The Phase Transition of Square Lattice Antiferromagnets at Finite Temperature

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The magnetic properties of the two-dimensional $J_1 - J_2$ model with both exchanges $J_1$ and $J_2$ being antiferromagnetic and a single-ion anisotropy at nonzero temperature are investigated. As $J_2/J_1 < 1/2$ ($> 1/2$), only the Néel (collinear) state exists. When $J_2/J_1 = 1/2$, both the Néel and collinear states can exist and have the same Néel temperature. The calculated free energies show that there can occur a phase transition between the two states below the Néel point when the single-ion anisotropy is strong enough. It is a first-order transition at nonzero temperature. It is possible that the doping in real materials can modify the ratio of $J_2/J_1$ to reach $1/2$ so as to implement the phase transition.

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A square lattice antiferromagnet can be described by the well-known two-dimensional (2D) $J_1 - J_2$ model. In this model both the nearest neighbor (nn) and next nearest neighbor (nnn) exchanges are antiferromagnetic (AF), so that it was believed a frustrated system. This model could be used to describe the structures in real materials. It was firstly related to the copper oxide monolayers in the Cu-based high temperature oxide superconductors[1], and then to magnetic planes in some other materials[2-6]. The most representative structure that could be well described by this model might be the Fe monolayers in the Fe-based superconductors La-O-Fe-As[7-13] and BaFe$_2$As$_2$[14]. Because of its importance, the $J_1 - J_2$ model has been carefully studied by various methods. However, the study has been mainly focused on its properties, especially its possible phase transition at 0K[15-17]. Investigations concerning nonzero temperature[18-24] have been comparatively much fewer, although the real materials are at finite temperature. Despite the already given physical results of the system by these investigations, there may be some interesting features still hidden at finite temperature.

Here we study the $J_1 - J_2$ model as a representative of such a Fe plane, focusing our attention on the properties at nonzero temperature. The physical quantities of the quantum model at finite temperature are calculated. A remarkable result is that when $J_2/J_1 = 1/2$, there may occur a phase transformation below Néel point $T_N$.

The AF Hamiltonian of a square lattice is $H = \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_i (\mathbf{S}^z_i)^2$. The first term is Heisenberg exchanges. Only the nn and nnn exchanges $J_1$ and $J_2$ are considered, both being positive. The second term presents a single-ion anisotropy. If a two-dimensional AFM system has no any anisotropy, there will be no spontaneous sublattice magnetizations in it[24,25]. It was indeed possible for the single-ion anisotropy to appear in real materials[26]. We term the exchanges $J_1$ and $J_2$ and anisotropy $D$ as Hamiltonian parameters. We let Boltzmann constant $k_B = 1$ so that all the quantities, including Hamiltonian parameters, temperature $T$, and sublattice magnetization $\langle S^z \rangle$, become dimensionless. $\langle S^z \rangle$ is the assembly thermostatical average of spin operator $S^z$. We fix $J_1 = 1$ and change $J_2$ value in computation. $D$ is assumed to be two or three orders of magnitude less than $J_1$. In the real La-O-Fe-As materials the spin quantum number might be larger than 1/2[27]. Therefore, the cases of some of the lowest spin quantum numbers are considered.

It was proposed that there might be four possible spin configurations[28,29], among which the two named as AF1 and AF2[30] had lower energies. They are depicted in Fig. 1, and were called as Néel state and collinear state, respectively. In either of the configurations, the lattice is divided into two sublattices. The spins within each sublattice are parallel to each other, and the spins of the two sublattices are antiparallel. The spin averages of the two sublattices are denoted as $\langle S^z_1 \rangle$ and $\langle S^z_2 \rangle$, respectively. Since there is no external field, $\langle S^z_1 \rangle = -\langle S^z_2 \rangle = \langle S^z \rangle$. We calculate the stable configurations by the many-body Green’s function method under random phase approximation[31]. According to our calculation results, when $J_2/J_1 < 1/2$, the stable state is AF1 configuration where the nn spins are antiparallel to each other, showing that the nn exchange is dominant. While for $J_2/J_1 > 1/2$, the stable state is AF2 where the nnn spins are antiparallel to each other. This conclusion holds at any temperature, for any $S$ and nonzero $D$ values. We also tested other ordered states including

