Rotationally invariant exchange interaction: The case of paramagnetic iron

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We present a generalization of the spin-fluctuation theory of magnetism which allows us to treat the full rotational invariance of the exchange interaction. The approach is formulated in terms of the local density approximation plus dynamical mean-field theory (LDA + DMFT), providing a systematic many-body treatment of the effect of spin-density fluctuations. This technique is employed to study the electronic and magnetic properties of paramagnetic α iron. Our result for the Curie temperature is in good agreement with experiment, while the calculations with the Ising-type exchange interaction yield almost twice the overestimated value.

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I. INTRODUCTION

The theoretical description of metallic magnets, especially those containing transition metals, is one of the central problems in condensed matter physics. Even today, in view of the great technological importance of such materials, a detailed understanding of their electronic, magnetic, and structural properties at finite temperatures remains problematic. This is mostly due to the presence of local magnetic moments above the magnetic ordering temperature which complicates the problem considerably and reduces the predictive power of first-principles calculations. Various properties of metallic magnets can be understood by using the spin-fluctuation theory[1] with its most general form based on a functional integral formulation.[2] This formulation was employed to describe the formation of local moments in paramagnetic metals[3] by reducing the many-body problem to a one-particle problem in a fluctuating external magnetic field and then evaluating the functional average. Stimulated by these results, different analytical and numerical methods have been developed, e.g., the well-known quantum Monte Carlo techniques.[4,5] By taking into account fluctuation corrections to the mean-field approximation, the spin-fluctuation theory has shown to provide a good qualitative description of the Curie-Weiss law behavior of magnetic susceptibility. However, applications of this technique to describe, e.g., the α-γ phase transition in iron, do not lead to satisfactory results.[6] In particular, it predicts the bcc-fcc phase transition to occur below the Curie temperature, T_C, while, in fact, this phase transition occurs 150 K above T_C.

The local density approximation plus dynamical mean-field theory (LDA + DMFT) method,[7] a combination of the *ab initio* local density approximation (LDA) of the density functional theory and dynamical mean-field theory (DMFT), nowadays has become a state-of-the-art approach for the calculation of electronic and magnetic properties of correlated electron compounds.[8] Applications of the LDA + DMFT to study transition-metal compounds have given a good quantitative description of localized as well as itinerant electron states.[9–11] These calculations predict the correct values of the local magnetic moment and magnetization, while the magnetic transition temperature turns out to be significantly overestimated. There are two reasons for the overestimation of the magnetic transition temperature. The first one is the single-site (local) nature of the DMFT approach, which is not able to capture the reduction of magnetic transition temperature due to long-wavelength spin waves.[9] The second reason comes from the approximate form of the local Coulomb repulsion restricted to the Ising-type exchange interaction. The model calculations for the Bethe lattice with the infinite coordination number (and thus exact in DMFT) show substantial overestimation of the Curie temperature for the density-density type of Coulomb interaction.[12] These problems can be repaired using DMFT extensions[13] and modern family of continuous-time quantum Monte Carlo (CT-QMC) solvers,[14] respectively. While the different DMFT extensions are actively developed and successfully applied to model systems, the implementations of these techniques to real materials (five-orbital systems) can really be counted by fingers.[15] The recently proposed continuous-time quantum Monte Carlo algorithms[14] as well as some other quantum impurity solvers[16] allow one to treat the Coulomb interaction in its general form retaining rotational spin symmetry. However, applications of these methods so far have been mostly limited to simple low degenerate model systems and only a few realistic calculations for the 3*d* compounds have recently appeared.[17] The main drawback of the CT-QMC algorithms with the full Coulomb interaction matrix is a high computational cost due to the exponential scaling with the number of orbitals. Therefore, at present, the density-density part of the Coulomb repulsion has been employed in the most material-specific calculations of the uniform magnetic susceptibility. However, as we will show below on the example of paramagnetic iron, retaining spin rotational symmetry is crucial for the correct description of magnetic properties.

In this paper, we present the spin-fluctuation theory of magnetism which is formulated in the framework of the LDA + DMFT method. The approach provides a systematic treatment of the effect of local electronic correlations by reducing the many-body problem to the functional integral over a fluctuating magnetic field on an effective impurity. The spin-fluctuation theory is generalized by replacing a scalar fluctuating magnetic field with a vector one. This allows one to take into account the full rotational invariance of the exchange interaction instead of the approximate Ising-type form. The

proposed method is employed to study the electronic and magnetic properties of paramagnetic α iron, resulting in a Curie temperature value which is in good agreement with experiment.

