Abnormal magnetism and phase transformation of a Heisenberg-like model with internal spin fluctuation

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Magnetic states of the system described by a Heisenberg-like model with on-site spin fluctuation are studied. Two features are shown for such a system. One is that the system may reveal ferromagnetism or antiferromagnetism in the intermediate temperature range between zero and order-disorder transition temperature. The other is that there may occur a first-order transformation below order-disorder transition temperature. These are effects of on-site spin fluctuation.

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The Heisenberg exchange model is famous as it describes magnetic systems very well. In this model, there is one spin on each lattice site with no more complex on-site structure. Recently, the spin fluctuation in lattice sites has been noticed. For transition-metal elements, the 3*d* orbitals usually split into two energy levels, i.e., double degeneration e_g and triple degeneration t_g orbitals, under the interaction of crystal field. Xia *et al.*[1] thought that this maybe suggested that the total atom spin was composed of two spins S^d and S^t contributed by the double and the triple states. It was believed that spin fluctuation inside the atom was unavoidable. Based on this consideration, they suggested a model Hamiltonian to mimic the spin fluctuation inside an atom in transition-metal element materials. This was an extended Heisenberg Hamiltonian in which there are two subspins in each lattice site, which, hereafter, will be called two-spin-per-site Heisenberg Hamiltonian (TSPSHH). They studied the spontaneous magnetization and susceptibility of ferromagnetism described by such a Hamiltonian as a comparison with the usual Heisenberg Hamiltonian. Jiang *et al.*[2] added a uniaxial anisotropy term to study its effect on the system, especially the shift of the Curie point. Both of the works merely studied the ferromagnetic state. However, because of the existence of two subspins in one lattice site, the possible states can be very complicated and there may be some new physical behavior.

In usual Heisenberg Hamiltonian, the magnetic state is mainly determined by two factors: spin quantum number S and the exchange interaction J if only the nearest-neighboring exchange is considered. In the case that the spin quantum number S on every site is the same, the phase diagram is simple. The possible state is determined only by the relative orientation of neighboring spins, or the sign of J. Below order-disorder transition temperature, it is either ferromagnetic (or antiferromagnetic) depending on the J value in a homogeneous bulk material. In the system described by TSPSHH, we have more factors to be considered: the spin quantum number of the two subspins (S^d, S^t) in each site and four exchange interactions between them. The possible state is determined not only by the relative orientation of neighboring spins but also the relative orientation of the subspins in one site.

In this report, we will see two properties according to TSPSHH. One is that the system can show magnetism in a intermediate temperature range between zero and the order-disorder transition point. Another is that a transformation can occur below the order-disorder transition point.

Our model assumes a simple cubic lattice. Because our discussion involves an antiferromagnetic case, the lattice is divided into two sublattices. Assuming that S^d and S^t are subspin quantum numbers of the two states of one atom, respectively, the TSPSHH reads

$$H=-\sum_{(ia,jb)}(\mathbf{S}_{ia}^{d}\mathbf{S}_{ja}^{t})\begin{pmatrix}J_1 & J_3\\ J_3 & J_2\end{pmatrix}\begin{pmatrix}\mathbf{S}_{ib}^{d}\\ \mathbf{S}_{jb}^{t}\end{pmatrix}-\frac{1}{2}\sum_{ia}(\mathbf{S}_{ia}^{d}\mathbf{S}_{ia}^{t})\times\begin{pmatrix}0 & J_0\\ J_0 & 0\end{pmatrix}\begin{pmatrix}\mathbf{S}_{ia}^{d}\\ \mathbf{S}_{ia}^{t}\end{pmatrix}-\frac{1}{2}\sum_{ib}(\mathbf{S}_{ib}^{d}\mathbf{S}_{ib}^{t})\begin{pmatrix}0 & J_0\\ J_0 & 0\end{pmatrix}\begin{pmatrix}\mathbf{S}_{ib}^{d}\\ \mathbf{S}_{ib}^{t}\end{pmatrix}. \tag{1}$$

