The single-ion anisotropy in LaFeAsO

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Abstract. We use Green’s function method to study the Heisenberg model of LaFeAsO with the striped antiferromagnetic collinear spin structure. In addition to the intra-layer spin couplings $J_{1a}, J_{1b}, J_2$ and the inter-layer coupling $J_c$, we further consider the contributions of the single-ion anisotropy $J_s$. The analytical expressions for the magnetic phase-transition temperature $T_N$ and the spin spectrum gap $\Delta$ are obtained. According to the experimental temperature $T_N = 138K$ and the previous estimations of the coupling interactions, we make a further discussion about the magnitude and the effects of the single-ion anisotropy $J_s$. We find that the magnitudes of $J_s$ and $J_c$ can compete. The dependences of the transition temperature $T_N$, the zero-temperature average spin and the spin spectrum gap on the single-ion anisotropy are investigated. We find they both increase as $J_s$ increases. The spin spectrum gap at low temperature $T \to 0$ is calculated as a function of $J_s$, the result of which is a useful reference for the future experimental researches.
1. Introduction

It was recently discovered that an iron-based material LaFeAsO shows high-temperature superconductivity when O atoms are partially substituted by F atoms[1]. This discovery has triggered great research interest on the FeAs-based pnictides superconductors and their undoped compounds. It has been theoretically and experimentally confirmed that these pure FeAs-based compounds have a ground state with collinear stripe-like antiferromagnetic(AF) spin order formed by Fe atoms[2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. Thus to establish an effective spin Hamiltonian for them and to elucidate the corresponding antiferromagnetism are helpful in understanding the underlying mechanism to make them superconducting upon doping.

For undoped LaFeAsO and other similar parent compounds, a Heisenberg exchange model was suggested to explain their AF structure[2, 4, 12, 13], and was used to explore their magnetic properties[14, 15, 16]. Figure 1 shows the unit cell of the orthorhombic AF spin structure of the Fe lattice. This orthorhombic structure exists below a structure transition temperature $T_S$, which is $15 \sim 20$K higher than the magnetic transition temperature $T_N$[6, 17]. Usually the nearest neighbor (NN) coupling $J_1$(including $J_{1a}$ and $J_{1b}$), and the next-nearest neighbor (NNN) coupling $J_2$ in FeAs layers are dominant and must be considered. The NN coupling $J_c$ between spins on neighboring layers is regarded to be much smaller than the in-plane couplings[2, 4]. However, $J_c$ was found to be essential for the existence of a non-zero magnetic transition temperature $T_N$[15]. A further consideration can include the single-ion anisotropy $J_s$. It was estimated to be even much smaller than $J_c$ in the model of SrFe$_2$As$_2$, but a spin spectrum gap was found to be produced by it[14]. For LaFeAsO, so far there is no research report about the magnitude or the effects of the single-ion anisotropy.

In this paper, we use Green’s function method[18] to study the Heisenberg model of FeAs-based pure parent compounds. The Hamiltonian of this model in a detailed form is

$$H = \frac{1}{2}J_{1a} \sum_{\langle ij \rangle} S_{1i} \cdot S_{1j} + \frac{1}{2}J_{1a} \sum_{\langle ij \rangle} S_{2i} \cdot S_{2j} + J_{1b} \sum_{\langle ij \rangle} S_{1i} \cdot S_{2j}$$

$$+ J_2 \sum_{\langle \langle ij \rangle \rangle} S_{1i} \cdot S_{2j} + J_c \sum_{\langle ij \rangle} S_{1i} \cdot S_{2j} - J_s \sum_{i} [(S_{1z}^2 + S_{2z}^2)]^2,\quad (1)$$

where the spin coupling $J_c$ between layers and the single-ion anisotropy $J_s$ are both considered. The subscripts 1 and 2 mean the sublattices 1 and 2 respectively. $\langle ij \rangle$ means NN spin pairs, and $\langle \langle ij \rangle \rangle$ means NNN spin pairs. The self-consistent equations for the average sublattice spin will be derived. An analytical expression for the magnetic transition temperature $T_N$ will be obtained. For LaFeAsO, according to the recent estimations of the strengths $J_{1a}, J_{1b}, J_2$ and $J_c$[15] with the experimental temperature $T_N = 138$K[6, 17], we shall make a further estimation of the single-ion anisotropy $J_s$. We find that the magnitude of $J_s$ can compete with $J_c$, and in some situations even bigger than $J_c$. The effects of the single-ion anisotropy on the transition temperature $T_N$, the zero-temperature average spin $\langle S_z \rangle_0$ and the spin spectrum gap are investigated. We
find they all increase as \( J \) increases. In section 2 we shall give our analytical results derived from the Green’s function method. In section 3 we shall present our numerical results. Finally a conclusion is given in section 4.

