op}} - h_{3\text{op}} - h^z_{\text{media}}, \\
|\psi_2\rangle &= \frac{1}{\sqrt{2}}\sqrt{1+\frac{h_{1\text{op}}-h_{3\text{op}}}{\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}}\left(|\uparrow_1\uparrow_2\downarrow_3\rangle+\frac{\delta}{J}|\uparrow_1\downarrow_2\uparrow_3\rangle\right) \\
&\quad+\frac{1}{\sqrt{2}}\sqrt{1-\frac{h_{1\text{op}}-h_{3\text{op}}}{\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}}\left(|\downarrow_1\uparrow_2\uparrow_3\rangle+\frac{\delta}{J}|\uparrow_1\downarrow_2\uparrow_3\rangle\right), \\
\epsilon_2 &= -\delta-\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}-h^z_{\text{media}}, \\
|\psi_3\rangle &= \frac{1}{\sqrt{2}}\sqrt{1-\frac{h_{1\text{op}}-h_{3\text{op}}}{\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}}\left(|\uparrow_1\uparrow_2\downarrow_3\rangle+\frac{\delta}{J}|\uparrow_1\downarrow_2\uparrow_3\rangle\right) \\
&\quad-\frac{1}{\sqrt{2}}\sqrt{1+\frac{h_{1\text{op}}-h_{3\text{op}}}{\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}}\left(|\downarrow_1\uparrow_2\uparrow_3\rangle+\frac{\delta}{J}|\uparrow_1\downarrow_2\uparrow_3\rangle\right), \\
\epsilon_3 &= -\delta+\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}-h^z_{\text{media}}, \\
|\psi_4\rangle &= |\downarrow_1\uparrow_2\downarrow_3\rangle+\frac{\delta}{J}|\downarrow_1\downarrow_2\uparrow_3\rangle+\frac{\delta}{J}|\uparrow_1\downarrow_2\downarrow_3\rangle,\ \epsilon_4 = h_{1\text{op}}+h_{3\text{op}}-h^z_{\text{media}}+2\delta,
\end{aligned} \tag{3.105}
$$

where

$$
\delta \equiv \frac{J^2}{-2h^z_{\text{media}}}. \tag{3.106}
$$

We thereby calculate the elements F_1, $p_{\uparrow\uparrow}$ and $p_{\downarrow\downarrow}$ of the density matrix ρ_{12}, which contribute to the concurrence. First, the leading terms of $p_{\uparrow\uparrow}$ and $p_{\downarrow\downarrow}$ are obtained as follows:

$$
p_{\uparrow\uparrow} = \frac{e^{-\beta\epsilon_1}}{Z_{\text{tot}}} \text{ and } p_{\downarrow\downarrow} = \frac{e^{-\beta\epsilon_4}}{Z_{\text{tot}}}. \tag{3.107}
$$

Therefore, $\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}$ is equal to $e^{\beta h^z_{\text{media}}}$ and does not depend on $h_{1\text{op}}$ and $h_{3\text{op}}$. Then the entanglement depends on $h_{1\text{op}}$ and $h_{3\text{op}}$ mainly through F_1 and F_2 as is shown in (3.8).

We obtain the leading term of F_1 as follows:

$$
F_1 = \frac{e^{\beta(h^z_{\text{media}}+\delta)}}{2Z_{\text{tot}}}\left(1-\frac{(h_{1\text{op}}-h_{3\text{op}})^2}{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}\right)\left(e^{\beta\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}-e^{-\beta\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}\right). \tag{3.108}
$$

Let us see how F_1 depends on the asymmetry $h_{1\text{op}} - h_{3\text{op}}$. We consider the following three terms of F_1 separately:

$$
\begin{aligned}
&1-\frac{(h_{1\text{op}}-h_{3\text{op}})^2}{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2} = 2\frac{1}{\sqrt{2}}\sqrt{1+\frac{h_{1\text{op}}-h_{3\text{op}}}{\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}}\cdot\frac{1}{\sqrt{2}}\sqrt{1-\frac{h_{1\text{op}}-h_{3\text{op}}}{\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}}, \\
&e^{\beta\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}} \text{ and } e^{-\beta\sqrt{(h_{1\text{op}}-h_{3\text{op}})^2+\delta^2}}.
\end{aligned} \tag{3.109}
$$

We start from the point $h_{1\text{op}} = h_{3\text{op}}$ and gradually increase the asymmetry $h_{1\text{op}} - h_{3\text{op}}$ to see how the above three terms affect the entanglement through the element F_1. We argue that there are two effects competing with each other, which may yield the phase boundary in the upper left corner of the phase diagram in Fig. 3.3.

The two factors on the right-hand side of the first term in (3.109) appear as coefficients in the states $|\psi_2\rangle$ and $|\psi_3\rangle$ in (3.105). As the asymmetry $h_{1\mathrm{op}} - h_{3\mathrm{op}}$ is increased, the second factor decreases and therefore the states $|\psi_2\rangle$ and $|\psi_3\rangle$ become dominated by the states $|\uparrow_1\uparrow_2\downarrow_3\rangle$ and $|\downarrow_1\uparrow_2\uparrow_3\rangle$, respectively; the first factor decreases when the asymmetry is reversed to $h_{3\mathrm{op}} - h_{1\mathrm{op}}$. Then the magnon becomes localized on the site 1 or 3 as the asymmetry $h_{1\mathrm{op}} - h_{3\mathrm{op}}$ is increased. This is a negative effect on the entanglement; the entanglement would be suppressed if the magnon does not move around the system. Indeed, the element F_1 in (3.108) and hence the entanglement in (3.8) can decrease because of the decrease of the first term in (3.109).

The second and third terms in (3.109) are the Boltzmann weights of the states $|\psi_2\rangle$ and $|\psi_3\rangle$. As the asymmetry $h_{1\mathrm{op}} - h_{3\mathrm{op}}$ is increased, the second term increases while the third term decreases exponentially. The state $|\psi_2\rangle$ has positive transverse correlations $\langle\sigma_1^x\sigma_3^x\rangle$ and $\langle\sigma_1^y\sigma_3^y\rangle$, while the state $|\psi_3\rangle$ has negative ones. As has been shown in the example of (3.101), the increase and decrease of the respective Boltzmann weights enhance the transverse correlations. This is a positive effect on the entanglement; the transverse correlations can promote the off-diagonal elements $\{F_1, F_2\}$ in Eq. (3.100). Indeed, the element F_1 in (3.108) can increase because of the exponential increase and the decrease of the second and third terms in (3.109).

The above negative and positive effects of the asymmetry on F_1 and the entanglement compete with each other. In other words, in some cases the entanglement is maximized without the asymmetry, while in other cases it is maximized by introducing the asymmetry. This may be the reason of the phase boundary in the upper left corner of the phase diagram in Fig. 3.3.

Second, we consider the case $h_{\mathrm{media}} > T \gg 1$, that is, the phase of the asymmetry in the upper right area of the phase diagram in Fig. 3.3. We numerically obtained $h_{1\mathrm{op}} \gg J$, $h_{3\mathrm{op}} \ll -J$ and $|h_{1\mathrm{op}}| \neq |h_{3\mathrm{op}}|$ in this area. We show that the asymmetry indeed promotes the entanglement by starting from the point $h_{1\mathrm{op}} = -h_{3\mathrm{op}}$ and increasing the asymmetry $h_{1\mathrm{op}} + h_{3\mathrm{op}}$ gradually. We argue that the increase of the entanglement is due to delocalization of the magnon. In order to calculate the entanglement in the present case, we regard the interaction between the focused spins and the media spins as perturbation.

Because $|h_{1\mathrm{op}}| \gg 1$ and $|h_{3\mathrm{op}}| \gg 1$, we consider only the two states $|\uparrow_1\uparrow_2\downarrow_3\rangle$ and $|\uparrow_1\downarrow_2\downarrow_3\rangle$ as the unperturbed states. Then, these two states in the first-order perturbation are given by

$$|\psi_1\rangle = |\uparrow_1\uparrow_2\downarrow_3\rangle + \frac{J}{-2(|h_{3\mathrm{op}}| + h^z_{\mathrm{media}})}|\uparrow_1\downarrow_2\uparrow_3\rangle + \frac{J^2}{4(|h_{3\mathrm{op}}| + h^z_{\mathrm{media}})(h_{1\mathrm{op}} + |h_{3\mathrm{op}}|)}|\downarrow_1\uparrow_2\uparrow_3\rangle,$$
$$\epsilon_1 = -h_{1\mathrm{op}} - |h_{3\mathrm{op}}| - h^z_{\mathrm{media}} \tag{3.110}$$

and

$$|\psi_2\rangle = |\uparrow_1\downarrow_2\downarrow_3\rangle + \frac{J}{-2(h_{1\mathrm{op}} + h^z_{\mathrm{media}})}|\downarrow_1\uparrow_2\downarrow_3\rangle + \frac{J^2}{4(h_{1\mathrm{op}} + h^z_{\mathrm{media}})(h_{1\mathrm{op}} + |h_{3\mathrm{op}}|)}|\downarrow_1\downarrow_2\uparrow_3\rangle,$$
$$\epsilon_2 = -h_{1\mathrm{op}} - |h_{3\mathrm{op}}| + h^z_{\mathrm{media}}. \tag{3.111}$$

From these expressions, we can calculate $\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}$ as

$$\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}} = e^{\beta(h_{1\mathrm{op}}+|h_{3\mathrm{op}}|)}\frac{J^2}{4(|h_{3\mathrm{op}}| + h^z_{\mathrm{media}})(h_{1\mathrm{op}} + h^z_{\mathrm{media}})}. \tag{3.112}$$

Similarly, the element F_1 is given by

$$F_1 = e^{\beta(h_{1\mathrm{op}}+|h_{3\mathrm{op}}|)}\left(\frac{J^2}{4(|h_{3\mathrm{op}}| + h^z_{\mathrm{media}})(h_{1\mathrm{op}} + |h_{3\mathrm{op}}|)}e^{\beta h^z_{\mathrm{media}}} + \frac{J^2}{4(h_{1\mathrm{op}} + h^z_{\mathrm{media}})(h_{1\mathrm{op}} + |h_{3\mathrm{op}}|)}e^{-\beta h^z_{\mathrm{media}}}\right). \tag{3.113}$$

To clarify the effect of the asymmetry $h_{1\mathrm{op}} + h_{3\mathrm{op}}$, we put

$$h_{1\mathrm{op}} = h_0 + \delta h \text{ and } h_{3\mathrm{op}} = -h_0 + \delta h, \tag{3.114}$$

where $h_0 \gg \delta h > 0$. Then, Eqs. (3.112) and (3.113) are recast into

$$\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}} = e^{2\beta h_0} \frac{J^2}{4\left[(h^z_{\mathrm{media}} + h_0)^2 - \delta h^2\right]} \tag{3.115}$$

and

$$F_1 = e^{2\beta h_0} \left(\frac{J^2}{8h_0(h_0 + h^z_{\mathrm{media}} - \delta h)} e^{\beta h^z_{\mathrm{media}}} + \frac{J^2}{8h_0(h_0 + h^z_{\mathrm{media}} + \delta h)} e^{-\beta h^z_{\mathrm{media}}} \right). \tag{3.116}$$

The former $\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}$ does not depend on the symmetry δh up to the first order. On the other hand, the latter F_1 and hence the entanglement in (3.8) increase as δh is increased; the increase of the first term on the right-hand side of (3.116) excels the decrease of the second term because of the difference in the Boltzmann weights.

The increase of F_1 can be related to delocalization of the magnon. Since $|h_{1\mathrm{op}}| \gg 1$ and $|h_{N\mathrm{op}}| \gg 1$, the magnon is almost localized in the site 3 in $|\psi_1\rangle$ of (3.110) and in the site 1 in $|\psi_2\rangle$ of (3.111), but slightly delocalized owing to the perturbation terms. As we increase δh, the perturbation terms of $|\psi_1\rangle$ increase, but those of $|\psi_2\rangle$ decrease. Therefore, the magnon is delocalized more in $|\psi_1\rangle$, while it is localized more in $|\psi_2\rangle$. This corresponds to the increase of the first term and the decrease of the second term in (3.116). Because the Boltzmann weight of the state $|\psi_1\rangle$ is greater than that of $|\psi_2\rangle$, the magnon delocalization in $|\psi_1\rangle$ excels the magnon localization in $|\psi_2\rangle$. This corresponds to the increase of F_1.

3.5 Entanglement maximization in four-spin chains

In the present section, we consider the maximization problem in four-spin chains (Fig. 3.4). As has been proved in Theorem 2, the four-spin chain is the shortest one in which the end-to-end entanglement cannot be generated in the high-temperature limit. We mainly discuss the critical temperature $\tilde{T}_\mathrm{c}$ and its dependence on the interaction of the spins.

3.5.1 Numerical results

In the present section, we consider the XY spin chains given by the Hamiltonian

$$H_{\mathrm{tot}} = \sum_{i=1}^{3} (J^x \sigma^x_i \sigma^x_{i+1} + J^y \sigma^y_i \sigma^y_{i+1}) + h^z_1 \sigma^z_1 + h^z_4 \sigma^z_2 + h^z_{\mathrm{media}}(\sigma^z_2 + \sigma^z_3). \tag{3.117}$$

We solve the entanglement maximization problem about the focused spins 1 and 4 by fixing the temperature T and the field h^z_{media} on the media spins 2 and 3. In order to solve this maximization problem numerically, we used the random search method and the Newton method together. According to Theorem 2, there always exists a critical temperature above which the maximized entanglement is exactly equal to zero because the focused spins 1 and 4 are separated by two spins.

We show the phase diagram of the XX spin chain and the XY spin chain in Fig. 3.5. As in the case of the three spins, there is an 'asymmetric' phase where the maximizing local field

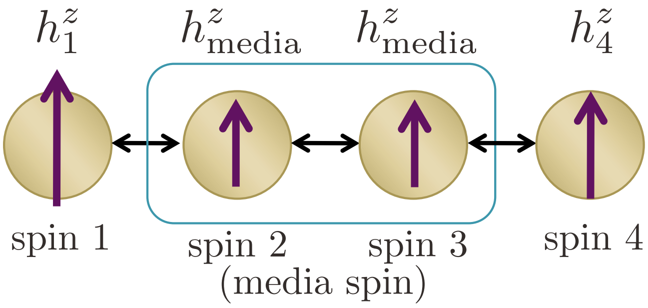


Figure 3.4: Schematic picture of four-spin chains. We define the spins 1 and 4 as the focused spins whereas the spins 2 and 3 as the media spins. The spins 1 and 4 indirectly interact with each other through the spins 2 and 3. In this system, the two focused spins are separated by two spins. Therefore, according to Theorem 2, there is a critical temperature above which the maximized entanglement is equal to zero.

$|h_{1\rm op}|$ is not equal to $|h_{4\rm op}|$. Moreover, the maximized entanglement is equal to zero in a region. The qualitatively different behavior of the critical temperature between the XX and the XY chains is due to the conservation of the angular momentum in the z direction, as we will argue in Section 3.5.2.