The first term is the Heisenberg exchange between the nearest-neighboring sites. The next two terms means the intra-atom exchange interaction between the two subspins. For convenience of the discussion below, we name J_1 and J_2 as the direct exchange between neighboring subspins $\mathbf{S}^d$ and $\mathbf{S}^t$, respectively, J_3 as the cross exchange, and J_0 as the on-site exchange. In Eq. (1), the subscripts a and b label the two sublattices and the summation (ia,jb) means that the summation is taken over all the nearest-neighbor pairs. The statistical averages of subspin operators are $\langle S_a^{dz}\rangle$, $\langle S_b^{dz}\rangle$, $\langle S_a^{tz}\rangle$, and $\langle S_b^{tz}\rangle$, respectively. The magnetizations of the two sublattices are $\langle S_a^z\rangle=\langle S_a^{dz}\rangle+\langle S_a^{tz}\rangle$ and $\langle S_b^z\rangle=\langle S_b^{dz}\rangle+\langle S_b^{tz}\rangle$. We let exchange parameters be dimensionless as in Refs. 1 and 2.

The method we use is the many-body Green's-function theory which has long been used to treat the Heisenberg model.[1,3,4] The retarded Green's functions are, according to Bogolyubov and Tyablikov,[5] as follows:

$$G_{ij}(t-t')=\langle\langle A_i;B_j\rangle\rangle=-i\theta(t-t')\langle A_iB_j-B_jA_i\rangle, \quad (2)$$

where the subscripts i and j label lattice sites. There is a well-known spectral theorem[3,6] to allow one to calculate the statistical average of the product of the operators $\langle B_jA_i\rangle$. In this report the operator A is taken as $A=S_a^{d+}$, S_b^{d+}, S_a^{t+}, S_b^{t+}, respectively. If we denote $(S_a^{d+}, S_b^{d+}, S_a^{t+}, S_b^{t+})=(S_1^+, S_2^+, S_3^+, S_4^+)$, or simply $A=S_\alpha^+$, $\alpha=1, 2, 3, 4$, then by the equation of motion of the Green's function, the statistical average of spin operators $\langle S_\alpha^z\rangle$ can be evaluated by the following formula:[7,4]

$$\langle S_\alpha^z\rangle=\frac{(S_\alpha-R_\alpha)(1+R_\alpha)^{2S_\alpha+1}+(1+S_\alpha+R_\alpha)R_\alpha^{2S_\alpha+1}}{(1+R_\alpha)^{2S_\alpha+1}-R_\alpha^{2S_\alpha+1}},$$

$$\alpha=1, 2, 3, 4, \quad (3)$$

where we denote spin averages $(\langle S_a^{dz}\rangle, \langle S_b^{dz}\rangle, \langle S_a^{tz}\rangle, \langle S_b^{tz}\rangle)=(\langle S_1^z\rangle, \langle S_2^z\rangle, \langle S_3^z\rangle, \langle S_4^z\rangle)$ and the subspin quantum numbers $(S_a^d, S_b^d, S_a^t, S_b^t)=(S_1, S_2, S_3, S_4)$, respectively. In derivation, the operator B in Eq. (2) is chosen as $B_\beta=(S_\beta^z)^nS_\beta^-$, $\beta=1,2,3,4$. Because the lattice is infinitely large and has translational invariance, the Green's function is Fourier transformed from real space to **k** space. Then, by standard derivation procedure of the Green's-function method, a secular equation can be obtained: $\Sigma_\mu(\omega_\tau\delta_{\nu\mu}-P_{\nu\mu})U_{\mu\tau}=0$. The order of matrix P is four, which is due to the fact that every crystal cell contains two sites a and b with each having S^d and S^t two subspins. ω_τ and U are the eigenvalues and eigenvectors of the matrix **P**. Thus, the R_α in Eq. (3) is given by

$$R_\alpha=\frac{1}{N}\sum_{\mathbf{k}}\sum_\tau\frac{U_{\alpha\tau}U_{\tau\alpha}^{-1}}{e^{\beta\omega_\tau}-1}, \quad \alpha=1, 2, 3, 4, \quad (4)$$

where U^{-1} is the inverse matrix of U. The summation of **k** means to do integration in the first Brillouin zone. Equations (3) and (4) are self-consistent equations for the calculation of magnetizations. In this report, the subspin quantum numbers for the two sublattices are taken as the same, that is to say, $S_a^d=S_b^d=S^d$ and $S_a^t=S_b^t=S^t$.