2. Green’s function derivation

According to the general scheme of Green’s function method to solve an antiferromagnetic spin model with two sublattices, we construct the following Green’s functions:

\[
G_{1k,1l}(\omega) = \langle \langle S^+_1; S^-_l \rangle \rangle, \quad G_{2k,1l}(\omega) = \langle \langle S^+_2; S^-_l \rangle \rangle.
\]  

(2)

The equation of motion is

\[
\omega \langle \langle A; S^-_l \rangle \rangle = \langle [A, S^-_l]; S^-_l \rangle + \langle \langle [A, H]; S^-_l \rangle \rangle,
\]  

(3)

where \( A \) represents the spin operator \( S^+_k \) or \( S^+_k \). The commutator \([A, H]\) can be derived using Hamiltonian (1) and the basic commutation relations of spin operators:

\[
[S^z_i, S^\pm_j] = \mp S^\pm_i \delta_{ij}, \quad [S^\pm_i, S^z_j] = \mp S^\pm_j \delta_{ij}, \quad \text{where} \quad S^\pm_i = S^x_i \pm iS^y_i.
\]

In order to close the system of equations, the so-called PRA or Tyablikov decoupling[18] is adopted for the terms stemming from the exchange couplings:

\[
\langle \langle S^-_i S^+_j; S^-_l \rangle \rangle \approx \langle S^-_i \rangle \langle \langle S^+_j; S^-_l \rangle \rangle, \quad i \neq j.
\]  

(4)

While for the terms stemming from the single-ion anisotropy, we adopt the Anderson-Callen(AC) decoupling[19]:

\[
\langle \langle S^z_i S^+_j + S^+_i S^z_j; S^-_l \rangle \rangle \approx 2 \langle S^z_i \rangle \Theta^{(z)}_i \langle \langle S^+_j; S^-_l \rangle \rangle,
\]  

(5)

where

\[
\Theta^{(z)}_i = 1 - \frac{1}{2S^2}[S(S+1) - \langle S^z_i S^z_i \rangle].
\]  

(6)

The AC decoupling has been demonstrated to be most adequate for the single-ion anisotropy much small compared to the exchange interactions[20, 21].

Figure 1. A unit cell of the orthorhombic Fe spin lattice. \( \mathbf{a}, \mathbf{b}, \mathbf{c} \) are the three base vectors. The lattice consists of two sublattices (distinguished by the color gray and black).
In order to write the decoupled equations of motion in the $k$ space, we take the following Fourier transformation:

$$G_{k,i}(\omega) = \frac{1}{N} \sum_k G(k, \omega) e^{i k (R_k - R_i)},$$

where $N$ is the number of sites in either sublattice, and the summation over $k$ is restricted to the first Brillouin zone of the sublattice. At the same time, the equation $\delta_{ij} = \frac{1}{N} \sum_k e^{i k (R_k - R_j)}$ is also used.

Because of the translation invariant, we have $\langle S^z_{1k} \rangle = \langle S^z \rangle$, $\langle S^z_{2k} \rangle = -\langle S^z \rangle$ and $\Theta^{(z)}_{1k} = \Theta^{(z)}_{2k} = \Theta^{(z)} = 1 - \frac{1}{2\pi^2} [S(S+1) - \langle S^z S^z \rangle]$. Finally, we obtain the decoupled equations of the two Green’s function in $k$ space:

$$[\omega - \langle S^z \rangle A_k]G_{11}(k, \omega) - \langle S^z \rangle B_k G_{21}(k, \omega) = 2\langle S^z \rangle,$$

and

$$[\omega + \langle S^z \rangle A_k]G_{21}(k, \omega) + \langle S^z \rangle B_k G_{11}(k, \omega) = 0,$$

where

$$A_k = 2J_{1a} \cos(k_x a) - 2J_{1a} + 2J_{1b} + 4J_2 + 2J_c + 2J_s \Theta^{(z)},$$