3.5.2 Difference between the XX and the XY model

We discuss the behavior of the critical temperature in the XX and XY chains. In the XX chain, the critical temperature increases as the media field $h^z_{\rm media}$ is increased, while in the XY chain it does not. This difference is attributed to the conservation of the angular momentum in the z direction. In the XX chain, we can suppress the mixture of the states with more than two magnons by choosing the fields as

$$\begin{aligned} h_1^z &= -h_0 \\ h_i^z &= h_0, \text{ for } i = 2, 3, \cdots N, \end{aligned} \tag{3.118}$$

and $h_0\beta \gg 1$. Then, the density matrix $e^{-\beta H_{\rm tot}}$ is almost equivalent to the ground state of $H_{\rm tot}$ and the mixture of the other states can be suppressed exponentially by increasing h_0. The ground state is given by the following form;

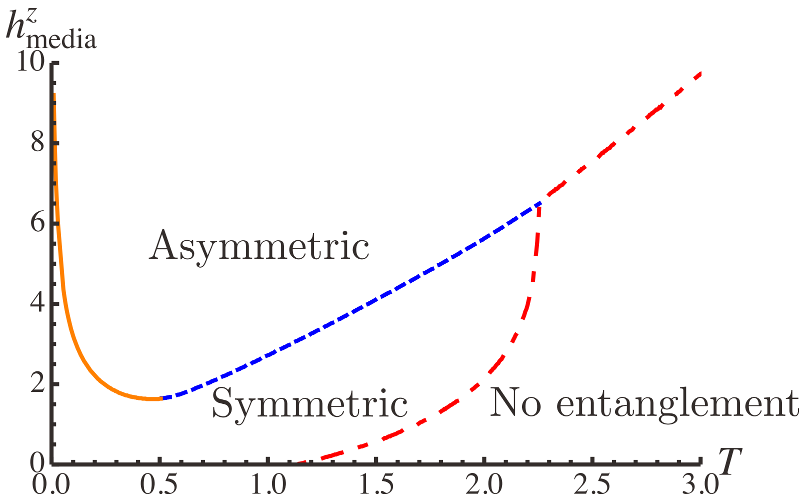
$$a_0|\downarrow_1\uparrow_2\cdots\uparrow_N\rangle + a_1|\uparrow_1\downarrow_2\cdots\uparrow_N\rangle + a_2|\uparrow_1\uparrow_2\downarrow_3\cdots\uparrow_N\rangle + \cdots + a_{N-1}|\uparrow_1\uparrow_2\cdots\downarrow_N\rangle, \tag{3.119}$$

where we can calculate a_k by the kth order perturbation to have

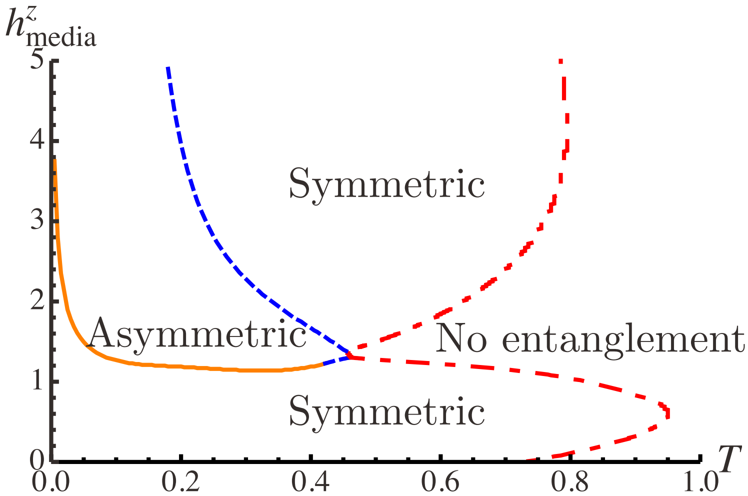
$$a_k = O\left(\frac{J^k}{h_0^k}\right) \tag{3.120}$$

for $k = 1, 2, \cdots N-1$ with the factor h_0^k coming from the energy denominator. In the ground state (3.119), the element $p_{\downarrow\downarrow}$ is equal to zero because there is no state with more than one down spins in (3.119). The entanglement between the spins 1 and N exists because $|F_1| \propto |a_{N-1}| \propto |J/h_0|^{N-1} > 0$. The mixture of the excited states generally destroys the entanglement, but is suppressed exponentially because of the Boltzmann weights. The entanglement between the spins 1 and N thereby survives.

On the other hand, in the XY chains, we cannot control the number of the magnons in the ground state by increasing the media fields. Therefore, $p_{\uparrow\uparrow}p_{\downarrow\downarrow}$ is not zero in the ground state, which invalidates the argument for the XX model. This may account for the fact that the critical temperature does not increase as the media fields $h^z_{\rm media}$ are increased.



(a)



(b)

Figure 3.5: The phase diagrams for the four-spin chain, (a) in the case of $J^x = J^y = 1$ and (b) in the case of $J^x = 1, J^y = 0.5$. In the 'asymmetric' phase, the maximizing fields $h_{1\rm op}$ and $h_{4\rm op}$ are asymmetric to each other as $|h_{1\rm op}| \neq |h_{4\rm op}|$, while they satisfy $|h_{1\rm op}| = |h_{4\rm op}|$ in the 'symmetric' phase. The asymmetry appears continuously on the solid line, while it appears discontinuously on the broken line. The maximized entanglement vanishes beyond the chained line.

3.5.3 Calculation of the critical temperature

Let us calculate the critical temperature analytically. It is generally difficult to solve the entanglement maximization analytically. At the critical temperature, however, we numerically confirmed that the maximizing local fields take the form $h_{1\mathrm{op}} = h_{4\mathrm{op}} \to \infty$ in some region of the 'symmetric' phases in Fig. 3.5. For the XX model, this happens everywhere in the 'symmetric' phase in Fig. 3.5 (a), whereas for the XY model it happens only in the lower one of the two 'symmetric' phases Fig. 3.5 (b). In such regions, we can derive the critical temperature analytically. In the 'asymmetric' phases in Fig. 3.5, on the other hand, the maximizing local fields do not have simple forms except in the case of the XX model, for which they approximately have the form of $h_{1\mathrm{op}} \to \infty$ and $h_{4\mathrm{op}} = -h^z_{\mathrm{media}}$. We consider the critical temperature in the 'asymmetric' phase in Fig. 3.5 (a) for the XX model in Appendix B.6.

Let us now derive the critical temperature in the limit $h_{1\mathrm{op}} = h_{4\mathrm{op}} \to \infty$. In order to calculate the entanglement in this limit, we regard the coupling Hamiltonian H_{couple} between the focused spins and the media spins as perturbation. Because of the condition $h_{1\mathrm{op}} = h_{4\mathrm{op}} \to \infty$, we consider only the following four unperturbed states:

$$|\uparrow_1\uparrow_4\rangle \otimes |\psi_1\rangle,\ |\uparrow_1\uparrow_4\rangle \otimes |\psi_2\rangle,\ |\uparrow_1\uparrow_4\rangle \otimes |\psi_3\rangle \text{ and } |\uparrow_1\uparrow_4\rangle \otimes |\psi_4\rangle, \tag{3.121}$$

where $\{|\psi_n\rangle\}_{n=1}^4$ are the eigenstates of the media spins. The states $\{|\psi_n\rangle\}_{n=1}^4$ and their corresponding eigenvalues are given by the solution of the bipartite XY spin chain:

$$\begin{aligned}
|\psi_1\rangle &= \frac{1}{\sqrt{2}}\big(|\uparrow_2\downarrow_3\rangle - |\downarrow_2\uparrow_3\rangle\big),\ \epsilon_1 = -J^x - J^y,\\
|\psi_2\rangle &= \frac{1}{\sqrt{2}}\big(|\uparrow_2\downarrow_3\rangle + |\downarrow_2\uparrow_3\rangle\big),\ \epsilon_2 = J^x + J^y,\\
|\psi_3\rangle &= a|\downarrow_3\downarrow_4\rangle - b|\uparrow_2\uparrow_3\rangle,\ \epsilon_3 = -\sqrt{4(h^z_{\mathrm{media}})^2 + (J^x - J^y)^2},\\
|\psi_4\rangle &= b|\downarrow_3\downarrow_4\rangle + a|\uparrow_2\uparrow_3\rangle,\ \epsilon_4 = \sqrt{4(h^z_{\mathrm{media}})^2 + (J^x - J^y)^2},
\end{aligned} \tag{3.122}$$

where we define a and b as

$$a = \frac{1}{\sqrt{2}}\sqrt{1 - \frac{2h^z_{\mathrm{media}}}{\sqrt{4(h^z_{\mathrm{media}})^2 + (J^x - J^y)^2}}},\ b = \frac{1}{\sqrt{2}}\sqrt{1 + \frac{2h^z_{\mathrm{media}}}{\sqrt{4(h^z_{\mathrm{media}})^2 + (J^x - J^y)^2}}}. \tag{3.123}$$

Next, we calculate the perturbation of $\{|\uparrow_1\uparrow_4\rangle \otimes |\psi_n\rangle\}_{n=1}^4$ as in the proof of Theorem 2. The perturbation contribution of $|\uparrow_1\uparrow_4\rangle \otimes |\psi_{n_0}\rangle$ to the leading term of the element F_1 is given by

$$\frac{J^2}{4h_{1\mathrm{op}}h_{4\mathrm{op}}}\Big(\gamma s_{n_0} w_{n_0} + u_{n_0} t_{n_0} + \gamma^2 t_{n_0} u_{n_0} + \gamma w_{n_0} s_{n_0}\Big), \tag{3.124}$$

where $\{s_n, t_n, u_n, w_n\}$ are defined as the coefficients of $\{|\psi_n\rangle\}_{n=1}^4$ in (3.122):

$$|\psi_n\rangle = s_n|\uparrow_2\uparrow_3\rangle + t_n|\uparrow_2\downarrow_3\rangle + u_n|\downarrow_2\uparrow_3\rangle + w_n|\downarrow_2\downarrow_3\rangle. \tag{3.125}$$

Equations (3.124) and (3.125) correspond to Eqs. (3.59) and (3.40), respectively. Similarly, the perturbation contribution of $|\uparrow_1\uparrow_4\rangle \otimes |\psi_{n_0}\rangle$ to the leading term of the element F_2 is given by

$$\frac{J^2}{4h_{1\mathrm{op}}h_{4\mathrm{op}}}\big(\gamma^2 w_{n_0} s_{n_0} + \gamma u_{n_0} t_{n_0} + \gamma t_{n_0} u_{n_0} + s_{n_0} w_{n_0}\big), \tag{3.126}$$

which corresponds to Eq. (3.60). On the other hand, Eqs. (3.56)–(3.58) give the perturbation contribution of $|\uparrow_1\uparrow_4\rangle \otimes |\psi_{n_0}\rangle$ to the leading terms of the elements $\{p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ as

$$\begin{aligned}
&\frac{J^2}{4h_{4\mathrm{op}}^2}\left(\gamma^2 s_{n_0}^2 + t_{n_0}^2 + \gamma^2 u_{n_0}^2 + w_{n_0}^2\right),\\
&\frac{J^2}{4h_{1\mathrm{op}}^2}\left(\gamma^2 s_{n_0}^2 + \gamma^2 t_{n_0}^2 + u_{n_0}^2 + w_{n_0}^2\right),\\
&\frac{J^4}{16h_{1\mathrm{op}}^2 h_{4\mathrm{op}}^2}\left(\gamma^4 s_{n_0}^2 + \gamma^2 t_{n_0}^2 + \gamma^2 u_{n_0}^2 + w_{n_0}^2\right),
\end{aligned} \tag{3.127}$$

respectively.

We have now all the ingredients for calculating the matrix elements $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ in the limit $h_{1\mathrm{op}} = h_{4\mathrm{op}} \to \infty$. We consider the mixture of the perturbed states of $|\uparrow_1\uparrow_4\rangle \otimes |\psi_1\rangle$, $|\uparrow_1\uparrow_4\rangle \otimes |\psi_2\rangle$, $|\uparrow_1\uparrow_4\rangle \otimes |\psi_3\rangle$ and $|\uparrow_1\uparrow_4\rangle \otimes |\psi_4\rangle$. These four states mix with the Boltzmann weights of $e^{-\beta(\epsilon_1+\delta\epsilon_1)}$, $e^{-\beta(\epsilon_2+\delta\epsilon_2)}$, $e^{-\beta(\epsilon_3+\delta\epsilon_3)}$ and $e^{-\beta(\epsilon_4+\delta\epsilon_4)}$, where we define the energy perturbations as $\{\delta\epsilon_i\}_{i=1}^4$. In the limit $h_{1\mathrm{op}} = h_{4\mathrm{op}} \to \infty$, we have to consider only the leading terms of $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$. Therefore, we ignore the energy perturbation $\{\delta\epsilon_i\}_{i=1}^4$. We thus arrive at the elements of the density matrix as

$$\begin{aligned}
Z_{\mathrm{tot}} F_1 &= \frac{J^2}{4h_0^2}\left[-4ab\gamma \sinh\beta|\epsilon_3| - (1+\gamma^2)\sinh\beta|\epsilon_1|\right],\\
Z_{\mathrm{tot}} F_2 &= \frac{J^2}{4h_0^2}\left[-2ab(1+\gamma^2)\sinh\beta|\epsilon_3| - 2\gamma\sinh\beta|\epsilon_1|\right],
\end{aligned} \tag{3.128}$$

and

$$\begin{aligned}
Z_{\mathrm{tot}} p_{\uparrow\uparrow} &= Z_{\mathrm{tot}} = 2\cosh\beta|\epsilon_1| + 2\cosh\beta|\epsilon_3|,\\
Z_{\mathrm{tot}} p_{\uparrow\downarrow} &= \frac{J^2}{4h_0^2}\Big[(\gamma^2 a^2 + b^2)e^{-\beta\epsilon_4} + (a^2 + \gamma^2 b^2)e^{-\beta\epsilon_3} + (\gamma^2+1)\cosh\beta|\epsilon_1|\Big],\\
Z_{\mathrm{tot}} p_{\downarrow\uparrow} &= \frac{J^2}{4h_0^2}\Big[(\gamma^2 a^2 + b^2)e^{-\beta\epsilon_4} + (a^2 + \gamma^2 b^2)e^{-\beta\epsilon_3} + (\gamma^2+1)\cosh\beta|\epsilon_1|\Big],\\
Z_{\mathrm{tot}} p_{\downarrow\downarrow} &= \frac{J^4}{16h_0^4}\Big[(\gamma^4 a^2 + b^2)e^{-\beta\epsilon_4} + (a^2 + \gamma^4 b^2)e^{-\beta\epsilon_3} + 2\gamma^2\cosh\beta|\epsilon_1|\Big],
\end{aligned} \tag{3.129}$$

where we defined $h_0 \equiv h_{1\mathrm{op}} = h_{4\mathrm{op}}$ with $h_0 \to \infty$.