By physical intuition, all the possible states are posted in Fig. 1. States *A* and *B* are ferromagnetic. States *C* and *D* are antiferromagnetic. States *E* is called as mixed states. Here the states mean the situations under order-disorder transition temperature which is the Curie point or Néel point. The transition temperature is determined by spin values and exchange parameters. Generally speaking, larger subspin values and stronger exchanges lead to higher order-disorder transition temperature.

If $J_1>0$, $J_2>0$, and $J_3=J_0=0$, then $\mathbf{S}_a^d$ is parallel to $\mathbf{S}_b^d$ and $\mathbf{S}_a^t$ is parallel to $\mathbf{S}_b^t$. The system can be either state *A* or *B*. It will be state *A* if both J_3 and J_0 are positive, and it will be state *B* if both J_3 and J_0 are negative. Now suppose that the signs of J_3 and J_0 are contrary, the state will be determined by the competition of the strength of these two parameters. This competition is also influenced by thermal motion.

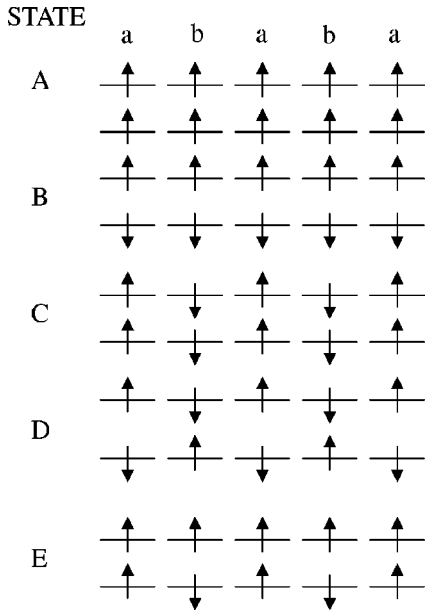

FIG. 1. Possible states. In each state, one line represents subspin S^d, and the other represents subspin S^t. The length of the arrows does not mean the dimension of the magnetization. The letters *a* and *b* label the two sublattices.

In state *B*, the neighboring subspin is parallel, while the on-site subspins are antiparallel. When $S^d=S^t$, $J_1=J_2$, the total magnetization is zero and the system will not exhibit magnetism at any temperature, as shown by the solid lines in Fig. 2. However, if $J_1\neq J_2$, the system will still be ferromagnetic, see the dashed lines in Fig. 2. At zero temperature, there is no thermal motion, the subspin orientations are uniquely determined by exchange interactions. As a result, the on-site subspins are antiparallel to each other and the system will not exhibit magnetism. As temperature ascends, the subspin with weaker direct exchange is more easily affected by thermal motion so that the subspin with weaker direct exchange has smaller magnetization. Therefore, the net magnetization is not zero and the system manifests ferromagnetism until the Curie point. Here we see a case that the system exhibits ferromagnetism in the intermediate range between zero temperature and the Curie point. Or in other words, the total magnetization decreases with decreasing temperature. This character is not possessed by the one-spin-per-site Heisenberg Hamiltonian system.

For states *A* and *B*, both direct exchanges are positive so that they are ferromagnetic. The condition for the system to be antiferromagnetic is that at least one of the direct exchanges must be negative. If both J_1 and J_2 are negative and $J_3=J_0=0$, then S_a^d is antiparallel to S_b^d and S_a^t is antiparallel to S_b^t. The system can be either state *C* or *D*. As the cross exchange and on-site exchange become nonzero, the competition of the two parameters determine the state of the system.

For state *D* when $S^d=S^t$, $J_1=J_2$, it is easily understood

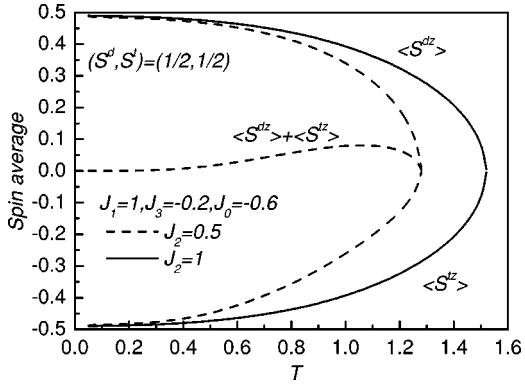

FIG. 2. Spin averages vs temperature with parameters $(S^d,S^t)=(1/2,1/2)$, $J_1=1$, $J_3=-0.2$, and $J_0=-0.6$. The state is B so that the two sublattices are identical. $\langle S^{dz}\rangle>0$ and $\langle S^{tz}\rangle<0$. For the solid lines: $J_2=1$. Because $J_1=J_2$, the two sublattices are identical and the total magnetization $\langle S^z\rangle=0$. While in the case presented by dashed lines, $J_2=0.5$. Because $J_1\neq J_2$, the system is ferromagnetic in the intermediate range.