and

$$B_k = 2J_{1b} \cos(k_y b) + 4J_2 \cos(k_x a) \cos(k_y b) + 2J_c \cos(k_z c),$$

in which $a, b, c$ are the three lattice constants. Solving equations (8) and (9), we obtain the Green’s function:

$$G_{11}(k, \omega) = \frac{\langle S^z \rangle}{\omega_k} \left[ \frac{A_k \langle S^z \rangle + \omega_k}{\omega - \omega_k} - \frac{A_k \langle S^z \rangle - \omega_k}{\omega + \omega_k} \right],$$

and the spin spectrum:

$$\omega_k = \langle S^z \rangle \sqrt{A^2_k - B^2_k}.$$  

When $k \to 0$, we obtain an expression for the spectrum gap:

$$\Delta = 2\langle S^z \rangle \sqrt{J_s \Theta^{(z)} [2J_{1b} + 4J_2 + 2J_c + J_s \Theta^{(z)}]},$$

which is similar with the expression given in ref[14] derived from the spin-wave theory, expect for the factor $\Theta^{(z)}$. From this expression for the gap, one see that the single-ion anisotropy is essential for the existence of the spectrum gap.

Then following the process of solving the average spin, we derive the correlation function $\langle S^- S^+ \rangle$ using the spectrum theorem:

$$\langle S^- S^+ \rangle = -\frac{1}{N \pi} \sum_k \int_{-\infty}^{\infty} d\omega \frac{\text{Im}G_{11}(k, \omega + i\epsilon)}{e^{\beta \omega} - 1}$$

$$= \frac{\langle S^z \rangle}{N} \sum_k \left[ \frac{A_k}{\sqrt{A^2_k - B^2_k}} \coth \frac{\beta \omega_k}{2} - 1 \right],$$

in which the equation $\frac{1}{x + i\epsilon} = P\left(\frac{1}{x}\right) - i\pi \delta(x)(P(\cdots)$ means taking the principle value) has been used to obtain the imaginary part of $G_{11}(\omega + i\epsilon)$, and $\beta = \frac{1}{k_B T}$, $k_B$ is the Boltzmann constant, $T$ is the temperature.
According to the theory of Callen\cite{22}, the average spin for arbitrary $S$ can be calculated using the following equation:

$$\langle S^z \rangle = \frac{(S - \Phi)(1 + \Phi)^{2S+1} + (S + 1 + \Phi)\Phi^{2S+1}}{(1 + \Phi)^{2S+1} - \Phi^{2S+1}},$$  \hspace{1cm} (16)$$

where

$$\Phi = \frac{\langle S^- S^+ \rangle}{2\langle S^z \rangle} = \frac{1}{2N} \sum_k \left[ \frac{A_k}{\sqrt{A_k^2 - B_k^2}} \coth \frac{\beta \omega_k}{2} - 1 \right].$$  \hspace{1cm} (17)$$

On the other hand, the correlation function $\langle S^z S^z \rangle$ can be calculated from the equation $\langle S^z S^z \rangle = S(S + 1) - (1 + 2\Phi)\langle S^z \rangle$. Using equation (16), we can relate $\Theta^{(z)}$ to $\Phi$ by

$$\Theta^{(z)} = 1 - \frac{\langle S^z \rangle}{2S^2}(1 + 2\Phi).$$  \hspace{1cm} (18)$$

Now the equations (16)(17)(18) can be solved self-consistently to obtain the average spin at any given temperature, provided we know the values of the exchange couplings $J_{1a}, J_{1b}, J_2, J_c$ and the single-ion anisotropy $J_s$.

When the temperature $T$ approaches zero, we obtain $\coth(\frac{\beta \omega_k}{2}) \to 1$. The equation (17) is reduced to

$$\Phi|_{T \to 0} = \frac{1}{2N} \sum_k \left[ \frac{A_k}{\sqrt{A_k^2 - B_k^2}} \coth \frac{\beta \omega_k}{2} - 1 \right].$$  \hspace{1cm} (19)$$

The zero-temperature average spin $\langle S^z \rangle_0$ can be obtained by self-consistently solving the equations (16)(18)(19).