By utilizing these parameters, we obtain a necessary and sufficient condition (3.9) for the existence of the entanglement in the form of the following two inequalities:

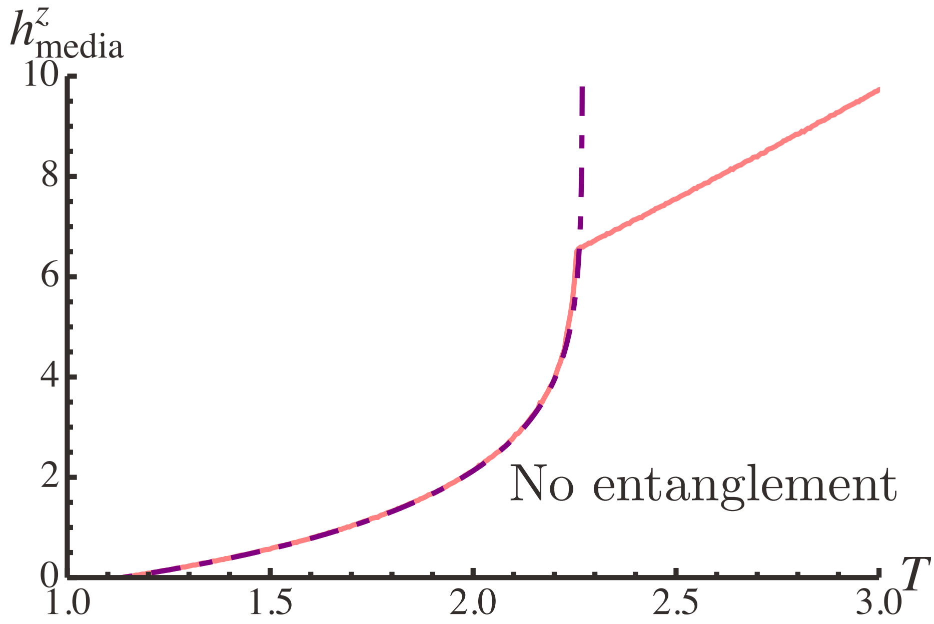
$$\begin{aligned}
Z_{\mathrm{tot}}^2(F_1^2 - p_{\uparrow\uparrow}p_{\downarrow\downarrow}) &= \left[4ab\gamma\sinh\beta|\epsilon_3| + (1+\gamma^2)\sinh\beta|\epsilon_1|\right]^2\\
&- Z_{\mathrm{tot}}\left[(\gamma^4 a^2 + b^2)e^{-\beta\epsilon_4} + (a^2 + \gamma^4 b^2)e^{-\beta\epsilon_3} + 2\gamma^2\cosh\beta|\epsilon_1|\right] > 0,
\end{aligned} \tag{3.130}$$

or

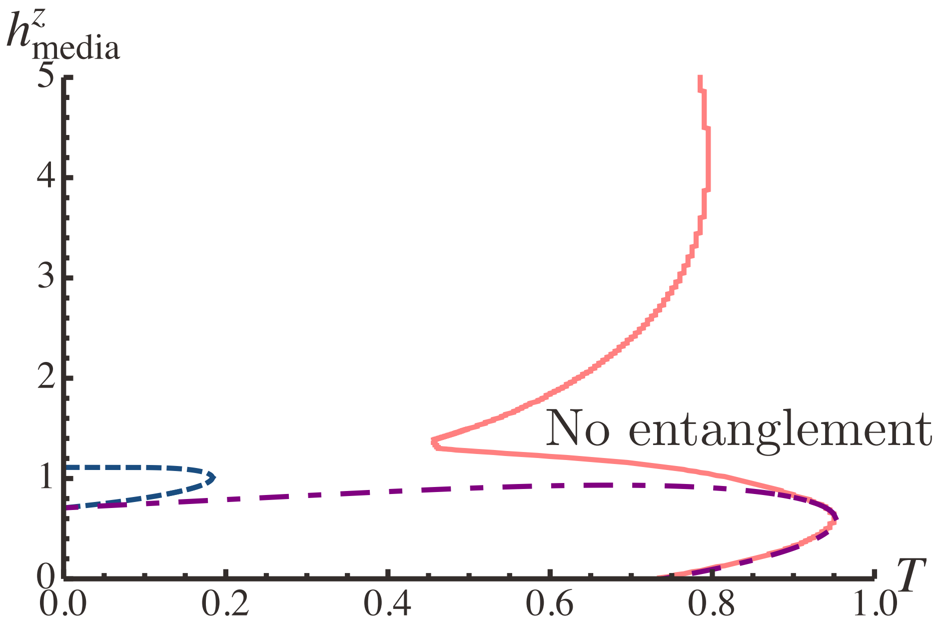
$$\begin{aligned}
Z_{\mathrm{tot}}^2(F_2^2 - p_{\uparrow\downarrow}p_{\downarrow\uparrow}) &= \left[2ab(1+\gamma^2)\sinh\beta|\epsilon_3| + 2\gamma\sinh\beta|\epsilon_1|\right]\\
&- \left[(\gamma^2 a^2 + b^2)e^{-\beta\epsilon_4} + (a^2 + \gamma^2 b^2)e^{-\beta\epsilon_3} + (\gamma^2+1)\cosh\beta|\epsilon_1|\right] > 0.
\end{aligned} \tag{3.131}$$

In the case of the XX model with $\gamma = 0$, in particular, the above condition becomes simpler; because we have $F_2 = 0$ for $\gamma = 0$, the inequality (3.131) is never satisfied. The condition (3.130), on the other hand, reduces to

$$\sinh^2(\beta|\epsilon 1|) - Z_{\mathrm{tot}}^{-\beta\epsilon_4} = \sinh^2(2\beta J) - Z_{\mathrm{tot}} e^{-2\beta h_{\mathrm{media}}^z} > 0, \tag{3.132}$$



(a)



(b)

Figure 3.6: Comparison of the phase boundary (a) in the case of $J^x = J^y = 1$ and (b) in the case of $J^x = 1, J^y = 0.5$. The critical temperature calculated according to Eq. (3.130) is indicated by the broken lines and the numerically calculated one by the solid lines, which are the same as in Fig. 3.5. The critical temperatures calculated by Eqs. (B.58) and (3.131) are also indicated by the chained line in (a) and the thin solid line in (b), respectively. In the XX spin chain, the approximation gives almost the same values as the numerical ones in the symmetric phase, while in the XY spin chain, the approximated values from (3.130) fit the numerical ones in the range of $0 \leq h^z_{\mathrm{media}} \leq 0.851$.

or

$$\frac{e^{2\beta h^z_{\text{media}}} \sinh^2(2\beta J)}{2\cosh(2\beta J) + 2\cosh(2\beta h^z_{\text{media}})} > 1. \tag{3.133}$$

In Fig. 3.6, we compare the critical temperatures calculated according to Eqs. (3.130) and (3.131) with the numerically calculated ones. We also plot in Fig. 3.6 (a) for the XX model, the critical temperature of the 'asymmetric' phase according to Appendix B.6. The approximations generally give precise estimates for the XX model, particularly for the 'symmetric' phase.

For the approximation for the 'asymmetric' phase, the consistency is fairly good in the range shown in Fig. 3.6 (a) as well as even in the range $h^z_{\text{media}} \gg J$ with a slight deviation. The deviation is because we used the approximation $h_{4\text{op}} = -h^z_{\text{media}}$, which is not correct in fact; for $h^z_{\text{media}} = 50$ and 100, for example, $h_{4\text{op}} = -59$ and $h_{4\text{op}} = -100$, respectively. The approximate estimates of the critical temperature are $\tilde{T}_{\text{c}} = 10.31$ and 18.05 for $h^z_{\text{media}} = 50$ and 100, respectively, while the numerical estimates are $\tilde{T}_{\text{c}} = 10.37$ and 18.20, respectively.

For the XY model, the approximate estimate shown in Fig. 3.6 (b) is due to (3.130) in the range bounded by the broken line and is due to (3.131) in the range bounded by the thin solid line. The former gives good estimates in the range of $0 \leq h^z_{\text{media}} \leq 0.851$.

3.6 Summary and conclusion

We have analytically and numerically studied the maximum value of the thermal entanglement between two spins which indirectly interact with each other. We showed two theorems on its general properties. First, if the indirect interaction satisfies the condition (3.10), the maximized entanglement is always equal to zero. We can say that in this case the interaction is not quantum but classical. This is one of the essential difference between the direct interaction and the indirect interaction; the direct interaction always generates non-zero entanglement. We also proved that the maximized entanglement is equal to zero above the critical temperature if the two spins are separated by two or more spins.

Secondly, in the three-spin chains and the four-spin chains, we showed properties of the maximized entanglement which is calculated numerically. In the three-spin chains, we showed that the maximizing local fields are not symmetric as $h_{1\text{op}} \neq h_{3\text{op}}$ (Fig. 3.3) in some parameter regions. We have explained the asymmetry qualitatively and quantitatively. We attributed the appearance of the asymmetry to the effects on the magnons, which mediate the indirect interaction. In other words, the local fields affect the effective interaction between the focused spins, while the direct interaction is determined independently of the external fields. In Eqs. (3.11)–(3.13), we also gave an example where the media fields also affect the interaction. In the four-spin chains, we demonstrated that the maximized entanglement vanishes above the certain critical temperature. Because of the difference in the symmetric properties between the XX and XY spin chains, the dependence of the critical temperatures on the media fields are qualitatively different between the two systems. We calculated the critical temperature analytically in some parameter regions.

In conclusion, we have clarified several properties of the entanglement which is generated from the indirect interaction. Our study has given several general limits for the entanglement generation. We have also shown some properties of the external fields which is closely related to the entanglement enhancement. However, there are many problems to be solved on the general relationship between the external fields and the entanglement enhancement. In particular, we have not shown the properties of the multipartite entanglement. It is obvious that the external fields also have effect on qualitative behaviors of the multipartite entanglement; the most famous one is the quantum phase transition of the transverse Ising model. In future, we plan to investigate the general properties of the enhancement of the multipartite entanglement by external fields.

Chapter 4

Summary and future works

In the present chapter, we conclude the present thesis and show the future works.

4.1 Conclusion

In the present thesis, we have given several answers to the following problems:

1. The general principles on the enhancement of (bipartite and multipartite) entanglement.
2. The general properties of the enhancement of the thermal entanglement at high temperatures.
3. The general properties of the long distance entanglement, as well as the method of generating it, if possible.

First, we have shown that the entanglement enhancement by external fields is attributed to the increase of the purity in the two-spin system. In the multipartite spin systems, the increase of the purity is not the only factor for the entanglement enhancement. In particular, in the three-spin chain, the enhancement of the entanglement can be brought about by the influence on the behavior of the magnons, which mediate the indirect interaction. So far, this results cannot be generalized to the other spin systems. However, we believe that the influence on a interaction by the external fields can play an essential role in the entanglement enhancement. Second, we have shown general properties of the maximized entanglement in the high-temperature limit. The protection of the thermal entanglement is impossible for any values of the local fields if the two spins are separated by two or more spins. Third, above a certain temperature, the long distance entanglement cannot be generated as a corollary of Theorem 2 in Chapter 3.

4.2 Future works

In the present thesis, we cannot give perfect answers to all the above problems. Therefore, we continue to study the above problems in more detail. First, we plan to research the generation of the long-distance entanglement by the local fields in the low-temperature limit. We have already obtained some results on it; in order to generate the long distance entanglement between the focused spins, we have to modulate the local fields on the focused spins and the neighboring spins (Fig. 4.1). Second, we also plan to investigate the general properties of the enhancement of the multipartite entanglement, which reflects the total quantumness of the system. The

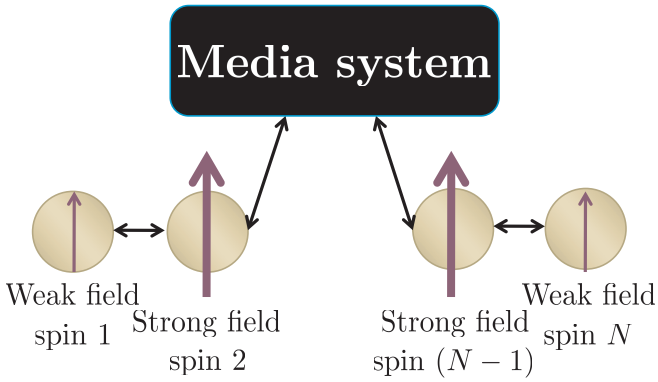


Figure 4.1: Schematic picture of the generation of the long distance entanglement. Four spins connect to the media system and we utilize two spins as one probe. We apply the local fields to these probe spins. One is very strong and the other is very weak. By choosing these local fields appropriately, we can generate the entanglement between the spins 1 and n.

research of the multipartite entanglement is essential to survey the relationship between the quantum phase transition and the enhancement of the quantumness. In the ground states, there are several useful measures of the multipartite entanglement [21, 23]. Therefore, we are going to work on the entanglement enhancement in the ground states.

Appendix A

Appendix for Chapter 2

A.1 Numerical comparison

In the present Appendix, we compare the asymptotes in Eqs. (2.11) and (2.13) with those in Eqs. (2.15) and (2.16) in the case of $\{J_x, J_y, J_z\} = \{1/3, 1/3, 1/3\}$. In this case, Eqs. (2.11), (2.13), (2.15) and (2.16), respectively, reduce to

$$e^{2h'_{(2.11)}} = \frac{12h'^2_{(2.11)}}{\beta}, \tag{A.1}$$

$$N_{(2.13)} = \beta\frac{1}{3h'_{(2.11)}} - 2e^{-2h'_{(2.11)}}, \tag{A.2}$$

$$h_{(2.15)} = \frac{\log 1/\beta}{2\beta}, \tag{A.3}$$

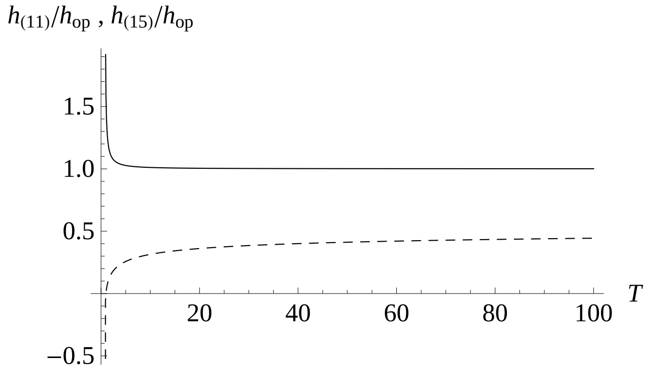
$$N_{(2.16)} = \beta\frac{2}{3\log 1/\beta}, \tag{A.4}$$

where the subscripts denote the equation number of the corresponding asymptotes. In Fig. A.1, we show the comparison of these asymptotes with the numerically rigorous estimates of h_{op} and N_{op} obtained from Eq. (2.95). We can see that the convergences of $h_{(2.15)}/h_{\text{op}}$ and $N_{(2.16)}/N_{\text{op}}$ are very slow, while the convergences of $h_{(2.11)}/h_{\text{op}}$ and $N_{(2.13)}/N_{\text{op}}$ are much faster. The convergence of $N(h_{(2.11)})/N_{\text{op}}$, where $N(h)$ is given in Eqs. (2.73) and (2.74), is even faster than that of $N_{(2.13)}/N_{\text{op}}$; at $T = 100$, the values of $N(h_{(2.11)})/N_{\text{op}}$ and $N_{(2.13)}/N_{\text{op}}$ are 0.999998 and 0.9994, respectively.

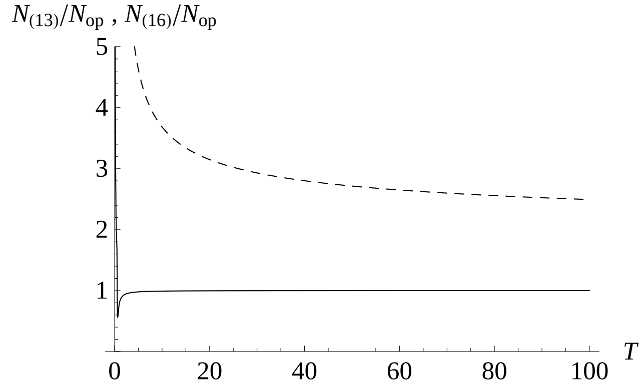
A.2 Lemma 2 in degenerate cases

In the proof of Lemma 2, we left out the cases of $\zeta = 0$ or $\zeta = \pm 1$ in Eq. (2.23). In the present Appendix, we prove that Lemma 2 still holds in these cases. First, the general form of Eq. (2.21) is given in the basis of $\{|\mu\rangle\}_{\mu=1}^4$ as follows:

$$Z\rho^{T_1} \xrightarrow{\beta\to 0} \begin{pmatrix} e^{2h'} & a_{12}f_{12}\beta & a_{31}f_{31}\beta & a_{32}f_{32}\beta \\ a_{21}f_{21}\beta & e^{2\zeta h'} & a_{41}f_{41}\beta & a_{42}f_{42}\beta \\ a_{13}f_{13}\beta & a_{14}f_{14}\beta & e^{-2\zeta h'} & a_{34}f_{34}\beta \\ a_{23}f_{23}\beta & a_{44}f_{42}\beta & a_{43}f_{43}\beta & e^{-2h'} \end{pmatrix}, \tag{A.5}$$



(a)



(b)

Figure A.1: The comparison between the asymptotes in Eqs. (2.11) and (2.13) and those in Eqs. (2.15) and (2.16). (a) for the ratios $h_{(2.11)}/h_{\rm op}$ (solid line) and $h_{(2.15)}/h_{\rm op}$ (dashed line), where $h_{(2.11)}$ and $h_{(2.15)}$ are derived from Eqs. (A.1) and (A.3), respectively, and $h_{\rm op}$ is the numerically rigorous value calculated from Eq. (2.95). At $T = 100$, the values of $h_{\rm op}^{(2.11)}/h_{\rm op}$ and $h_{\rm op}^{(2.15)}/h_{\rm op}$ are 1.0007 and 0.4438, respectively. (b) for the ratios $N_{(2.13)}/N_{\rm op}$ (solid line) and $N_{(2.16)}/N_{\rm op}$ (dashed line), where $N_{(2.13)}$ and $N_{(2.16)}$ are derived from Eqs. (A.2) and (A.4), respectively, and $N_{\rm op}$ is the numerically rigorous value calculated from Eq. (2.95). At $T = 100$, the values of $N_{(2.13)}/N_{\rm op}$ and $N_{(2.16)}/N_{\rm op}$ are 0.9994 and 2.494, respectively.

where $\{a_{ij}\}$ are constants of order 1 and $\{f_{\mu\nu}\}$ are defined in Eq. (2.22). Note that on the diagonal of Eq. (A.5), the second term of Eq. (2.21) is neglected in comparison to the first term. In the cases of $\zeta \neq 0$ and $\zeta \neq \pm 1$, Eq. (A.5) reduces to Eq. (2.23).