that $\langle S_a^{dz}\rangle=-\langle S_a^{tz}\rangle$ and $\langle S_b^{tz}\rangle=-\langle S_b^{dz}\rangle$ so that $\langle S_a^z\rangle=\langle S_b^z\rangle=0$. The total magnetization is zero and the system will not exhibit magnetism at any temperature. However, if $J_1\neq J_2$, the system will still be antiferromagnetic, see Fig. 3. At zero temperature, the two subspins are antiparallel to each other and the total magnetism at each site is zero. As temperature increases, due to the influence of thermal motion, the absolute values of the statistical averages of the two subspins in one site are not the same. Therefore, the net magnetization of each sublattice is not zero and the system manifests antiferromagnetism until the Néel point. Thus, there can occur a case that the system exhibits antiferromagnetism in the intermediate range between zero temperature and the Néel point.

If the two direct exchanges have contrast signs, the system is of state E. For instance, if $J_1>0$, $J_2<0$, and $J_3=J_0=0$, then S_a^d is parallel to S_b^d and S_a^t is antiparallel to S_b^t. When cross exchange and on-site exchange become nonzero, the situation become complicated, because any of them

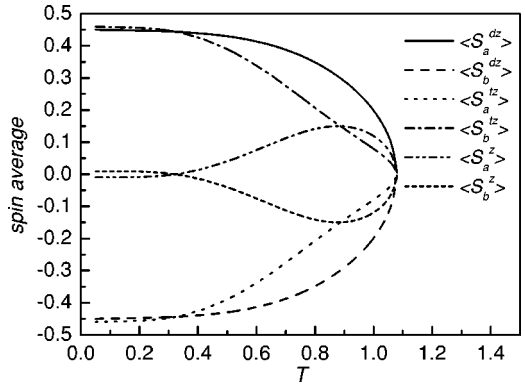

FIG. 3. Spin averages vs temperature of state D with parameters $(S^d,S^t)=(1/2,1/2)$, $J_1=-1$, $J_2=-0.5$, and $J_3=0.2$ and $J_0=0.6$. Because $J_1\neq J_2$, $\langle S^{dz}\rangle\neq-\langle S^{tz}\rangle$ and the system is antiferromagnetic in the intermediate temperature range. If $J_2=-1$, then because $J_1=J_2$, $\langle S^{dz}\rangle=-\langle S^{tz}\rangle$ and the total magnetization of each sublattice is zero (not shown).

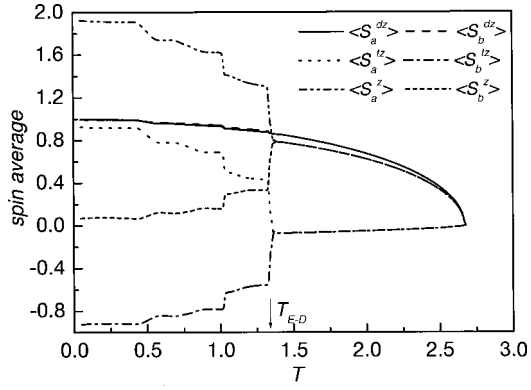

FIG. 4. Spin averages vs temperature to show $E-B$ transformation with parameters $(S^d,S^t)=(1,1)$, $J_1=1$, $J_2=-0.5$, $J_3=0$, and $J_0=-0.5$. At $E-B$ transformation temperature, $T_{E-B}=1.34$, the magnetization has a drop, indicating a first-order transformation.

has a contrary effect at the configurations of S^d and S^t. If J_3 or J_0 is beneficial for the configuration of S^d, then it will be against the configuration of neighboring S^t, and vice versa. Therefore, the configuration of state E may change unless J_3 and J_0 are vanishing. In this sense, the state E is not a stable one. As the temperature departs from zero, all states $A, B, C, D,$ and E are possible depending on the parameters of the subspin quantum number and four exchanges. For some parameters, the state turns to $A, B, C,$ or D even at zero temperature although the signs of J_1 and J_2 are contrary. Hence, that the two direct exchanges hold contrary signs is the necessary but not sufficient condition of state E. Here we stress the cases that a transformation may occur below order-disorder transition temperature.