When the temperature $T$ approaches the magnetic transition temperature $T_N$, the average spin $\langle S^z \rangle$ as well as the spectrum $\omega_k$ will approach zero. Expanding $\coth(\frac{\beta \omega_k}{2})$ in the equation (17), we obtain

$$\Phi|_{T \to T_N} \approx \frac{\Gamma}{\beta \langle S^z \rangle} - \frac{1}{2},$$  \hspace{1cm} (20)$$

where $\Gamma = \frac{1}{N} \sum_k \frac{A_k}{A_k^2 - B_k^2}$. Inserting (20) into (16), and expanding the terms in the denominator and the numerator as the series of $\langle S^z \rangle$, we finally derive

$$\langle S^z \rangle \approx \sqrt{\frac{12(\Gamma k_BT_N)^2}{S(2S - 1)}} \left(1 - \frac{T}{T_N}\right),$$  \hspace{1cm} (21)$$

where

$$T_N = \frac{S(S + 1)}{3k_B\Gamma}.$$  \hspace{1cm} (22)$$

On the other hand, inserting (20) into (18), and using the equation (22), we obtain the reduced expression for $\Theta^{(z)}$ near the temperature $T_N$:

$$\Theta^{(z)}|_{T \to T_N} \approx \frac{2S - 1}{3S}.$$  \hspace{1cm} (23)$$
Figure 2. The transition temperature $T_N$ as a function of the single-ion anisotropy $J_s$ for $J_{1a} = 0.98$, $J_2 = 0.52$ and $J_c = 0.0004$ in the unit of $J_{1b} = 50$ meV. The black point correspond to the experimental temperature $T_N = 0.238$ in the unit of $J_{1b}/k_B$.

3. Numerical results and discussions

So far there is no consensus on the magnitudes of the exchange couplings $J_{1a}$, $J_{1b}$, $J_2$ and $J_c$, because of the unclear microscopic origin of the observed AF spin structure. Here we prefer the estimations in ref[15], which gave $J_{1b} = 50 \pm 10$ meV, $J_{1a} = 49 \pm 10$ meV, $J_2 = 26 \pm 5$ meV and $J_c = 0.020 \pm 0.015$ meV by using the experimental transition temperature $T_N = 138K$ of pure LaFeAsO. The main purpose of this paper is to investigate the magnitude and the effects of the single-ion anisotropy $J_s$ in LaFeAsO. Through out our numerical calculation, we take $J_{1b} = 50$ meV, $J_{1a} = 49$ meV, $J_2 = 26$ meV and the spin $S = 1$. The result $J_{1b} \sim 50$meV are obtained from the first-principle calculating[4, 23]. In the present systems of units, the Boltzmann constant is taken as $k_B = 0.086$ meV/K.

Figure 2 shows the effect of the single-ion anisotropy $J_s$ on the transition temperature $T_N$. We can see that $T_N$ increases as $J_s$ increases. This means that the single-ion anisotropy term is in favor of the AF spin structure. It can be understood from the expression of the single-ion anisotropy term in the Hamiltonian (11). Increasing the magnitude of $J_s$ will make the spins incline to align along the $z$ axis, and give a lower total energy, which make the system more stable. To one’s surprise, the magnitude of $J_s$ corresponding to the experimental transition temperature $T_N$ is about 0.00143$J_{1b}$, which is much bigger than the magnitude of the exchange coupling $J_c = 0.0004J_{1b}$ estimated in ref[15]. Furthermore, we see from figure 3 that the variation range of $\langle S^z \rangle_0$ with $J_s$ varying from 0 to 0.1 is almost the same as the one produced by $J_c$ in ref[15]. All these results imply that the magnitude of $J_s$ is probably not much small compared with $J_c$. So the estimation of $J_c$ maybe need to be adjusted if the single-ion anisotropy term is considered. As to the estimations of the other exchange couplings $J_{1a}$, $J_{1b}$ and $J_2$, we think there are still reasonable.
Figure 3. The zero-temperature average spin as a function of the single-ion anisotropy $J_s$ for $J_{1a} = 0.98$, $J_2 = 0.52$ and $J_c = 0.0004$ in the unit of $J_{1b} = 50$ meV.

Figure 4. The competing relation of $J_c$ and $J_s$. The curve is depicted by using the equation (22) for $T_N = 0.238(J_{1b}/k_B)$, $J_{1a} = 0.98J_{1b}$ and $J_2 = 0.52J_{1b}$.

Figure 4 shows the competing relation of $J_c$ and $J_s$ when the transition temperature is fixed at the experimental value. The increase of $J_c$ is accompanied by the decrease of $J_s$, and vice versa. From figure 4 we can see that the ranges of their corresponding variations are at the same magnitude, which implies they probably have the same status in the viewpoint of theoretical study. As to revealing the actual magnitudes of the two parameters $J_c$ and $J_s$, we think it is not enough to use only the experimental transition temperature $T_N$.