II. METHOD

We start with the simple Hamiltonian of the Coulomb interaction in the following form:

$$\hat{H}_{\text{Coul}} = \frac{1}{2}\sum_{\mu,\nu,\sigma} U\hat{n}_{\mu\sigma}\hat{n}_{\nu\bar{\sigma}} + \frac{1}{2}\sum_{\substack{\mu,\nu,\sigma \\ \mu\neq\nu}} (U-J)\hat{n}_{\mu\sigma}\hat{n}_{\nu\sigma}, \quad (1)$$

where $\hat{n}_{\mu\sigma}$ denotes the electron number operator with spin σ $(=\uparrow, \downarrow)$ at orbital μ. Using the total electron number operator, $\hat{N} = \sum_{\mu\sigma}\hat{n}_{\mu\sigma}$, and the z projection of the spin operator, $\hat{S}_z = \sum_{\mu}(\hat{n}_{\mu\uparrow} - \hat{n}_{\mu\downarrow})/2$, the Hamiltonian can be rewritten as

$$\hat{H}_{\text{Coul}} = \tfrac{1}{2}\bar{U}\hat{N}(\hat{N}-1) + \tfrac{1}{4}J\hat{N} - J\hat{S}_z^2, \quad (2)$$

where $\bar{U} = U - J/2$ is the average value of the Coulomb interaction. This Hamiltonian represents the density-density part of the Coulomb interaction and contains the exchange interaction in the Ising-type form. To restore the spin rotational symmetry, one should replace the z projection of the spin operator, $\hat{S}_z$, to the vector spin operator, $\hat{\vec{S}}$. Therefore, the Hamiltonian with the rotationally invariant exchange interaction reads

$$\hat{H}_{\text{Coul}} = \tfrac{1}{2}\bar{U}\hat{N}(\hat{N}-1) + \tfrac{1}{4}J\hat{N} - J\hat{\vec{S}}^2. \quad (3)$$

Following the spin-fluctuation theory, we assume that the charge fluctuations are of high frequency and thus the characteristic time of the charge fluctuations is substantially smaller than that of the spin fluctuations. This makes it reasonable to average (not neglect) the contribution from the charge fluctuations and focus on the spin dynamics. The first quadratic term in Eq. (3) can be treated as $\hat{N}^2 \to \hat{N}n_d$, where $n_d = \langle \hat{N} \rangle$ is the number of $3d$ electrons averaged over spin fluctuation time. The Hamiltonian of the system can be expressed as

$$\hat{H} = \hat{H}_{\text{LDA}} + \bar{U}(n_d - n_{d0})\hat{N} - J\hat{\vec{S}}^2, \quad (4)$$

where the first term, $\hat{H}_{\text{LDA}}$, is a kinetic contribution borrowed from the LDA. The second term is a combination of the LDA double counting term (n_{d0} is the LDA value for n_d) and static charge term discussed above. The last term is responsible for spin fluctuations.

In the DMFT approach the lattice problem with Hamiltonian (4) is mapped onto a quantum impurity model. Using the general form of the Hubbard-Stratonovich transformation,[2] the partition function can be expressed as a functional integral

$$Z = \int D\vec{\xi}(\tau)\exp\left[-\frac{\pi}{\beta}\int_0^\beta \vec{\xi}^{\,2}(\tau)d\tau\right] Z(\vec{\xi}), \quad (5)$$

where

$$Z(\vec{\xi}) = \text{Tr}\left\{T_\tau \exp\left[-\beta\hat{H}_{\text{LDA}} - \beta\bar{U}(n_d - n_{d0})\hat{N} + 2c\int_0^\beta \vec{\xi}(\tau)\hat{\vec{S}}\,d\tau\right]\right\}. \quad (6)$$

Here, T_τ denotes the time-ordering operator, β the inverse temperature, and $c = \sqrt{\pi J/\beta}$. Function $\vec{\xi}(\tau)$ stands for an effective magnetic field resulting in the potential $\hat{V}(\tau) = 2c\,\vec{\xi}(\tau)\hat{\vec{S}}$. The functional integral over all fluctuating fields gives a solution of the impurity problem.

In the functional integral formulation of the conventional spin-fluctuation theory, the fluctuating magnetic field in Eq. (5) is considered to be scalar. The generalization to a vector field corresponds to the transition from Eq. (2) to Eq. (3) and allows one to take into account the spin-rotational symmetry, thereby extending the theory from the Ising-type exchange interaction to the full rotationally invariant one. Dividing the imaginary time interval $[0,\beta]$ on L slices of length $\Delta\tau$ and using the Trotter breakup for the exponential operator in Eq. (6), the partition function $Z(\vec{\xi})$ for a given $\vec{\xi}(\tau)$ can be expressed as

$$Z(\vec{\xi}) \simeq \text{Tr}\left\{T_\tau \prod_{l=1}^{L}(\exp[-\Delta\tau\hat{H}_0]\exp[\hat{V}(\tau_l)])\right\}, \quad (7)$$

where $\hat{H}_0 = \hat{H}_{\text{LDA}} + U(n_d - n_{d0})\hat{N}$ is the $\vec{\xi}$-independent part of the Hamiltonian. These equations are similar to those of the Hirsch-Fye quantum Monte Carlo (HF-QMC) method.[5] The partition function can be written as

$$Z = \sum_{\{\vec{\xi}\}}\exp\left[-\frac{\pi}{L}\sum_{l=1}^{L}\vec{\xi}^{\,2}(\tau_l)\right]\prod_{\mu}\det\left[G_\mu^{-1}(\vec{\xi})\right]. \quad (8)$$