![FIG. 1: (a) AF1 and (b) AF2 configurations.](image-url)
FIG. 2: $\langle S^z \rangle$ vs. $T$ curves for $S=1$ and $D=0.01$. The numbers labeling the curves are the $J_2$ values. When $J_2 < 0.5 (> 0.5)$, the state is AF1 (AF2). When $J_2 = 0.5$, both states exist.

the two suggested in Refs. [28,29] and none of them were stable at nonzero temperature.

In Fig. 2 we plot the curves of spin average $\langle S^z \rangle$ vs. $T$ at various $J_2$ values. In this and following figures, we always use the solid and dashed lines to represent the results of AF1 and AF2 configurations, respectively. The temperature at which $\langle S^z \rangle$ becomes zero is Néel point, denoted as $T_N$. Because we merely research the case of nonzero temperature, when we mention zero temperature, we actually mean the temperature very close to zero, which is denoted by $0^+$. To describe the dependence of the curves on $J_2$ value, we concentrate our attention to two physical quantities: Néel temperature $T_N$ and the spin average $\langle S^z \rangle$ at $0^+$K, hereafter denoted as $\langle S^z(0^+) \rangle$. As $J_2$ increases from zero to 0.5, both Néel temperature $T_N$ and $\langle S^z(0^+) \rangle$ decreases. At $J_2 = 0$, it is an ordinary nn AF exchange system, and there is no competition to cause frustration. As $J_2$ increases from 0, the competition between $J_2$ and $J_1$ emerges and becomes stronger. This results in the drop of both $T_N$ and $\langle S^z(0^+) \rangle$.

When $J_2 = 0.5$, the competition between $J_1$ and $J_2$ is the strongest. As $J_2$ rises from 0.5, the role of $J_2$ becomes more important and the competition becomes comparatively weaker. As a consequence, both $T_N$ and $\langle S^z(0^+) \rangle$ increase.

Figure 3 plots the results of $T_N$ as a function of $J_2$ at different $S$ and $D$ values. This figure is in fact a phase diagram that contains three phases: AF1, AF2 and paramagnetic (P) phases. A solid line is the border line between phases AF1 and P, and a dashed one is the border between AF2 and P. As $J_2/J_1$ approaches 1/2 from either side, the competition between $J_1$ and $J_2$ lowers $\langle S^z(0^+) \rangle$ value.

As $J_2 > 1$, the curves of $\langle S^z(0^+) \rangle$ vs. $J_2$ value are almost flat. This means that when $J_2$ is sufficiently large, it is predominant compared to $J_1$ value, so that the quantum fluctuation at $0^+$K caused by the competition between $J_1$ and $J_2$ is almost unchanged with the variation of $J_2$ value. Nevertheless, no matter how large the $J_2$ value is, $\langle S^z \rangle$ of AF2 is always smaller than that of AF1 when $J_2=0$. This is because when $J_2 > 0$, there is always a competition between $J_1$ and $J_2$. While in the case of $J_2=0$, there is no such a competition, and only the nn AF exchange plays a role.

FIG. 3: (a) Néel temperature vs. $J_2$ value. (b) Enlargement of the region around $J_2 = 0.5$. The lines are to guide eyes.

FIG. 4: (a) $\langle S^z(0^+) \rangle$ vs. $J_2$ value. (b) Enlargement of the region around $J_2 = 0.5$. The lines are to guide eyes.
The effect of the anisotropy $D$ value and spin quantum number $S$ is embodied in Figs. 3 and 4. As $D$ or $S$ value is smaller, $(S^z)$ value at any temperature is smaller, and Néel point $T_N$ is as well.