In the case of $\zeta = 0$, we have $\{E_1', E_2', E_3', E_4'\} = \{2h', 0, 0, -2h'\}$ and $f_{23} = f_{32} = e^{-E_2'} = 1$, and hence Eq. (A.5) reduces to

$$Z\rho^{T_1} \xrightarrow{\beta\to 0} \begin{pmatrix} e^{2h'} & a_{12}\frac{\beta e^{2h'}}{h'} & a_{31}\frac{\beta e^{2h'}}{h'} & a_{32}\beta \\ a_{21}\frac{\beta e^{2h'}}{h'} & 1 & a_{41}\frac{\beta e^{2h'}}{h'} & a_{42}\frac{\beta}{h'} \\ a_{13}\frac{\beta e^{2h'}}{h'} & a_{14}\frac{\beta e^{2h'}}{h'} & 1 & a_{34}\frac{\beta}{h'} \\ a_{23}\beta & a_{24}\frac{\beta}{h'} & a_{43}\frac{\beta}{h'} & e^{-2h'} \end{pmatrix}, \tag{A.6}$$

In this case, the product of the diagonal elements (PD) of $Z\rho^{T_1}$ is 1, whereas the maximum of the absolute values of the products including off-diagonal elements (POD) is of order $e^{4h'}\beta^2/h'^2$, which comes from the product $-e^{2h'} \times a_{41}\frac{\beta e^{2h'}}{h'} \times a_{14}\frac{\beta e^{2h'}}{h'} \times e^{-2h'}$. Therefore, it is necessary for $\det \rho^{T_1} < 0$ that the order of $e^{4h'}\beta^2/h'^2$ is greater or of order 1, which leads to

$$\beta h = h' > \frac{\log 1/\beta}{2} \tag{A.7}$$

as in Eq. (2.24). Thus, Lemma 2 is proved in the case of $\zeta = 0$.

The proofs for the cases of $\zeta = 1$ and $\zeta = -1$, or the cases of $\{h_1, h_2\} = \{2h, 0\}$ and $\{h_1, h_2\} = \{0, 2h\}$, are essentially the same. We here present the proof only for the case of $\zeta = 1$. In this case, we have $\{E_1', E_2', E_3', E_4'\} = \{2h', 2h', -2h', -2h'\}$, $f_{12} = f_{21} = e^{2h'}$ and $f_{34} = f_{43} = e^{-2h'}$, and hence Eq. (A.5) reduces to

$$Z\rho^{T_1} \xrightarrow{\beta\to 0} \begin{pmatrix} e^{2h'} & a_{12}\beta e^{2h'} & a_{31}\frac{\beta e^{2h'}}{h'} & a_{32}\frac{\beta e^{2h'}}{h'} \\ a_{21}\beta e^{2h'} & e^{2h'} & a_{41}\frac{\beta e^{2h'}}{h'} & a_{42}\frac{\beta e^{2h'}}{h'} \\ a_{13}\frac{\beta e^{2h'}}{h'} & a_{14}\frac{\beta e^{2h'}}{h'} & e^{-2h'} & a_{34}\beta e^{-2h'} \\ a_{23}\frac{\beta e^{2h'}}{h'} & a_{24}\frac{\beta e^{2h'}}{h'} & a_{43}\beta e^{-2h'} & e^{-2h'} \end{pmatrix}, \tag{A.8}$$

The PD of $Z\rho^{T_1}$ is 1, whereas the maximum of the absolute values of the PODs is of order $e^{4h'}\beta^2/h'^2$ or of order $e^{8h'}\beta^4/h'^4$, which come from $-e^{2h'} \times a_{41}\frac{\beta e^{2h'}}{h'} \times a_{14}\frac{\beta e^{2h'}}{h'} \times e^{-2h'}$ and $a_{32}\frac{\beta e^{2h'}}{h'} \times a_{41}\frac{\beta e^{2h'}}{h'} \times a_{14}\frac{\beta e^{2h'}}{h'} \times a_{23}\frac{\beta e^{2h'}}{h'}$, respectively. Therefore, it is also necessary for $\det \rho^{T_1} < 0$ that $e^{4h'}\beta^2/h'^2$ is greater or of order 1, which again leads to Eq. (A.7). Thus, Lemma 2 is also proved in the case of $\zeta = 1$.

A.3 Proof of Eq. (2.67)

In order to prove Eq. (2.67), we begin with the standard operator expansion of an arbitrary $2 \otimes 2$ operator Q:

$$Q = \frac{1}{4} \sum_{i,j=0,x,y,z} q_{ij} \sigma_1^i \otimes \sigma_2^j, \tag{A.9}$$

where $\sigma_1^0 = \sigma_2^0 = I$ is the two-dimensional identity operator. The coefficients q_{ij} are given by

$$q_{ij} = \mathrm{tr}\big(Q\sigma_1^i \otimes \sigma_2^j\big) \tag{A.10}$$

because $\mathrm{tr}(I \otimes I) = 4$ and the other terms are traceless.

Symmetries that the Hamiltonian (2.59) possesses eliminate many of the coefficients $\{q_{ij}\}$ of the expansion of operators with the same symmetries, such as $\exp(-\beta x H_{\mathrm{tot}}^{\mathrm{op}})$. First, a straightforward calculation shows that the Hamiltonian (2.59) commutes with the global phase flip

$$\begin{aligned} U_{\mathrm{flip}} &= e^{i(\pi/2)\sigma_1^z} \otimes e^{i(\pi/2)\sigma_2^z} \\ &= -\sigma_1^z \otimes \sigma_2^z. \end{aligned} \tag{A.11}$$

This operator flips the signs of σ^x and σ^y. For an operator Q that commutes with U_{flip}, the coefficients $\{q_{0x}, q_{0y}, q_{x0}, q_{y0}, q_{xz}, q_{yz}, q_{zx}, q_{zy}\}$ vanish. For example, we have

$$\begin{aligned} q_{xz} &= \mathrm{tr}[Q(\sigma_1^x \otimes \sigma_2^z)] \\ &= \mathrm{tr}[{U_{\mathrm{flip}}}^{-1} Q U_{\mathrm{flip}} {U_{\mathrm{flip}}}^{-1} (\sigma_1^x \otimes \sigma_2^z) U_{\mathrm{flip}}] \\ &= \mathrm{tr}[Q((-\sigma_1^x) \otimes \sigma_2^z)] \\ &= -q_{xz} = 0. \end{aligned} \tag{A.12}$$

The same argument gives $q_{0x} = q_{0y} = q_{x0} = q_{y0} = q_{xz} = q_{yz} = q_{zx} = q_{zy} = 0$.

Next, the Hamiltonian (2.59) is a real matrix in the σ^z basis. Noting that only σ^y has imaginary elements in this representation, we have, for an operator Q with the symmetry $Q^* = Q$,

$$\begin{aligned} (q_{xy})^* &= \mathrm{tr}[Q^*((\sigma_1^x)^* \otimes (\sigma_2^y)^*)] \\ &= \mathrm{tr}[Q(\sigma_1^x \otimes (-\sigma_2^y))] \\ &= -q_{xy}. \end{aligned} \tag{A.13}$$

On the other hand, the Hermiticity of an operator Q is followed by

$$\begin{aligned} (q_{xy})^* &= \mathrm{tr}[((\sigma_1^x)^\dagger \otimes (\sigma_2^y)^\dagger) Q^\dagger] \\ &= \mathrm{tr}[Q(\sigma_1^x \otimes \sigma_2^y)] \\ &= q_{xy}. \end{aligned} \tag{A.14}$$

The above argument shows $q_{xy} = q_{yx} = 0$.

Finally, the Hamiltonian (2.59) is symmetric with respect to the following set of operations:

$$\begin{aligned} U_{12} &= (e^{i(\pi/2)\sigma_1^x} \otimes e^{i(\pi/2)\sigma_2^x}) P_{12} \\ &= -(\sigma_1^x \otimes \sigma_2^x) P_{12}, \end{aligned} \tag{A.15}$$

where P_{12} is the permutation of the spins 1 and 2. The operator $\sigma_1^x \otimes \sigma_2^x$ flips the signs of σ_1^z and σ_2^z but the permutation P_{12} makes the signs back to the original ones, because the local fields are in the opposite directions in the Hamiltonian (2.59). For an operator Q that commutes with U_{12}, we have

$$\begin{aligned} q_{z0} &= \mathrm{tr}[Q(\sigma_1^z \otimes I)] \\ &= \mathrm{tr}[{U_{12}}^{-1} Q U_{12} {U_{12}}^{-1} (\sigma_1^z \otimes I) U_{12}] \\ &= \mathrm{tr}[Q(I \otimes (-\sigma_2^z))] \\ &= -q_{0z}. \end{aligned} \tag{A.16}$$

To summarize, an operator with the same symmetries as the Hamiltonian (2.59) is expanded in the form

$$Q = \frac{1}{4}\Bigg[q_{00} I \otimes I + q_{z0}(\sigma_1^z \otimes I - I \otimes \sigma_2^z) + \sum_{i=x,y,z} q_{ii}\sigma_1^i \otimes \sigma_2^i\Bigg]. \tag{A.17}$$

In (2.66), the operators $e^{-\beta x H_{\rm tot}^{\rm op}}$ and $e^{-\beta(1-x)H_{\rm tot}^{\rm op}}$ have the same symmetries as the Hamiltonian $H_{\rm tot}^{\rm op}$ and hence are given in the form (A.17).

Since the density operator $\rho = e^{-\beta H_{\rm tot}^{\rm op}}$ is given in the form (A.17), the partial transpose ρ^{T_1} is also of the form (A.17); in the σ_z basis, the partial transpose T_1 only flips the sign of σ_1^y and hence changes only the sign of q_{yy} in the expansion, not the symmetries nor the form of the expansion.

The state $|\phi_-\rangle$ is a non-degenerate eigenstate of the operator ρ^{T_1} if the minimum eigenvalue λ_- is negative. Suppose that the operator ρ^{T_1} commutes with a symmetry operator U. Then the projection operator $|\phi_-\rangle\langle\phi_-|$ should have the same symmetry. This is shown as follows. Since we have

$$\rho^{T_1} U|\phi_-\rangle = U\rho^{T_1}|\phi_-\rangle = \lambda_- U|\phi_-\rangle \tag{A.18}$$

and $|\phi_-\rangle$ is non-degenerate, the vector $U|\phi_-\rangle$ must be the same vector as $|\phi_-\rangle$ except for a phase: $U|\phi_-\rangle = e^{i\theta}|\phi_-\rangle$. Therefore, the projection operator $|\phi_-\rangle\langle\phi_-|$ commutes with U if the negativity is non-zero. This means that $|\phi_-\rangle\langle\phi_-|$ as well as $(|\phi_-\rangle\langle\phi_-|)^{T_1}$ have the same symmetries as the Hamiltonian $H_{\rm tot}^{\rm op}$ and are expanded in the form (A.17).

We thereby arrive at the conclusion that the operator

$$\begin{aligned} &e^{-\beta x H_{\rm tot}^{\rm op}} \hat{n} e^{-\beta(1-x)H_{\rm tot}^{\rm op}} \\ =&e^{-\beta x H_{\rm tot}^{\rm op}}\Big[N(\rho_{\rm op})\big(I\otimes I\big) + 2\big(|\phi_-\rangle\langle\phi_-|\big)^{T_1}\Big]e^{-\beta(1-x)H_{\rm tot}^{\rm op}} \end{aligned} \tag{A.19}$$

has the same symmetries as the Hamiltonian $H_{\rm tot}^{\rm op}$ and hence is expanded in the form (A.17).

A.4 The Eigenvalues of (2.70)

In this section, we prove that in the eigenvalues of the matrix (2.70), only $a_1 - |a_2|$ can have a negative value for $\{J_x, J_y\} \geq J_z \geq 0$ and $0 \geq J_z \geq \{J_x, J_y\}$. The four eigenvalues are given in (2.72). Because $a_1 > 0$, $|a_2| > 0$ and $b_2^2 + b_3^2 > 0$, we obviously have

$$a_1 + |a_2| > 0, \quad b_1 + \sqrt{b_2^2 + b_3^2} > 0. \tag{A.20}$$

Therefore, we only have to prove that $b_1 - \sqrt{b_2^2 + b_3^2} > 0$.

First, we prove this inequality for $h = 0$. For $h = 0$, the eigenvalue $b_1 - \sqrt{b_2^2 + b_3^2}$ reduces to

$$\begin{aligned} &b_1 - \sqrt{b_2^2 + b_3^2} \\ &= e^{\beta J_z}\cosh\big[\beta(J_x + J_y)\big] - e^{-\beta J_z}\sinh\big[\beta|J_x - J_y|\big] \\ &= \frac{1}{2}\big(e^{\beta(J_x+J_y+J_z)} + e^{\beta(-J_x-J_y+J_z)} \\ &\quad - e^{\beta(|J_x-J_y|-J_z)} + e^{\beta(-|J_x-J_y|-J_z)}\big). \end{aligned} \tag{A.21}$$

For $\{J_x, J_y\} \geq J_z \geq 0$, we have

$$e^{\beta(J_x+J_y+J_z)} - e^{\beta(|J_x-J_y|-J_z)} \geq 0, \tag{A.22}$$

which leads to $b_1 - \sqrt{b_2^2+b_3^2} > 0$. For $0 \geq J_z \geq \{J_x, J_y\}$, we have

$$\begin{aligned} & e^{\beta(-J_x-J_y+J_z)} - e^{\beta(|J_x-J_y|-J_z)} \\ =& \begin{cases} 2e^{-\beta J_y} \sinh\bigl[\beta(-J_x+J_z)\bigr] & \text{for } 0 \geq J_z \geq J_x \geq J_y, \\ 2e^{-\beta J_x} \sinh\bigl[\beta(-J_y+J_z)\bigr] & \text{for } 0 \geq J_z \geq J_y \geq J_x. \end{cases} \end{aligned} \tag{A.23}$$

Because $-J_x + J_z \geq 0$ and $-J_y + J_z \geq 0$,

$$e^{\beta(-J_x-J_y+J_z)} - e^{\beta(|J_x-J_y|-J_z)} \geq 0 \tag{A.24}$$

for $0 \geq J_z \geq \{J_x, J_y\}$, which also leads to $b_1 - \sqrt{b_2^2+b_3^2} > 0$. Thus, $b_1 - \sqrt{b_2^2+b_3^2} > 0$ is proved for $h = 0$.

Next, we prove $b_1^2 - b_2^2 - b_3^2 > 0$ for arbitrary h, which is equivalent to $b_1 - \sqrt{b_2^2+b_3^2} > 0$ because $b_1 + \sqrt{b_2^2+b_3^2} > 0$. The value of $b_1^2 - b_2^2 - b_3^2$ is calculated as follows:

$$\begin{aligned} & b_1^2 - b_2^2 - b_3^2 \\ =& e^{2\beta J_z}\left(\cosh^2 \beta J_2 - \frac{4h^2}{J_2^2}\sinh^2 \beta J_2\right) - e^{-2\beta J_z}\sinh^2 \beta J_1 \\ =& e^{2\beta J_z}\left[1 + \left(1 - \frac{4h^2}{J_2^2}\right)\sinh^2 \beta J_2\right] - e^{-2\beta J_z}\sinh^2 \beta J_1 \\ =& e^{2\beta J_z} + \frac{e^{2\beta J_z}(J_x+J_y)^2}{J_2^2}\sinh^2 \beta J_2 - e^{-2\beta J_z}\sinh^2 \beta J_1. \end{aligned} \tag{A.25}$$

Only the second term depends on h through $J_2 = \sqrt{4h^2 + (J_x+J_y)^2}$. The term $(\sinh \beta J_2 / J_2)^2$ is a monotonically increasing function of J_2 for $J_2 > 0$, while J_2 is a monotonically increasing function of h^2. Therefore, $b_1^2 - b_2^2 - b_3^2$ is also a monotonically increasing function of h^2. Since we already proved that $b_1^2 - b_2^2 - b_3^2$ is positive for $h = 0$, we obtain $b_1^2 - b_2^2 - b_3^2 > 0$ for any values of h, and thus $b_1 - \sqrt{b_2^2+b_3^2} > 0$ is proved.