Figure 5 shows the effect of the single-ion anisotropy $J_s$ on the spectrum gap at low temperature. The gap vanishes as $J_s$ vanishes, and increases as $J_s$ increases. We find the effect of $J_c$ on the gap is very trivial. The curves for different values of $J_c$ between $(0.0001, 0.01)$ are almost the same, while the single-ion anisotropy affects the gap apparently. Considering the gap can be obtained from inelastic neutron-scattering
Figure 5. The spin spectrum gaps at low temperature $T \to 0$ as functions of the single-ion anisotropy $J_s$ for $J_{1a} = 0.98J_{1b}$, $J_2 = 0.52J_{1b}$ and $J_c = 0.0001J_{1b}$ (the below curve), $J_c = 0.01J_{1b}$ (the above curve).

experiment\[14\], we suggest that the magnitude of the single-ion anisotropy be estimated from the future experimental results of the spectrum gap. For example, if $J_c = 0.0004J_{1b}$, we obtain $J_s = 0.00143J_{1b}$ from the experimental transition temperature $T_N = 138$K. Then calculating the spectrum gap with $J_s = 0.00143J_{1b}$, we obtain the magnitude of the gap $\Delta \approx 3.4$ meV, which can be compared with the future experimental result.

4. Conclusion

We use Green’s function method to study the Heisenberg model (1) of LaFeAsO with the striped AF spin structure as shown in figure 1. The main purpose of this paper is to investigate the magnitude and the effects of the single-ion anisotropy $J_s$. We derive the self-consistent equations for the average spin, and obtained the analytical expressions for the spin spectrum gap $\Delta$, and the magnetic transition temperature $T_N$. We find that the transition temperature $T_N$, the zero-temperature average spin $\langle S^z \rangle_0$ and the spin spectrum gap $\Delta$ are all increasing functions of the single-ion anisotropy $J_s$. From our numerical results by using $T_N = 138$K and the previous estimations of $J_{1a}, J_{1b}, J_2$ and $J_c$ in ref[15], we find that the magnitude of $J_s$ is probably not much small compared with $J_c$. Because the single-ion anisotropy is essential for the existence of the spin spectrum gap, we suggest using the experimental result of the spin spectrum gap to fix the magnitude of the single-ion anisotropy $J_s$ in the future.

References

[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Yildirim T 2008 Phys. Rev. Lett. 101 057010
[3] Cao C, Hirschfeld P J and Cheng H P 2008 Phys. Rev. B 77 220506(R)
[4] Ma F and Lu Z Y 2008 Phys. Rev. B 78 033111
[5] Dong J et al 2008 Europhys. Lett. 83 27006
[6] de la Cruz C et al 2008 Nature 453 899
[7] Chen Y et al 2008 Phys. Rev. B 78 064515
[8] Huang Q et al 2008 Phys. Rev. Lett. 101 257003
[9] Zhao J et al 2008 Phys. Rev. B 78 140504(R)
[10] Goldman A I et al 2008 Phys. Rev. B 78 106506(R)
[11] McGuire M A et al 2008 Phys. Rev. B 78 094517; [arXiv:0804.0796]
[12] Si Q and Abrahams E 2008 Phys. Rev. Lett. 101 076401
[13] Fang C et al 2008 Phys. Rev. B 77 224509
[14] Zhao J et al 2008 Phys. Rev. Lett. 101 167203
[15] Liu G B and Liu B G 2009 J. Phys.:Condens. Matter 21 195701
[16] Yao D X and Carlson E W 2008 Phys. Rev. B 72 052507
[17] Klauss H H et al 2008 Phys. Rev. Lett. 101 077005
[18] Tyablikov S V 1959 Ukr. Mat. Zh. 11 289; S. V. Tyablikov 1967 Methods in the Quantum Theory of Magnetism (New York: Plenum Press)
[19] Anderson F B and Callen H B 1964 Phys. Rev. 136 A1068
[20] Fröbrich P, Jensen P J and Kuntz P J 2000 Eur. Phys. J.B 13 477
[21] Henelius P, Fröbrich P, Kuntz P J et al 2002 Phys. Rev. B 66 094407
[22] Callen H B 1963 Phys. Rev. 130 890
[23] Yin Z P et al 2008 Phys. Rev. Lett. 101 047001