Now let us discuss the case of $J_2/J_1 = 1/2$. At first thought, in this case the competition between $J_1$ and $J_2$ are strongest so that the AF configurations may be totally frustrated and either AF configuration is difficult to hold. Indeed, researches[24] showed that as long as there was no single-ion anisotropy, both configurations could not exist at finite temperature when $J_2/J_1 = 1/2$. However, our calculation shows that for any nonzero $D$ value, both AF1 and AF2 states can exist as displayed by Figs. 2 to 4. In Fig. 2, the solid and dashed lines marked by '0.5' show that both the states can exist and have the same $T_N$ point under the parameter $J_2/J_1 = 1/2$. In Fig. 3, it is shown that both AF1 and AF2 reach the same Néel temperature when $J_2/J_1$ reaches 1/2 for various $S$ and $D$ values. Figure 4(b) reveals that both AF1 and AF2 have nonzero $(S^z(0^+))$ as $J_2/J_1$ reaches 1/2.

Since as $J_2/J_1$ goes to 1/2 from either side, $T_N$ reaches the same value, see Fig. 3(b), it is understood that the Néel point is uniquely determined by the Hamiltonian parameters, although there may be more than one state.

Figure 4(b) shows that when $J_2/J_1 = 1/2$, $(S^z(0^+))$ values of the two states are not the same, and that of AF2 is higher. This can be explained from the pictures in Fig. 1. In AF2 configuration, every spin is parallel to a half of its nn spins and antiparallel to another half, respectively, while in AF1, every spin is antiparallel to all of its nn spins. Thus the quantum fluctuation of AF2 at 0°K is stronger than AF1.

Since both configurations can exist, one may ask which one is stabler. At fixed temperature and volume, the state with lower free energy is stabler. The free energy can be evaluated numerically by means of the internal energy via $F(T) = E(0^+) = T \int_0^T E(T') - E(0^+) dT'$. Before calculating the free energy, one has to compute the internal energy which is defined as the thermostatistical average of Hamiltonian, $E = \langle H \rangle / N$, where $N$ is the site number in the 2-D plane. The correlation functions

$$\frac{1}{2} \sum_{i,j} J_{ij} S_i^+ S_j^-$$

involved in the energy are carefully calculated by use of the spectral theorem[32]. Figure 5 plots $E(T)$ for $S = 3/2$ and $D = 0.01$ and 0.05. $E(T)$ increases with temperature monotonically as it should be. In Fig. 5, $E_2(T) > E_1(T)$, but the internal energy cannot be used to determine which stable is stabler, since the entropies of the two states are different. The corresponding free energies are plotted in Fig. 5. $F(T)$ decreases monotonically with temperature. It is seen that $F_1(0^+) < F_2(0^+)$, which means that near zero temperature AF1 configuration is stabler. However, $F_2(T)$ drops faster than $F_1(T)$ and the two curves cross at a temperature, which means that above this temperature AF2 is stabler. Thus it is concluded that below $T_N$, an AF1-AF2 phase transformation may occur. By comparison of Figs. 5(a) and (b), it is seen that as the anisotropy $D$ is raised from 0.01 to 0.05, both $F_1(0^+)$ and $F_2(0^+)$ lower, the former decreasing more, and thus the AF1-AF2 phase transformation point rises.