Appendix B

Appendix for Chapter 3

B.1 Relation between Theorem 1 and the quantum discord

In Theorem 1, we show a necessary condition for the entanglement entanglement to be generated by the maximizing local fields. In this section, we answer the following question; if the condition (3.10) is satisfied, can the quantum discord still exist? The answer to this question is yes and we show an example in the following.

First, we review the definition of the quantum discord [57]. The quantum discord $\mathcal{Q}(\rho_{12})$ between the spins 1 and 2 is defined as follows:

$$\mathcal{Q}(\rho_{12}) \equiv \mathcal{I}(\rho_{12}) - \mathcal{J}(\rho_{12}), \tag{B.1}$$

where $\mathcal{I}(\rho)$ is the quantum mutual information defined by

$$\mathcal{I}(\rho) \equiv S(\rho_1) + S(\rho_2) - S(\rho_{12}) \tag{B.2}$$

with $S(\rho)$ the von Neumann entropy $S(\rho) \equiv \mathrm{tr}(\rho \ln \rho)$. On the other hand, $\mathcal{J}(\rho)$ is the optimized classical mutual information, which is the maximum information obtained from the measurement of the spins 1 or 2, and is defined by

$$\mathcal{J}(\rho) \equiv S(\rho_2) - \min_{\Pi_j} \sum_j p_j S(\rho_{2|\Pi_j}), \tag{B.3}$$

where $S(\rho_2)$ is the initial von Neumann entropy of the spin 2 and $\sum_j p_j S(\rho_{2|\Pi_j})$ is the average of the von Neumann entropy after the measurement of the spin 1 in the basis of Π_j. If the quantum discord (B.1) has a non-zero value, the correlation between these two spins may not be explained by classical theory.

Let us consider the Hamiltonian

$$H_{\mathrm{int}} = \sigma_1^x \sigma_3^x + \sigma_3^x \sigma_2^x. \tag{B.4}$$

This is a transverse Ising chain and satisfies the condition (3.10) as

$$\begin{aligned} &[H_A(\sigma_1), H_B(\sigma_2)] = 0, \\ &H_A = \sigma_1^x \sigma_3^x, \\ &H_B = \sigma_3^x \sigma_2^x. \end{aligned} \tag{B.5}$$

Therefore, the entanglement can never exist between the spins 1 and 2 in the thermal state of $H_{\mathrm{int}} + h_1^z\sigma_1^z + h_2^z\sigma_2^z$ however we modulate h_1^z and h_3^z. Indeed, the density matrix ρ_{12} is

$$\rho_{12} = \begin{pmatrix} 0.6627 & 0 & 0 & 0.09865 \\ 0 & 0.1513 & 0.09865 & 0 \\ 0 & 0.09865 & 0.1513 & 0 \\ 0.09865 & 0 & 0 & 0.03456 \end{pmatrix}. \tag{B.6}$$

for $h_1^z = h_2^z = 1$ and $T = 1$. This system has no entanglement. However, it has a non-zero quantum discord. We utilize the criterion in Ref. [58] to prove this. First, we separate the density matrix into the following four blocks:

$$\begin{aligned} \rho^{11} &= \mathrm{tr}^1\left(|\uparrow_1\rangle\langle\uparrow_1|\rho_{12}\right) = \begin{pmatrix} 0.6627 & 0 \\ 0 & 0.1513 \end{pmatrix}, \ \rho^{12} = \mathrm{tr}^1\left(|\uparrow_1\rangle\langle\downarrow_1|\rho_{12}\right) = \begin{pmatrix} 0 & 0.09865 \\ 0.09865 & 0 \end{pmatrix}, \\ \rho^{21} &= \mathrm{tr}^1\left(|\downarrow_1\rangle\langle\uparrow_1|\rho_{12}\right) = \begin{pmatrix} 0 & 0.09865 \\ 0.09865 & 0 \end{pmatrix}, \ \rho^{22} = \mathrm{tr}^1\left(|\downarrow_1\rangle\langle\downarrow_1|\rho_{12}\right) = \begin{pmatrix} 0.1513 & 0 \\ 0 & 0.03456 \end{pmatrix}, \end{aligned} \tag{B.7}$$

where tr^1 denotes the trace operation only on the spin 1. A necessary and sufficient condition for zero discord is given by the following two statements:

$$[\rho^{ij}, (\rho^{ij})^\dagger] = 0 \text{ for } i, j = 1, 2 \tag{B.8}$$

and

$$[\rho^{ij}, \rho^{i'j'}] = 0 \text{ for } i, j, i', j' = 1, 2. \tag{B.9}$$

The density matrix (B.6) satisfies the first condition (B.8) because it is a real matrix. However, the second (B.9) condition is not satisfied. Indeed,

$$\rho^{11}\rho^{12} = \begin{pmatrix} 0 & 0.0653822 \\ 0.0149309 & 0 \end{pmatrix}, \ \rho^{12}\rho^{11} = \begin{pmatrix} 0 & 0.0149309 \\ 0.0653822 & 0 \end{pmatrix}, \tag{B.10}$$

and we have $\rho^{11}\rho^{12} \neq \rho^{12}\rho^{11}$. Therefore, there exists a quantum discord between the spins 1 and 2. This shows that the condition in Theorem 1 is applicable only to the existence of the entanglement. So far, we are not sure whether there exists a condition for the indirect interaction to generate a quantum discord.

B.2 In the case $h_{1\mathbf{op}} - h_{N\mathbf{op}} = O(\beta^{-\tilde{\kappa}})$ with $\tilde{\kappa} \leq 0$ in the case (b)

Here, we discuss the case of $h_{1\mathrm{op}} - h_{N\mathrm{op}} = O(\beta^{-\tilde{\kappa}})$ with $\tilde{\kappa} \leq 0$ in the case (b). In this case, we cannot consider the unperturbed states $|\uparrow_1\downarrow_N\rangle \otimes |\psi^n_{\mathrm{media}}\rangle$ and $|\downarrow_1\uparrow_N\rangle \otimes |\psi^n_{\mathrm{media}}\rangle$ independently because their eigenvalues are almost degenerate. Then, the magnitudes of the elements F_1 and F_2 can be different from the ones in (3.34). We can still apply the same calculation to the other parameters $\{P^{\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}, P^{\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}\}$ as in the case $\tilde{\kappa} > 0$; they are of order $\beta^{2\kappa_1+2\kappa_N}$.

We here prove that F_1^2 and F_2^2 are of order $\beta^{2\kappa_1+2\kappa_N}$ or higher. In order to prove this, we separate H_1+H_N as follows;

$$\begin{aligned}
H_1+H_N &= H_{\rm LO}+\delta H_{\rm LO},\\
H_{\rm LO} &= \frac{1}{2}(h_{\rm 1op}+h_{N\rm op}+h_0)\sigma_1^z+\frac{1}{2}(h_{\rm 1op}+h_{N\rm op}-h_0)\sigma_N^z\\
&= -(h_{\rm 1op}+h_{N\rm op})\big(|\uparrow_1\uparrow_N\rangle\langle\uparrow_1\uparrow_N|-|\downarrow_1\downarrow_N\rangle\langle\downarrow_1\downarrow_N|\big)-h_0\big(|\uparrow_1\downarrow_N\rangle\langle\uparrow_1\downarrow_N|-|\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N|\big),\\
\delta H_{\rm LO} &= -(h_{\rm 1op}-h_{N\rm op}-h_0)\big(|\uparrow_1\downarrow_N\rangle\langle\uparrow_1\downarrow_N|-|\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N|\big),\\
&= \frac{1}{2}(h_{\rm 1op}-h_{N\rm op}-h_0)(\sigma_1^z-\sigma_N^z),
\end{aligned} \tag{B.11}$$

where we define as $h_0=O(\beta^{\tilde{\kappa_0}})$ with $0<\tilde{\kappa_0}<1$, and regard $\beta\delta H_{\rm LO}$ as perturbation. The unperturbed density matrix $\tilde{\rho}_{\rm tot}^{(0)}$ is given by

$$\tilde{\rho}_{\rm tot}^{(0)}=e^{-\beta\tilde{H}_{\rm tot}^{(0)}}, \tag{B.12}$$

where $\tilde{H}_{\rm tot}^{(0)}$ is defined by

$$\tilde{H}_{\rm tot}^{(0)}=H_{\rm media}+H_{\rm couple}+H_{\rm LO}. \tag{B.13}$$

Because $h_0=O(\beta^{-\tilde{\kappa}_0})$ with $0<\tilde{\kappa}_0<1$, the magnitudes of the unperturbed elements $\{F_1^{(0)},F_2^{(0)}\}$ of $\tilde{\rho}_{\rm tot}^{(0)}$ are given by

$$O(\beta^{\kappa_1+\kappa_N+\kappa_0'})\text{ and }O(\beta^{\kappa_1+\kappa_N+\kappa_0}), \tag{B.14}$$

where

$$\kappa_0=\min(\kappa_N,1)\text{ and }\kappa_0'=\min(\kappa_N,\tilde{\kappa}_0,1). \tag{B.15}$$

The density matrix in the first-order perturbation is given by

$$\rho_{\rm tot}=\frac{1}{Z^{(0)}+\delta Z}\left(e^{-\beta\tilde{H}_{\rm tot}^{(0)}}+\beta\int_0^1 e^{-\beta x\tilde{H}_{\rm tot}^{(0)}}\delta H_{\rm LO}e^{-\beta(1-x)\tilde{H}_{\rm tot}^{(0)}}dx\right), \tag{B.16}$$

where $Z^{(0)}$ is the partition function of the density matrix $\tilde{\rho}_{\rm tot}^{(0)}$, while $Z^{(0)}+\delta Z$ is the partition function of $\rho_{\rm tot}$. The elements F_1 and F_2 are given by

$$\begin{aligned}
F_1&={\rm tr}_{1N}\langle\uparrow_1\downarrow_N|\rho_{\rm tot}|\downarrow_1\uparrow_N\rangle,\\
F_2&={\rm tr}_{1N}\langle\uparrow_1\uparrow_N|\rho_{\rm tot}|\downarrow_1\downarrow_N\rangle,
\end{aligned} \tag{B.17}$$

where ${\rm tr}_{1N}$ denotes the trace operation on the spins except the focused spins 1 and N. The first-order perturbations of the elements F_1 and F_2 are

$$\begin{aligned}
-\beta(h_{\rm 1op}-h_{N\rm op}-h_0){\rm tr}_{1N}\int_0^1 dx\Big(&\langle\uparrow_1\downarrow_N|e^{-\beta x\tilde{H}_{\rm tot}^{(0)}}|\uparrow_1\downarrow_N\rangle\langle\uparrow_1\downarrow_N|e^{-\beta(1-x)\tilde{H}_{\rm tot}^{(0)}}|\downarrow_1\uparrow_N\rangle\\
&-\langle\uparrow_1\downarrow_N|e^{-\beta x\tilde{H}_{\rm tot}^{(0)}}|\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N|e^{-\beta(1-x)\tilde{H}_{\rm tot}^{(0)}}|\downarrow_1\uparrow_N\rangle\Big)
\end{aligned} \tag{B.18}$$

and

$$-\beta(h_{1\mathrm{op}} - h_{N\mathrm{op}} - h_0)\mathrm{tr}_{1N}\int_0^1 dx\Big(\langle\uparrow_1\uparrow_N|e^{-\beta x\tilde{H}^{(0)}_{\mathrm{tot}}}|\uparrow_1\downarrow_N\rangle\langle\uparrow_1\downarrow_N|e^{-\beta(1-x)\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\downarrow_N\rangle$$
$$-\langle\uparrow_1\uparrow_N|e^{-\beta x\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N|e^{-\beta(1-x)\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\downarrow_N\rangle\Big), \quad \text{(B.19)}$$

respectively.

In order to estimate the order of the first-order perturbations of F_1 and F_2, we introduce $\{|\psi^{(0),n,\xi}_{\mathrm{tot},\xi}\rangle\}$ and $\{E^{(0),n,\xi}_{\mathrm{tot}}\}$ as defined in Eqs. (3.42) and (3.43), but for $\tilde{H}^{(0)}_{\mathrm{tot}}$. First, we have

$$\langle\uparrow_1\downarrow_N|e^{-\beta(1-x)\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\uparrow_N\rangle = \sum_{\substack{n=1\\ \xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow}}^{2^{N-2}} e^{-\beta(1-x)E^{(0),n,\xi}_{\mathrm{tot}}}|\psi^{(0),n,\xi}_{\mathrm{media},\uparrow\downarrow}\rangle\langle\psi^{(0),n,\xi}_{\mathrm{media},\downarrow\uparrow}|. \quad \text{(B.20)}$$

From the calculation in Appendix B.3, we obtain

$$\Big|\Big||\psi^{(0),n,\xi}_{\mathrm{media},\uparrow\downarrow}\rangle\langle\psi^{(0),n,\xi}_{\mathrm{media},\downarrow\uparrow}|\Big|\Big| = O(\beta^{\kappa_1+\kappa_N}). \quad \text{(B.21)}$$

Therefore, we have

$$\frac{1}{Z^{(0)}(x)}\big|\big|\langle\uparrow_1\downarrow_N|e^{-\beta(1-x)\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\uparrow_N\rangle\big|\big| = O(\beta^{\kappa_1+\kappa_N}), \quad \text{(B.22)}$$

where

$$\frac{1}{Z^{(0)}(x)} \equiv \mathrm{tr}\big(e^{-\beta x\tilde{H}^{(0)}_{\mathrm{tot}}}\big). \quad \text{(B.23)}$$

Similarly, we obtain

$$\begin{aligned}
&\frac{1}{Z^{(0)}(x)}\big|\big|\langle\uparrow_1\uparrow_N|e^{-\beta x\tilde{H}^{(0)}_{\mathrm{tot}}}|\uparrow_1\downarrow_N\rangle\big|\big| = O(\beta^{\kappa_N}),\\
&\frac{1}{Z^{(0)}(1-x)}\big|\big|\langle\uparrow_1\downarrow_N|e^{-\beta(1-x)\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\downarrow_N\rangle\big|\big| = O(\beta^{\kappa_1}),\\
&\frac{1}{Z^{(0)}(x)}\big|\big|\langle\uparrow_1\uparrow_N|e^{-\beta x\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\uparrow_N\rangle\big|\big| = O(\beta^{\kappa_1}),\\
&\frac{1}{Z^{(0)}(1-x)}\big|\big|\langle\downarrow_1\uparrow_N|e^{-\beta(1-x)\tilde{H}^{(0)}_{\mathrm{tot}}}|\downarrow_1\uparrow_N\rangle\big|\big| = O(\beta^{\kappa_N}).
\end{aligned} \quad \text{(B.24)}$$

As a result, we obtain the first-order perturbations of F_1 and F_2 as

$$O(\beta^{\kappa_1+\kappa_N+1-\tilde{\kappa}_0}), \quad \text{(B.25)}$$

where we utilized $\beta(h_{1\mathrm{op}} - h_{N\mathrm{op}} - h_0) = O(\beta^{1-\tilde{\kappa}_0})$. We can similarly calculate higher-order perturbations of F_1 and F_2 to see that they are of order higher than (B.25). Thus, in the case $h_{1\mathrm{op}} - h_{N\mathrm{op}} = O(\beta^{-\tilde{\kappa}})$ with $\tilde{\kappa} \le 0$, F_1^2 and F_2^2 are of order higher than $\{P^{\uparrow\uparrow}_{\uparrow\downarrow,\downarrow\uparrow}, P^{\uparrow\uparrow}_{\uparrow\uparrow,\downarrow\downarrow}\}$.