In Fig. 4, we show $E-B$ transformation. The parameters are $(S^d,S^t)=(1,1)$, $J_1=1$, $J_2=-0.5$, $J_3=0$, and $J_0=-0.5$. That $J_1=1$ keeps the subspins S_a^{dz} and S_b^{dz} parallel to each other and that $J_0=-0.5$ tends to make the subspins in one site antiparallel to each other. If $J_2=0$, it would be state B. Now $J_2=-0.5$, which makes S_a^{tz} and S_b^{tz} antiparallel. Supposing that S_a^{dz} and S_b^{dz} point up and S_b^{tz} points down, let us see how the orientation of S_a^{tz} is determined. We can qualitatively discuss it by means of the concept of the molecular field (MF). It can be recognized that S_a^t is affected by two MF's: One is from its neighboring S_b^t, $6J_2\langle S_b^{tz}\rangle$, called nearest-neighbor MF (NNMF) and the other is from the subspin S_a^d at the same site, $J_0\langle S_b^{tz}\rangle$, called on-site MF (OSMF). Here we note that there are six nearest neighbors in simple cubic and $J_2=J_0$. At zero temperature, all the z-component averages of subspins are near to one, see Fig. 4. The S_a^t is affected by six nearest-neighbor S_b^t and only one S_a^d. Therefore, NNMF prevails and S_a^{tz} should point up, i.e., it is the state E. Because $|J_2|<J_1$, the averages $\langle S_a^{tz}\rangle$ and $|\langle S_b^{tz}\rangle|$ decrease with temperature more rapidly than $\langle S_a^{dz}\rangle$ and $\langle S_b^{dz}\rangle$, see dotted and dash-dotted lines in Fig. 4. At temperature T_{E-B}, $\langle S_b^{tz}\rangle$ is weak enough while $\langle S_a^{dz}\rangle$ is still large (about 0.86). Therefore, the effect OSMF become dominant so that the subspin flip down. This is a transformation from state E to state B. At the transformation point, the average of the flipped subspin $\langle S_a^{tz}\rangle$, as well as its neighboring $\langle S_b^{tz}\rangle$,

has a sudden drop, and after the transformation, the combined effect of J_2, J_0, and thermal motion keeps them rather weak. The averages of subspins $\langle S^{dz} \rangle$, are not influenced by the transformation. This is a first-order transformation because the total magnetization is discontinuous.

If there are two contradictory factors acted on spins, a transition may take place. The discussion above provides an example. Another example is the magnetization reorientation transition of ferromagnetic ultrathin films.[8–12] It is believed that the transition occurs because of the competition between single-ion anisotropy and dipole interaction.[13–15] The former tends to make spins perpendicular to, while the latter tends to make the spins parallel to, the film plane.

Calculation shows that all four kinds of $E-X$ transformation, where X represents A, B, C, or D states, can occur if we select appropriate parameters. The features are quite similar to Fig. 4. They are outlined as follows. The spin average of subspin with stronger direct exchange is not influenced. Only the subspin with weaker direct exchange flips. At the transformation point, the average of the flipped subspin, as well as its neighbor, has a sudden drop, and after the transformation they are rather weak. The transformation belongs to the first-order one because the magnetization, as a order parameter, is discontinuous at transformation temperature. Calculation shows that the larger the subspin values or the stronger the exchanges, the higher the transformation temperature T_{E-X} will be. The reason is that the subspin has a stronger power against thermal motion if it has a larger direct exchange or a larger spin quantum number.

In summary, we have seen two features of the system described by TSPSHH. One is that the system may reveal ferromagnetism or antiferromagnetism in the intermediate temperature range between zero and order-disorder transition temperature. The other is that there may occur a first-order transformation below order-disorder transition temperature. These are two believed properties of magnetic systems and are effects of intra-atom spin fluctuation.

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