In Fig. 6, we plot the free energy curves for $S = 1, 2, 5/2, 3$ each with two $D$ values. The common features in this figure and Fig. 5 are that $F_2(T)$ always decreases faster than $F_1(T)$, and as $D$ rises, both $F_1(0^+)$ and $F_2(0^+)$. drop, the former dropping more. In Fig. 6(a), where $S = 1$, for either $D$ value, $F_1(0^+)$ is much lower than $F_2(0^+)$, and up to $T_N$, the two curves do not cross. Therefore, in this case, there is no phase transformation below Néel point. As for $S = 3/2$, the AF1-AF2 transformation may occur, as having been revealed by Fig. 5. In Fig. 6(b), as $D = 0.01$, $F_1(0^+)$ is higher than $F_2(0^+)$, and up to $T_N$, the two curves do not cross, indicating AF2 being always stabler and lack of the phase transformation. While when $D$ is raised to 0.05, $F_1(0^+)$ drops to such a position that $F_1(0^+) < F_2(0^+)$, and the two curves $F_1(T)$ and $F_2(T)$ cross below $T_N$. Therefore, a phase transformation may occur. The analysis of Fig. 6(c) is similar to that of Fig. 6(b).

It is deduced from Figs. 5 and 6 that the condition for the AF1-AF2 phase transformation to occur is that the $D$ value should be large enough so that $F_1(0^+) < F_2(0^+)$. Otherwise, $F_1(0^+) > F_2(0^+)$ and there is no phase transformation, because $F_2(T)$ always decrease with temper-

![FIG. 5: The internal energies $E(T)$ (ascending curves) and free energies $F(T)$ (descending curves) at $J_2 = 0.5$ and $S = 3/2$. (a) $D = 0.01$ and (b) $D = 0.05$.](image1)

![FIG. 6: The free energies at $J_2 = 0.5$ and several $S$ and $D$ values. (a) $S = 1$, (b) $S = 2$ and (c) $S = 5/2$.](image2)
ature faster than \( F_1(T) \). In the case of \( S = 1 \), when the \( D \) value continues to increase, then both the solid and dashed lines lower, and if \( D \) is strong enough, it is expected that the two line will cross and the phase transition will occur.

It should be noted that both AF1 and AF2 are stable states but with different energies, so that it is a first-order transition. There is certainly an energy barrier between the two states. Unlike a classical system, the energy barrier in the present quantum system is difficult to reckon since it involves non-equilibrium states.

A question arises that how to actualized the AF1-AF2 phase transformation. For instance, in the case of Fig. 5(b), suppose that the system is initially under room temperature. When temperature is decreased to below Néel point, the state becomes AF2. As temperature reaches the AF1-AF2 transformation point, how the AF2 configuration can overcomes the barrier to get to AF1 configuration? We suggest that applying a strong impulsive magnetic field along the direction that is perpendicular to the spin direction can make the system to reach the stabler state.

Finally, we would like to briefly discuss the possibility of adjusting the ratio \( J_2/J_1 \) to become 1/2 in real materials. The results from the band structure calculation of LaFeAsO were that \( J_2/J_1 > 1/2 \)[27,29,30,33], thus the Fe planes in this crystal were in AF2 state. In this kind of materials both the nn and nnn exchanges between Fe atoms were mediated by As atoms[27]. Because the Fe 2-D plane was sandwiched by As atom monolayers, As atoms played a key bridge role in the indirect exchanges. Since in \( \text{AFeAsO} (\text{A}=\text{La, Ce, Pr, Nd, Sm}) \)[10,34] and \( \text{BFe2As2} (\text{B}=\text{Ca, Sr, Ba}) \)[35] the Fe-As sandwich structures were the same, the Fe planes in all these crystals were in AF2 states. While LaFePO, which had the same crystal structure as LaFeAsO except that As was replaced by P, exhibited paramagnetism in the normal conducting state[36]. This prompted us that the ability of the P atoms in the Fe-P sandwich structure to mediate the exchange interaction was rather weak. Based on this fact, we conjecture that the appropriate doping in As monolayers could modify the ratio \( J_2/J_1 \) value and possibly to reach 1/2. It therefore deserves to explore the new material to observe the expected phase transition.

In summary, we find that in a 2D AF system described by the \( J_1 - J_2 \) model, there can occur the phase transformation between the collinear and Néel states under the condition that \( J_2/J_1 = 1/2 \).

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