B.3 Calculation in the case (b)

Here, we derive the approximated form of $|\psi_{\rm tot}^{n_0,\uparrow\uparrow}\rangle$ in Eq. (3.42). In order to calculate perturbation of $|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle$, we employ the general perturbation theory,

$$|\psi\rangle = |\psi^{(0)}\rangle + \big(I_{\rm tot} - |\psi^{(0)}\rangle\langle\psi^{(0)}|\big)\frac{1}{E^{(0)} - H^{(0)}}\delta H|\psi\rangle, \tag{B.26}$$

where $H^{(0)}$ is the unperturbed Hamiltonian, $|\psi^{(0)}\rangle$ and $E^{(0)}$ are the unperturbed eigenstate and the unperturbed eigenvalue, respectively, and δH is the perturbative Hamiltonian. We calculate each element of $|\psi_{\rm media}^{n_0,\uparrow\uparrow}\rangle$, namely $\{|\psi_{{\rm media},\xi}^{n_0,\uparrow\uparrow}\rangle\}$ in Eq. (3.42) with $\xi = \uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow, \downarrow\downarrow$. We first calculate $|\psi_{{\rm media},\downarrow\uparrow}^{n_0,\uparrow\uparrow}\rangle$, which is given by the first-order perturbation;

$$\begin{aligned}
&|\psi_{{\rm media},\downarrow\uparrow}^{n_0,\uparrow\uparrow}\rangle = |\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N| \otimes I_{\rm media} \\
&\quad\times \Big[I_{\rm tot} - \big(|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle\big)\big(\langle\uparrow_1\uparrow_N| \otimes \langle\psi_{\rm media}^{n_0}|\big)\Big]\frac{1}{E^{(0)} - H^{(0)}}H_{\rm couple}|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle \\
&= \sum_{n=1}^{2^{N-2}} |\downarrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n}\rangle \frac{\big(\langle\downarrow_1\uparrow_N| \otimes \langle\psi_{\rm media}^{n}|\big)H_{\rm couple}\big(|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle\big)}{E_{\rm media}^{n_0} - h_{1{\rm op}} - h_{N{\rm op}} - (E_{\rm media}^{n} + h_{1{\rm op}} - h_{N{\rm op}})},
\end{aligned} \tag{B.27}$$

where we put $E^{(0)}$ to $E_{\rm media}^{n_0} - h_{1{\rm op}} - h_{N{\rm op}}$, $H^{(0)}$ to $H_1 + H_N + H_{\rm media}$, $|\psi^{(0)}\rangle$ to $|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle$ and δH to $H_{\rm couple}$ in Eq. (B.26). As has been stated, we assume $h_{1{\rm op}} = O(\beta^{-\kappa_1})$ and $h_{N{\rm op}} = O(\beta^{-\kappa_N})$ with $\kappa_1, \kappa_N > 0$. On the other hand, the eigenvalues $\{E_{\rm media}^{n}\}$ are of order β^0 because the media-spin Hamiltonian $H_{\rm media}$ is fixed. Therefore, Eq. (B.27) can be approximated by

$$\begin{aligned}
&|\psi_{{\rm media},\downarrow\uparrow}^{n_0,\uparrow\uparrow}\rangle \\
&= \sum_{n=1}^{2^{N-2}} |\downarrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n}\rangle \frac{\big(\langle\downarrow_1\uparrow_N| \otimes \langle\psi_{\rm media}^{n}|\big)H_{\rm couple}\big(|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle\big)}{-2h_{1{\rm op}}}\Big(1 + O(\beta^{\kappa_1})\Big).
\end{aligned} \tag{B.28}$$

We can sum the leading term over the label n to obtain

$$|\psi_{{\rm media},\downarrow\uparrow}^{n_0,\uparrow\uparrow}\rangle = \frac{1}{-2h_{1{\rm op}}}\big(|\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N| \otimes I_{\rm media}\big)H_{\rm couple}\big(|\uparrow_1\uparrow_N\rangle \otimes |\psi_{\rm media}^{n_0}\rangle\big), \tag{B.29}$$

where

$$\sum_{n=1}^{2^{N-2}} |\psi_{\rm media}^{n}\rangle\langle\psi_{\rm media}^{n}| = I_{\rm media} \tag{B.30}$$

is the identity operator in the whole space of the media spins. Using Eqs. (3.40) and (3.53) in Eq. (B.28), we have

$$\begin{aligned}
|\psi_{{\rm media},\downarrow\uparrow}^{n_0,\uparrow\uparrow}\rangle = \frac{J}{-2h_{1{\rm op}}}|\downarrow_1\uparrow_N\rangle \otimes \Big(&\gamma s_{n_0}|\downarrow_2\rangle|\tilde{\psi}_{n_0}^{\uparrow\uparrow}\rangle|\uparrow_{N-1}\rangle + \gamma t_{n_0}|\downarrow_2\rangle|\tilde{\psi}_{n_0}^{\uparrow\downarrow}\rangle|\downarrow_{N-1}\rangle \\
&+ u_{n_0}|\uparrow_2\rangle|\tilde{\psi}_{n_0}^{\downarrow\uparrow}\rangle|\uparrow_{N-1}\rangle + w_{n_0}|\uparrow_2\rangle|\tilde{\psi}_{n_0}^{\downarrow\downarrow}\rangle|\downarrow_{N-1}\rangle + O(\beta^{\kappa_1})\Big),
\end{aligned} \tag{B.31}$$

Second, the leading term of $|\psi^{n_0,\uparrow\uparrow}_{\mathrm{media},\uparrow\downarrow}\rangle$ is similarly given as follows:

$$
\begin{aligned}
&|\psi^{n_0,\uparrow\uparrow}_{\mathrm{media},\uparrow\downarrow}\rangle \\
&=\sum_{n=1}^{2^{N-2}} |\uparrow_1\downarrow_N\rangle\otimes|\psi^{n}_{\mathrm{media}}\rangle \frac{\big(\langle\uparrow_1\downarrow_N|\otimes\langle\psi^{n}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{-2h_{N\mathrm{op}}}\Big(1+O(\beta^{\kappa_N})\Big) \\
&=\frac{1}{-2h_{N\mathrm{op}}}\big(|\uparrow_1\downarrow_N\rangle\langle\uparrow_1\downarrow_N|\otimes I_{\mathrm{media}}\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big) \\
&=\frac{J}{-2h_{N\mathrm{op}}}|\uparrow_1\downarrow_N\rangle\otimes\Big(\gamma s_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|\downarrow_{N-1}\rangle+t_{n_0}|\uparrow_2\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|\uparrow_{N-1}\rangle \\
&\quad+\gamma u_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|\downarrow_{N-1}\rangle+w_{n_0}|\downarrow_2\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|\uparrow_{N-1}\rangle+O(\beta^{\kappa_N})\Big).
\end{aligned}
\tag{B.32}
$$

Third, we calculate $|\psi^{n_0,\uparrow\uparrow}_{\mathrm{media},\downarrow\downarrow}\rangle$, which is given by the second-order perturbation;

$$
\begin{aligned}
&|\psi^{n_0,\uparrow\uparrow}_{\mathrm{media},\downarrow\downarrow}\rangle=\sum_{n,n'=1}^{2^{N-2}}|\downarrow_1\downarrow_N\rangle\otimes|\psi^{n}_{\mathrm{media}}\rangle \\
&\Bigg[\frac{\big(\langle\downarrow_1\downarrow_N|\otimes\langle\psi^{n}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\downarrow_N\rangle\otimes|\psi^{n'}_{\mathrm{media}}\rangle\big)}{E^{n_0}_{\mathrm{media}}-h_{1\mathrm{op}}-h_{N\mathrm{op}}-(E^{n}_{\mathrm{media}}+h_{1\mathrm{op}}+h_{N\mathrm{op}})}\cdot\frac{\big(\langle\uparrow_1\downarrow_N|\otimes\langle\psi^{n'}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{E^{n_0}_{\mathrm{media}}-h_{1\mathrm{op}}-h_{N\mathrm{op}}-(E^{n'}_{\mathrm{media}}-h_{1\mathrm{op}}+h_{N\mathrm{op}})} \\
&+\frac{\big(\langle\downarrow_1\downarrow_N|\otimes\langle\psi^{n}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\downarrow_1\uparrow_N\rangle\otimes|\psi^{n'}_{\mathrm{media}}\rangle\big)}{E^{n_0}_{\mathrm{media}}-h_{1\mathrm{op}}-h_{N\mathrm{op}}-(E^{n}_{\mathrm{media}}+h_{1\mathrm{op}}+h_{N\mathrm{op}})}\cdot\frac{\big(\langle\downarrow_1\uparrow_N|\otimes\langle\psi^{n'}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{E^{n_0}_{\mathrm{media}}-h_{1\mathrm{op}}-h_{N\mathrm{op}}-(E^{n'}_{\mathrm{media}}+h_{1\mathrm{op}}-h_{N\mathrm{op}})}\Bigg].
\end{aligned}
\tag{B.33}
$$

By utilizing the assumptions $h_{1\mathrm{op}}=O(\beta^{-\kappa_1})>0$, $h_{N\mathrm{op}}=O(\beta^{-\kappa_N})>0$ with $\kappa_1\geq\kappa_N>0$ and $E^{n}_{\mathrm{media}}=O(\beta^0)$, we can approximate (B.33) as

$$
\begin{aligned}
|\psi^{n_0,\uparrow\uparrow}_{\mathrm{media},\downarrow\downarrow}\rangle=\sum_{n,n'=1}^{2^{N-2}}|\downarrow_1\downarrow_N\rangle\otimes|\psi^{n}_{\mathrm{media}}\rangle&\Bigg[\frac{\big(\langle\downarrow_1\downarrow_N|\otimes\langle\psi^{n}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\downarrow_N\rangle\otimes|\psi^{n'}_{\mathrm{media}}\rangle\big)}{-2(h_{1\mathrm{op}}+h_{N\mathrm{op}})} \\
&\times\frac{\big(\langle\uparrow_1\downarrow_N|\otimes\langle\psi^{n'}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{-2h_{N\mathrm{op}}}\Big(1+O(\beta^{\kappa_N})\Big) \\
&+\frac{\big(\langle\downarrow_1\downarrow_N|\otimes\langle\psi^{n}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\downarrow_1\uparrow_N\rangle\otimes|\psi^{n'}_{\mathrm{media}}\rangle\big)}{-2(h_{1\mathrm{op}}+h_{N\mathrm{op}})} \\
&\times\frac{\big(\langle\downarrow_1\uparrow_N|\otimes\langle\psi^{n'}_{\mathrm{media}}|\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{-2h_{1\mathrm{op}}}\Big(1+O(\beta^{\kappa_1})\Big)\Bigg].
\end{aligned}
\tag{B.34}
$$

We can calculate the leading terms of (B.34) as

$$
\begin{aligned}
&\frac{\big(|\downarrow_1\downarrow_N\rangle\langle\downarrow_1\downarrow_N|\otimes I_{\mathrm{media}}\big)H_{\mathrm{couple}}\big(|\uparrow_1\downarrow_N\rangle\langle\uparrow_1\downarrow_N|\otimes I_{\mathrm{media}}\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{4h_{N\mathrm{op}}(h_{1\mathrm{op}}+h_{N\mathrm{op}})} \\
&+\frac{\big(|\downarrow_1\downarrow_N\rangle\langle\downarrow_1\downarrow_N|\otimes I_{\mathrm{media}}\big)H_{\mathrm{couple}}\big(|\downarrow_1\uparrow_N\rangle\langle\downarrow_1\uparrow_N|\otimes I_{\mathrm{media}}\big)H_{\mathrm{couple}}\big(|\uparrow_1\uparrow_N\rangle\otimes|\psi^{n_0}_{\mathrm{media}}\rangle\big)}{4h_{1\mathrm{op}}(h_{1\mathrm{op}}+h_{N\mathrm{op}})}.
\end{aligned}
\tag{B.35}
$$

Upon using Eqs. (3.40) and (3.53), the numerator of the first term in (B.35) reduces to

$$
\begin{aligned}
&\big(|{\downarrow_1\downarrow_N}\rangle\langle{\downarrow_1\downarrow_N}|\otimes I_{\text{media}}\big)H_{\text{couple}}\big(|{\uparrow_1\downarrow_N}\rangle\langle{\uparrow_1\downarrow_N}|\otimes I_{\text{media}}\big)H_{\text{couple}}\big(|{\uparrow_1\uparrow_N}\rangle\otimes|\psi^{n_0}_{\text{media}}\rangle\big)\\
=&\big(|{\downarrow_1\downarrow_N}\rangle\langle{\downarrow_1\downarrow_N}|\otimes I_{\text{media}}\big)H_{\text{couple}}J\\
\times&\Big[|{\uparrow_1\downarrow_N}\rangle\otimes\big(\gamma s_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+t_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle+\gamma u_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+w_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\big)\Big]\\
=&J^2|{\downarrow_1\downarrow_N}\rangle\otimes\big(\gamma^2 s_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+\gamma t_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\\
&+\gamma u_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+w_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\big).
\end{aligned}
\tag{B.36}
$$

Similarly, the numerator of the second term in (B.35) reduces to

$$
\begin{aligned}
&\big(|{\downarrow_1\downarrow_N}\rangle\langle{\downarrow_1\downarrow_N}|\otimes I_{\text{media}}\big)H_{\text{couple}}\big(|{\downarrow_1\uparrow_N}\rangle\langle{\downarrow_1\uparrow_N}|\otimes I_{\text{media}}\big)H_{\text{couple}}\big(|{\uparrow_1\uparrow_N}\rangle\otimes|\psi^{n_0}_{\text{media}}\rangle\big)\\
=&\big(|{\downarrow_1\downarrow_N}\rangle\langle{\downarrow_1\downarrow_N}|\otimes I_{\text{media}}\big)H_{\text{couple}}J\Big[|{\downarrow_1\uparrow_N}\rangle\otimes\big(\gamma s_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle+\gamma t_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle\\
&+u_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle+w_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle\big)\Big]\\
=&J^2|{\downarrow_1\downarrow_N}\rangle\otimes\big(\gamma^2 s_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+\gamma t_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\\
&+\gamma u_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+w_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\big),
\end{aligned}
\tag{B.37}
$$

which is equal to (B.36). As a result, we arrive at

$$
\begin{aligned}
|\psi^{n_0,\uparrow\uparrow}_{\text{media},\downarrow\downarrow}\rangle=&\frac{J^2}{4h_{1\text{op}}h_{N\text{op}}}\Big[\big(\gamma^2 s_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+\gamma t_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\\
&+\gamma u_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+w_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\big)+O(\beta^{\kappa_N})\Big],
\end{aligned}
\tag{B.38}
$$

Next, we calculate each element of $|\psi^{n_0,\uparrow\downarrow}_{\text{tot}}\rangle$, namely $\{|\psi^{n_0,\uparrow\downarrow}_{\text{tot},\xi}\rangle\}$ in Eq. (3.42) with $\xi=\uparrow\uparrow,\uparrow\downarrow,\downarrow\uparrow,\downarrow\downarrow$. By similar calculations, we obtain

$$
\begin{aligned}
|\psi^{n_0,\uparrow\downarrow}_{\text{media},\uparrow\uparrow}\rangle=&\frac{J}{2h_{N\text{op}}}\Big[\big(s_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+\gamma t_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\\
&+u_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle+\gamma w_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle\big)+O(\beta^{\kappa_N})\Big]\\
|\psi^{n_0,\uparrow\downarrow}_{\text{media},\downarrow\downarrow}\rangle=&\frac{J}{-2h_{1\text{op}}}\Big[\big(\gamma s_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle+\gamma t_{n_0}|{\downarrow_2}\rangle|\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle\\
&+u_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle|{\uparrow_{N-1}}\rangle+w_{n_0}|{\uparrow_2}\rangle|\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle|{\downarrow_{N-1}}\rangle\big)+O(\beta^{\kappa_1})\Big].
\end{aligned}
\tag{B.39}
$$

The element $|\psi^{n_0,\uparrow\downarrow}_{\text{media},\downarrow\uparrow}\rangle$ can be calculated from

$$
\begin{aligned}
&|\psi^{n_0,\uparrow\downarrow}_{\text{media},\downarrow\uparrow}\rangle=\sum_{n,n'=1}^{2^{N-2}}|{\downarrow_1\downarrow_N}\rangle\otimes|\psi^{n}_{\text{media}}\rangle\\
&\Bigg[\frac{\big(\langle{\downarrow_1\uparrow_N}|\otimes\langle\psi^{n}_{\text{media}}|\big)H_{\text{couple}}\big(|{\uparrow_1\uparrow_N}\rangle\otimes|\psi^{n'}_{\text{media}}\rangle\big)}{E^{n_0}_{\text{media}}-h_{1\text{op}}+h_{N\text{op}}-(E^{n}_{\text{media}}+h_{1\text{op}}-h_{N\text{op}})}\cdot\frac{\big(\langle{\uparrow_1\uparrow_N}|\otimes\langle\psi^{n'}_{\text{media}}|\big)H_{\text{couple}}\big(|{\uparrow_1\downarrow_N}\rangle\otimes|\psi^{n_0}_{\text{media}}\rangle\big)}{E^{n_0}_{\text{media}}-h_{1\text{op}}+h_{N\text{op}}-(E^{n'}_{\text{media}}-h_{1\text{op}}-h_{N\text{op}})}\\
&+\frac{\big(\langle{\downarrow_1\uparrow_N}|\otimes\langle\psi^{n}_{\text{media}}|\big)H_{\text{couple}}\big(|{\downarrow_1\downarrow_N}\rangle\otimes|\psi^{n'}_{\text{media}}\rangle\big)}{E^{n_0}_{\text{media}}-h_{1\text{op}}+h_{N\text{op}}-(E^{n}_{\text{media}}+h_{1\text{op}}-h_{N\text{op}})}\cdot\frac{\big(\langle{\downarrow_1\downarrow_N}|\otimes\langle\psi^{n'}_{\text{media}}|\big)H_{\text{couple}}\big(|{\uparrow_1\downarrow_N}\rangle\otimes|\psi^{n_0}_{\text{media}}\rangle\big)}{E^{n_0}_{\text{media}}-h_{1\text{op}}+h_{N\text{op}}-(E^{n'}_{\text{media}}+h_{1\text{op}}+h_{N\text{op}})}\Bigg].
\end{aligned}
\tag{B.40}
$$

By utilizing the assumptions $h_{1\mathrm{op}} = O(\beta^{-\kappa_1})$, $h_{N\mathrm{op}} = O(\beta^{-\kappa_N})$ with $\kappa_1 \geq \kappa_N > 0$, $h_{1\mathrm{op}} - h_{N\mathrm{op}} = O(\beta^{-\tilde{\kappa}})$ with $\tilde{\kappa} > 0$ and $E^n_{\mathrm{media}} = O(\beta^0)$, we can approximate (B.40) as

$$
\begin{aligned}
|\psi^{n_0,\uparrow\downarrow}_{\mathrm{media},\downarrow\uparrow}\rangle = \sum_{n,n'=1}^{2^{N-2}} |\downarrow_1\downarrow_N\rangle \otimes |\psi^n_{\mathrm{media}}\rangle \Bigg[& \frac{\big(\langle\downarrow_1\uparrow_N| \otimes \langle\psi^n_{\mathrm{media}}|\big) H_{\mathrm{couple}} \big(|\uparrow_1\uparrow_N\rangle \otimes |\psi^{n'}_{\mathrm{media}}\rangle\big)}{-2(h_{1\mathrm{op}} - h_{N\mathrm{op}})} \\
& \times \frac{\big(\langle\uparrow_1\uparrow_N| \otimes \langle\psi^{n'}_{\mathrm{media}}|\big) H_{\mathrm{couple}} \big(|\uparrow_1\downarrow_N\rangle \otimes |\psi^{n_0}_{\mathrm{media}}\rangle\big)}{-2h_{N\mathrm{op}}} \Big(1 + O(\beta^{\kappa_N}) + O(\beta^{\tilde{\kappa}})\Big) \\
& + \frac{\big(\langle\downarrow_1\uparrow_N| \otimes \langle\psi^n_{\mathrm{media}}|\big) H_{\mathrm{couple}} \big(|\downarrow_1\downarrow_N\rangle \otimes |\psi^{n'}_{\mathrm{media}}\rangle\big)}{-2(h_{1\mathrm{op}} - h_{N\mathrm{op}})} \\
& \times \frac{\big(\langle\downarrow_1\downarrow_N| \otimes \langle\psi^{n'}_{\mathrm{media}}|\big) H_{\mathrm{couple}} \big(|\uparrow_1\downarrow_N\rangle \otimes |\psi^{n_0}_{\mathrm{media}}\rangle\big)}{-2h_{1\mathrm{op}}} \Big(1 + O(\beta^{\kappa_1}) + O(\beta^{\tilde{\kappa}})\Big) \Bigg]. \quad \text{(B.41)}
\end{aligned}
$$

We finally arrive at

$$
\begin{aligned}
|\psi^{n_0,\uparrow\downarrow}_{\mathrm{media},\downarrow\uparrow}\rangle = & \frac{J^2}{4h_{1\mathrm{op}}h_{N\mathrm{op}}} \Big[\big(\gamma s_{n_0} |\downarrow_2\rangle |\tilde{\psi}^{\uparrow\uparrow}_{n_0}\rangle |\downarrow_{N-1}\rangle + \gamma^2 t_{n_0} |\downarrow_2\rangle |\tilde{\psi}^{\uparrow\downarrow}_{n_0}\rangle |\uparrow_{N-1}\rangle \\
& + u_{n_0} |\uparrow_2\rangle |\tilde{\psi}^{\downarrow\uparrow}_{n_0}\rangle |\downarrow_{N-1}\rangle + \gamma w_{n_0} |\uparrow_2\rangle |\tilde{\psi}^{\downarrow\downarrow}_{n_0}\rangle |\uparrow_{N-1}\rangle\big) + O(\beta^{\kappa_N}) + O(\beta^{\tilde{\kappa}}) \Big], \quad \text{(B.42)}
\end{aligned}
$$

where κ' is defined in (3.35).

B.4 Calculation in the case (c)

Here, we derive the perturbed form of $|\phi^{n_0,\uparrow}_{\mathrm{tot}}\rangle$ in Eq. (3.82). Using Eq. (B.26), the leading term of $|\phi^{n_0,\uparrow}_{\mathrm{tot},\downarrow}\rangle$ is given by

$$
|\phi^{n_0,\uparrow}_{\mathrm{tot},\downarrow}\rangle = \sum_{n=1}^{2^{N-1}} |\downarrow_1\rangle \otimes |\phi^n_{\mathrm{media}}\rangle \frac{\big(\langle\downarrow_1| \otimes \langle\phi^n_{\mathrm{media}}|\big) H'_{\mathrm{couple}} \big(|\uparrow_1\rangle \otimes |\phi^{n_0}_{\mathrm{media}}\rangle\big)}{E'^{n_0}_{\mathrm{media}} - h_{1\mathrm{op}} - (E'^{n}_{\mathrm{media}} + h_{1\mathrm{op}})}, \quad \text{(B.43)}
$$

where $h_{1\mathrm{op}} = O(\beta^{-\kappa_1})$ with $\kappa_1 \geq 1$, but $\{E'^n_{\mathrm{media}}\}$ are of order β^0. Then we obtain

$$
|\phi^{n_0,\uparrow}_{\mathrm{tot},\downarrow}\rangle = \sum_{n=1}^{2^{N-1}} |\downarrow_1\rangle \otimes |\phi^n_{\mathrm{media}}\rangle \frac{\big(\langle\downarrow_1| \otimes \langle\phi^n_{\mathrm{media}}|\big) H'_{\mathrm{couple}} \big(|\uparrow_1\rangle \otimes |\phi^{n_0}_{\mathrm{media}}\rangle\big)}{-2h_{1\mathrm{op}}} \Big(1 + O(\beta^{\kappa_1})\Big). \quad \text{(B.44)}
$$

We thereby obtain $|\phi^{n_0,\uparrow}_{\mathrm{tot}}\rangle$ as

$$
\begin{aligned}
|\phi^{n_0,\uparrow}_{\mathrm{tot}}\rangle = |\uparrow_1\rangle \otimes |\phi^{n_0}_{\mathrm{media}}\rangle + \frac{J}{-2h_{1\mathrm{op}}} |\downarrow_1\rangle \otimes \Big(& \gamma s'_{n_0} |\downarrow_2\rangle |\tilde{\phi}^{\uparrow\uparrow}_{n_0}\rangle |\uparrow_N\rangle + \gamma t'_{n_0} |\downarrow_2\rangle |\tilde{\phi}^{\uparrow\downarrow}_{n_0}\rangle |\downarrow_N\rangle \\
& + u'_{n_0} |\uparrow_2\rangle |\tilde{\phi}^{\downarrow\uparrow}_{n_0}\rangle |\uparrow_N\rangle + w'_{n_0} |\uparrow_2\rangle |\tilde{\phi}^{\downarrow\downarrow}_{n_0}\rangle |\downarrow_N\rangle + O(\beta^{\kappa_1}) \Big). \quad \text{(B.45)}
\end{aligned}
$$

B.5 Proof for the existence of the maximized entanglement in three-spin systems

In this section, we prove that the maximized entanglement always exists in the systems with the Hamiltonian (3.99) in the high-temperature limit $\beta \to 0$. We prove this statement by showing that the entanglement exists by letting h_1^z and h_3^z

$$h_1^z = h_3^z = h_0(\beta), \tag{B.46}$$

where we assume

$$h_0(\beta) = O(\beta^{-\kappa_0}) \text{ with } \kappa_0 < -1. \tag{B.47}$$

We regard the interaction Hamiltonian as perturbation and calculate the leading order of the elements $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$ of the density matrix. Then we show the inequality (3.9), which is a necessary and sufficient condition for the existence of the entanglement.

First, we separate the Hamiltonian (3.99) as follows:

$$H_{\text{tot}} = \tilde{H}_{\text{LO}} + \tilde{H}_{\text{int}}, \tag{B.48}$$

where

$$\begin{aligned}
\tilde{H}_{\text{LO}} &= h_1^z\sigma_1^z + h_3^z\sigma_3^z + h_{\text{media}}^z\sigma_2^z + J^z\sigma_1^z\sigma_2^z + J^z\sigma_2^z\sigma_3^z, \\
\tilde{H}_{\text{int}} &= (J^x\sigma_1^x\sigma_2^x + J^y\sigma_1^y\sigma_2^y) + (J^x\sigma_2^x\sigma_3^x + J^y\sigma_2^y\sigma_3^y) \\
&= \Big\{ J\big[\sigma_1^+\sigma_2^- + \sigma_1^-\sigma_2^+ + \gamma(\sigma_1^+\sigma_2^+ + \sigma_1^-\sigma_2^-)\big] + J\big[\sigma_2^+\sigma_3^- + \sigma_2^-\sigma_3^+ + \gamma(\sigma_2^+\sigma_3^+ + \sigma_2^-\sigma_3^-)\big] \Big\},
\end{aligned} \tag{B.49}$$

h_{media}^z has an arbitrary value, $J^x = J(1+\gamma)/2$, $J^y = J(1-\gamma)/2$, and we consider $\tilde{H}_{\text{int}}$ as perturbation. Because $\beta h_0(\beta) \to \infty$ as $\beta \to 0$, we only have to consider the ground state and the first excited state as the unperturbed states, which are given by

$$|\uparrow_1\uparrow_2\uparrow_3\rangle \text{ and } |\uparrow_1\downarrow_2\uparrow_3\rangle \tag{B.50}$$

with the corresponding eigenvalues

$$\epsilon_1 = -2h_0 - h_{\text{media}}^z + 2J^z \text{ and } \epsilon_2 = -2h_0 + h_{\text{media}}^z - 2J^z, \tag{B.51}$$

where we assume $h_{\text{media}}^z > 0$, but the following discussion is also applicable to the case of $h_{\text{media}}^z < 0$. Other excited states have the eigenvalues $\epsilon_1 + O(\beta^{-\kappa_0})$ and hence their thermal mixing can be ignored in the limit $\beta \to 0$.

We consider the states (B.50) up to the second-order perturbation of $\tilde{H}_{\text{int}}$:

$$\begin{aligned}
k_1\Bigg(&|\uparrow_1\uparrow_2\uparrow_3\rangle - \frac{J\gamma}{2h_0 + 2h_{\text{media}}^z - 2J^z}|\downarrow_1\downarrow_2\uparrow_3\rangle \\
&- \frac{J\gamma}{2h_0 + 2h_{\text{media}}^z - 2J^z}|\uparrow_1\downarrow_2\downarrow_3\rangle + \frac{J^2\gamma}{(2h_0 - 2J^z)(2h_0 + 2h_{\text{media}}^z - 2J^z)}|\downarrow_1\uparrow_2\downarrow_3\rangle \Bigg)
\end{aligned} \tag{B.52}$$

and

$$\begin{aligned}
k_2\Bigg(&|\uparrow_1\downarrow_2\uparrow_3\rangle - \frac{J}{2h_0 - 2h_{\text{media}}^z + 2J^z}|\downarrow_1\uparrow_2\uparrow_3\rangle \\
&- \frac{J}{2h_0 - 2h_{\text{media}}^z + 2J^z}|\uparrow_1\uparrow_2\downarrow_3\rangle + \frac{J^2\gamma}{(2h_0 + 2J^z)(2h_0 - 2h_{\text{media}}^z + 2J^z)}|\downarrow_1\downarrow_2\downarrow_3\rangle \Bigg),
\end{aligned} \tag{B.53}$$

where k_1 and k_2 are the respective normalization factor. According to the above expressions, k_1 and k_2 are of order of $1 + O(\beta^{2\kappa_0})$. By mixing these two states with the Boltzmann weights $e^{-\beta\epsilon_1}$ and $e^{-\beta\epsilon_2}$, we obtain the matrix elements $\{F_1, F_2, p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$.

Let us first consider the case $\gamma \neq 1$. In this case, the leading terms of F_1 and $\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}}$ are given as follows;

$$F_1 = \frac{J^2(1+\gamma^2)}{8h_0^2} + O(\beta^{1+2\kappa_0}),$$
$$\sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}} = \frac{J^2\gamma}{4h_0^2} + O(\beta^{1+2\kappa_0}), \tag{B.54}$$

which yields

$$F_1 - \sqrt{p_{\uparrow\uparrow}p_{\downarrow\downarrow}} = \frac{J^2(1-\gamma)^2}{8h_0^2} + O(\beta^{1+2\kappa_0}) > 0 \tag{B.55}$$

in the limit $\beta \to 0$. This gives a non-zero value of the concurrence (3.8) of order $\beta^{2\kappa_0}$ in the case $\gamma \neq 0$.

Next, we consider the case $\gamma = 1$. Because we assumed $J^x \geq J^y \geq J^z$, the equality $J^y = J^z = 0$ is satisfied in the case $\gamma = 1$. In this case, we have to take higher-order approximation because the first term of Eq. (B.55) vanishes. The expansions of F_2 and $\sqrt{p_{\uparrow\downarrow}p_{\downarrow\uparrow}}$ are given by

$$F_2 = \frac{J^2}{4h_0^2} + \frac{(h^z_{\text{media}})^2J^2}{8}\frac{\beta}{h_0^2} - \frac{(h^z_{\text{media}})^2J^2}{4}\frac{\beta}{h_0^3} + O(\beta^{2+3\kappa_0}),$$
$$\sqrt{p_{\downarrow\uparrow}p_{\uparrow\downarrow}} = \frac{J^2}{4h_0^2} + \frac{(h^z_{\text{media}})^2J^2}{8}\frac{\beta}{h_0^2} - \frac{(h^z_{\text{media}})^2J^2}{2}\frac{\beta}{h_0^3} + O(\beta^{2+3\kappa_0}), \tag{B.56}$$

which is followed by

$$F_2 - \sqrt{p_{\uparrow\downarrow}p_{\downarrow\uparrow}} = \frac{\beta}{4h_0^3}J^2(h^z_{\text{media}})^2 + O(\beta^{2+3\kappa_0}). \tag{B.57}$$

If the media field h^z_{media} is equal to zero, the entanglement vanishes for any values of the local fields h^z_1 and h^z_3; this is consistent with Theorem 1. For $h^z_{\text{media}} \neq 0$, the concurrence (3.8) is of order $\beta^{1+3\kappa_0}$ in the case $\gamma = 1$ and increases as the media field h^z_{media} is increased. We have thereby proved that the entanglement in a three-spin system with Hamiltonian (3.99) has always a non-zero value if we choose the local fields properly.

B.6 The critical temperature in the asymmetric phase

In this section, we show analytical calculation of the critical temperature between the phase with the asymmetry $h_{1\text{op}} \neq h_{4\text{op}}$ and the phase with the no entanglement in the four-spin XX chain, that is, on the phase boundary in the upper right area of the phase diagram in Fig. 3.5 (a). For the XX model, the element F_2 vanishes, and hence we obtain the critical temperature as a solution of

$$F_1^2 - p_{\uparrow\uparrow}p_{\downarrow\downarrow} = 0. \tag{B.58}$$

We calculate the elements F_1, $p_{\uparrow\uparrow}$ and $p_{\downarrow\downarrow}$ from perturbation calculations in two steps. Numerical calculation suggests on the phase boundary in the area that the maximizing local fields behave as $h_{1\text{op}} \to \infty$ and $h_{4\text{op}} \simeq h^z_{\text{media}}$ and that $h^z_{\text{media}} \gg J$ and $h^z_{\text{media}} \gg T$. We thereby regard the coupling between the spin 1 and the rest of the system

$$J_x\sigma_1^x\sigma_2^x + J_y\sigma_1^y\sigma_2^y = J(\sigma_1^+\sigma_2^- + \sigma_1^-\sigma_2^+) \tag{B.59}$$

as perturbation in the first step, where $J_x = J_y = J/2$. Then, we regard the interaction Hamiltonian

$$\tilde{H}_{\rm int} = \frac{1}{2}(J\sigma_2^x\sigma_3^x + J\sigma_2^y\sigma_3^y) + \frac{1}{2}(J\sigma_3^x\sigma_4^x + J\sigma_3^y\sigma_4^y)$$
$$= J(\sigma_2^+\sigma_3^- + \sigma_2^-\sigma_3^+) + J(\sigma_3^+\sigma_4^- + \sigma_3^-\sigma_4^+) \tag{B.60}$$

as perturbation in the second step. As the unperturbed states in the limit $h_{1\rm op} \to \infty$, we first use

$$\{|\uparrow_1\rangle \otimes |\phi_n\rangle\}_{n=1}^{8}, \tag{B.61}$$

where $|\phi_n\rangle$ denotes the eigenstates of the system of the spins 2, 3 and 4 and is defined by

$$|\phi_n\rangle = s_n'|\uparrow_2\rangle|\tilde{\phi}_n^{\uparrow\uparrow}\rangle|\uparrow_4\rangle + t_n'|\uparrow_2\rangle|\tilde{\phi}_n^{\uparrow\downarrow}\rangle|\downarrow_4\rangle + u_n'|\downarrow_2\rangle|\tilde{\phi}_n^{\downarrow\uparrow}\rangle|\uparrow_4\rangle + w_n'|\downarrow_2\rangle|\tilde{\phi}_n^{\downarrow\downarrow}\rangle|\downarrow_4\rangle \tag{B.62}$$

for $1 \le n \le 8$, which is the same as in Eq. (3.81). We define them so that their eigenvalues $\{\epsilon_n\}$ are in the non-descending order.

We thereby calculate the first-order contribution of $|\uparrow_1\rangle \otimes |\phi_{n_0}\rangle$ to the elements $\{F_1, F_2\}$ and $\{p_{\uparrow\uparrow}, p_{\uparrow\downarrow}, p_{\downarrow\uparrow}, p_{\downarrow\downarrow}\}$, which we define as $\{F_1^{n_0,\uparrow}, F_2^{n_0,\uparrow}\}$ and $\{p_{\uparrow\uparrow}^{n_0,\uparrow}, p_{\uparrow\downarrow}^{n_0,\uparrow}, p_{\downarrow\uparrow}^{n_0,\uparrow}, p_{\downarrow\downarrow}^{n_0,\uparrow}\}$. We utilize the perturbation calculation in the proof of Theorem 2. From Eqs. (3.86) and (3.87), we have the elements $\{F_1^{n_0,\uparrow}, F_2^{n_0,\uparrow}\}$ as

$$\frac{J}{-2h_{1\rm op}}\Big(\gamma s_{n_0}' w_{n_0}' \langle\phi_{n_0}^{\downarrow\downarrow}|\phi_{n_0}^{\uparrow\uparrow}\rangle + u_{n_0}' t_{n_0}' \langle\phi_{n_0}^{\downarrow\uparrow}|\phi_{n_0}^{\uparrow\downarrow}\rangle\Big) \tag{B.63}$$

and

$$\frac{J}{-2h_{1\rm op}}\Big(\gamma t_{n_0}' u_{n_0}' \langle\phi_{n_0}^{\downarrow\uparrow}|\phi_{n_0}^{\uparrow\downarrow}\rangle + w_{n_0}' s_{n_0}' \langle\phi_{n_0}^{\uparrow\uparrow}|\phi_{n_0}^{\downarrow\downarrow}\rangle\Big). \tag{B.64}$$

From Eq. (3.88), we also have the elements $\{p_{\uparrow\uparrow}^{n_0,\uparrow}, p_{\uparrow\downarrow}^{n_0,\uparrow}, p_{\downarrow\uparrow}^{n_0,\uparrow}, p_{\downarrow\downarrow}^{n_0,\uparrow}\}$ as

$$s_{n_0}'^2 + u_{n_0}'^2,\ t_{n_0}'^2 + w_{n_0}'^2,\ \frac{J^2}{4h_{1\rm op}^2}(\gamma^2 s_{n_0}'^2 + u_{n_0}'^2) \text{ and } \frac{J^2}{4h_{1\rm op}^2}(\gamma^2 t_{n_0}'^2 + w_{n_0}'^2). \tag{B.65}$$

Now, we move to the perturbation in the second step to obtain the eigenstates $\{|\phi_n\rangle\}$ explicitly. Because $h_{\rm media}^z \gg T$, out of eight states $\{|\phi_n\rangle\}$ we consider the perturbations of only the four states $|\uparrow_2\uparrow_3\downarrow_4\rangle$, $|\downarrow_2\uparrow_3\downarrow_4\rangle$, $|\uparrow_2\downarrow_3\downarrow_4\rangle$ and $|\uparrow_2\uparrow_3\uparrow_4\rangle$ with the corresponding eigenvalues $\{h_{4\rm op} - 2h_{\rm media}^z, h_{4\rm op}, h_{4\rm op}, -h_{4\rm op} - 2h_{\rm media}^z\}$. We ignore the perturbation of the other states $|\downarrow_2\downarrow_3\uparrow_4\rangle$, $|\uparrow_2\downarrow_3\uparrow_4\rangle$, $|\downarrow_2\uparrow_3\uparrow_4\rangle$ and $|\downarrow_2\downarrow_3\downarrow_4\rangle$ with the corresponding eigenvalues $\{-h_{4\rm op} + 2h_{\rm media}^z, -h_{4\rm op}, -h_{4\rm op}, h_{4\rm op} + 2h_{\rm media}^z\}$.

The perturbed form of the eigenstate $|\uparrow_2\uparrow_3\downarrow_4\rangle$ up to the second order of $\tilde{H}_{\rm int}$ in (B.60) is given by

$$|\uparrow_2\uparrow_3\downarrow_4\rangle + \frac{J}{2h_{4\rm op} - 2h_{\rm media}^z}|\uparrow_2\downarrow_3\uparrow_4\rangle + \frac{J^2}{(-2h_{4\rm op} + 2h_{\rm media}^z)^2}|\downarrow_2\uparrow_3\uparrow_4\rangle, \tag{B.66}$$

with the eigenvalue change

$$\delta\epsilon_1 \equiv \frac{J^2}{-2h_{4\rm op} + 2h_{\rm media}^z}. \tag{B.67}$$

Suppose here that we choose $h_{4\text{op}} = -h^z_{\text{media}}$. Then, Eq. (B.66) reduces to

$$|\uparrow_2\uparrow_3\downarrow_4\rangle + \frac{J}{-4h^z_{\text{media}}}|\uparrow_2\downarrow_3\uparrow_4\rangle + \frac{J^2}{16h^{z2}_{\text{media}}}|\downarrow_2\uparrow_3\uparrow_4\rangle. \tag{B.68}$$

In the case of $h_{4\text{op}} = -h^z_{\text{media}}$, the eigenvalues of the three zeroth-order states $|\downarrow_2\uparrow_3\downarrow_4\rangle$, $|\uparrow_2\downarrow_3\downarrow_4\rangle$ and $|\uparrow_2\uparrow_3\uparrow_4\rangle$ are degenerate into $-h^z_{\text{media}}$; we define them as $\{\phi^{(0)}_i\}$. The first order contribution of the other states to the states $\{\phi^{(0)}_i\}$ with $i = 2, 3, 4$ are given in the forms

$$\phi^{(1)}_2 = |\downarrow_2\uparrow_3\downarrow_4\rangle,\ \phi^{(1)}_3 = |\downarrow_2\uparrow_3\downarrow_4\rangle + \frac{J}{-4h^z_{\text{media}}}|\downarrow_2\downarrow_3\uparrow_4\rangle,\ \phi^{(1)}_4 = |\uparrow_2\uparrow_3\uparrow_4\rangle \tag{B.69}$$

We define the perturbation matrix as

$$\langle\phi^{(0)}_i|\delta H|\phi^{(1)}_j\rangle, \tag{B.70}$$

where δH is the perturbation Hamiltonian, namely the interaction $\tilde{H}_{\text{int}}$. This is given by

$$\begin{pmatrix} 0 & J & 0 \\ J & -J^2/4h^z_{\text{media}} & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{B.71}$$

By diagonalizing this matrix, we thereby obtain the three states in the form

$$\mathcal{U}_n|\uparrow_2\downarrow_3\downarrow_4\rangle + \mathcal{V}_n\left(|\downarrow_2\uparrow_3\downarrow_4\rangle + \frac{J}{-4h^z_{\text{media}}}|\downarrow_2\downarrow_3\uparrow_4\rangle\right) + \mathcal{W}_n|\uparrow_2\uparrow_3\uparrow_4\rangle, \tag{B.72}$$

where

$$\begin{aligned} &\{\mathcal{U}, \mathcal{V}, \mathcal{W}\} \\ \equiv&\left\{-\frac{1}{\sqrt{2}}\left(1 - \frac{J}{16h^z_{\text{media}}}\right), \frac{1}{\sqrt{2}}\left(1 + \frac{J}{16h^z_{\text{media}}}\right), 0\right\},\ \{0, 0, 1\}, \\ &\left\{\frac{1}{\sqrt{2}}\left(1 + \frac{J}{16h^z_{\text{media}}}\right), \frac{1}{\sqrt{2}}\left(1 - \frac{J}{16h^z_{\text{media}}}\right), 0\right\}, \end{aligned} \tag{B.73}$$

with the eigenvalues

$$\{\epsilon_2, \epsilon_3, \epsilon_4\} = \left\{-J - \frac{J^2}{8h^z_{\text{media}}}, 0, J - \frac{J^2}{8h^z_{\text{media}}}\right\}, \tag{B.74}$$

respectively.

Now, we can calculate the explicit forms of the eigenstates $\{|\phi_n\rangle\}^8_{n=1}$ and the corresponding eigenvalues $\{\epsilon_n\}_{n=1}^{8}$:

$$
\begin{aligned}
&|\phi_1\rangle = |{\uparrow_2}{\uparrow_3}{\downarrow_4}\rangle + \frac{J}{-4h^z_{\text{media}}}|{\uparrow_2}{\downarrow_3}{\uparrow_4}\rangle + \frac{J^2}{16h^{z2}_{\text{media}}}|{\downarrow_2}{\uparrow_3}{\uparrow_4}\rangle,\\
&\epsilon_1 = -3h^z_{\text{media}} + \frac{J^2}{-4h^z_{\text{media}}},\\
&|\phi_2\rangle = -\frac{1}{\sqrt{2}}\left(1 - \frac{J}{16h^z_{\text{media}}}\right)|{\uparrow_2}{\downarrow_3}{\downarrow_4}\rangle + \frac{1}{\sqrt{2}}\left(1 + \frac{J}{16h^z_{\text{media}}}\right)\left(|{\downarrow_2}{\uparrow_3}{\downarrow_4}\rangle + \frac{J}{-4h^z_{\text{media}}}|{\downarrow_2}{\downarrow_3}{\uparrow_4}\rangle\right),\\
&\epsilon_2 = -h^z_{\text{media}} - J - \frac{J^2}{8h^z_{\text{media}}},\\
&|\phi_3\rangle = |{\uparrow_2}{\uparrow_3}{\uparrow_4}\rangle,\ \epsilon_2 = -h^z_{\text{media}},\\
&|\phi_4\rangle = \frac{1}{\sqrt{2}}\left(1 - \frac{J}{16h^z_{\text{media}}}\right)|{\uparrow_2}{\downarrow_3}{\downarrow_4}\rangle + \frac{1}{\sqrt{2}}\left(1 + \frac{J}{16h^z_{\text{media}}}\right)\left(|{\downarrow_2}{\uparrow_3}{\downarrow_4}\rangle + \frac{J}{-4h^z_{\text{media}}}|{\downarrow_2}{\downarrow_3}{\uparrow_4}\rangle\right),\\
&\epsilon_4 = -h^z_{\text{media}} + J - \frac{J^2}{8h^z_{\text{media}}},\\
&|\phi_5\rangle = |{\downarrow_2}{\downarrow_3}{\downarrow_4}\rangle,\ \epsilon_5 = h^z_{\text{media}},\\
&|\phi_6\rangle = |{\uparrow_2}{\downarrow_3}{\uparrow_4}\rangle,\ \epsilon_6 = h^z_{\text{media}},\\
&|\phi_7\rangle = |{\downarrow_2}{\uparrow_3}{\uparrow_4}\rangle,\ \epsilon_7 = h^z_{\text{media}},\\
&|\phi_8\rangle = |{\downarrow_2}{\downarrow_3}{\uparrow_4}\rangle,\ \epsilon_8 = 3h^z_{\text{media}}.
\end{aligned}
\tag{B.75}
$$

Using Eqs. (B.63)–(B.65) and (B.75), we obtain the matrix elements $\{F_1, p_{\uparrow\uparrow}, p_{\downarrow\downarrow}\}$ to calculate Eq. (B.58):

$$
F_1 = \frac{J}{-2h_{1\text{op}}}\left(e^{\beta(3h^z_{\text{media}}+J^2/4h^z_{\text{media}})}\frac{J^2}{16(h^z_{\text{media}})^2} + e^{\beta(h^z_{\text{media}}+J^2/8h^z_{\text{media}})}\frac{J}{4h^z_{\text{media}}}\sinh\beta J\right) \tag{B.76}
$$

and

$$
\begin{aligned}
p_{\uparrow\uparrow} &= e^{\beta(3h^z_{\text{media}}+J^2/4h^z_{\text{media}})}\frac{J^2}{16(h^z_{\text{media}})^2} + e^{\beta h^z_{\text{media}}} + 2e^{-\beta h^z_{\text{media}}},\\
p_{\downarrow\downarrow} &= \frac{J^2}{4h^2_{1\text{op}}}\left(e^{\beta(h^z_{\text{media}}+J^2/8h^z_{\text{media}})}\cosh(\beta J) + \frac{J}{8h^z_{\text{media}}}e^{\beta(h^z_{\text{media}}+J^2/8h^z_{\text{media}})}\sinh(\beta J) + e^{-\beta h^z_{\text{media}}}\right).
\end{aligned}
\tag{B.77}
$$

Substituting these expressions into (B.58), we obtain the approximate phase boundary in the upper right area of the phase diagram in Fig. 3.6 (a).

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