However, instead of a single spin flip as in the HF-QMC method, here one should stochastically change the value of the field $\vec{\xi}(\tau)$ for a random value of imaginary time. Due to the vector nature of the spin operator $\hat{\vec{S}}$, the instant Green function for a given fluctuation field configuration becomes nondiagonal in spin indexes. The interacting Green function restores the spin symmetry after averaging over all fluctuating fields. The computational scheme where the partition function is calculated with an auxiliary vector magnetic field is referred to below as $\vec{J}$-QMC. By taking into account only the z component of the field, the approximate form of the local Coulomb interaction, limited to the Ising-type exchange interaction, is assumed (referred to as J_z-QMC).

III. RESULTS AND DISCUSSION

Elemental iron is one of the most famous itinerant-electron ferromagnets which exhibits localized moment behavior above the Curie temperature T_C. Although various properties of the low-temperature ferromagnetic state of Fe can be understood within the density functional theory,[18] applications of these techniques to describe the *paramagnetic* state do not lead to satisfactory results. Clearly, an overall understanding of the properties of iron requires a formalism which takes into account the existence of local magnetic moments above T_C.[19] Recent applications of the LDA + DMFT have shown to provide a qualitatively correct description of the electronic, magnetic, and structural properties of paramagnetic iron.[9,11,20–22] However, a quantitative agreement has been achieved only in terms of the reduced temperature T/T_C, while the calculated Curie temperature[9] was found to be about twice larger than the experimental value of 1043 K.[23] The average Coulomb

interaction in the Fe $3d$ shell is considerably smaller than the bandwidth, showing no evidence for the formation of Hubbard bands in the spectral function. However, due to the strong exchange interaction,[11] local magnetic moments are formed that are accompanied by loss of coherence for metallic states due to scattering of electrons on the fluctuating spins. The charge fluctuations in paramagnetic iron are of high frequency with $3d$ electrons being far from the localization limit. In addition, body-centered cubic iron has a large coordination number, $z = 8$, implying small error in Curie temperature due to the local nature of the DMFT. These arguments make iron an ideal candidate for our study.

To calculate the electronic structure of paramagnetic α iron within the LDA, the tight-binding linear muffin-tin orbital (TB-LMTO) method was employed.[24] The low-energy Hamiltonian containing the $4s$, $4p$, and $3d$ states has been constructed with the use of an Nth-order muffin-tin orbital (NMTO) method.[25] In our calculations, we used the value of the screened Coulomb interaction, $U = 2.3$ eV, and the value of Hund's exchange, $J = 0.9$ eV, which are consistent with the previous estimations.[9,11,20,26]

In Fig. 1 we present the partial densities of states and the corresponding imaginary parts of the self-energies obtained by the LDA + DMFT at $\beta = 10$ eV^{-1}. The HF-QMC and $\vec{J}$-QMC calculations give qualitatively similar results, reproducing the splitting in the density of states of the e_g orbitals near the Fermi level caused by exchange interaction.[11] The splitting in the density of states of the t_{2g} orbitals is found to occur in the LDA calculation and hence can be attributed to the band-structure effects. In both approaches, the self-energies for the e_g orbitals diverge at low frequencies forming a non-Fermi-liquid state.

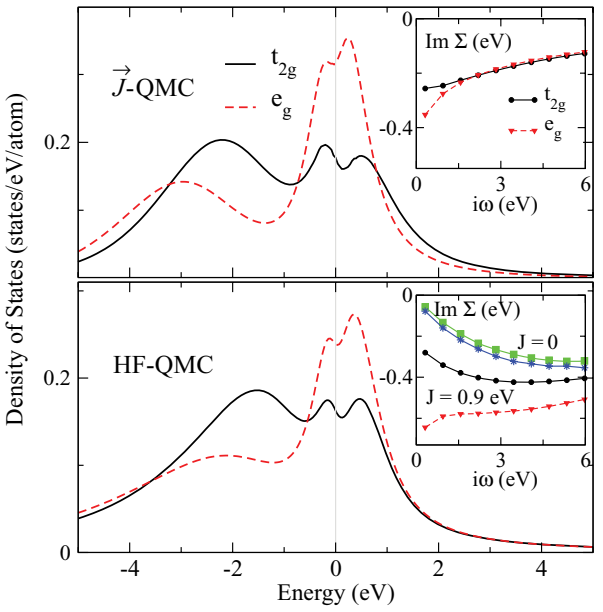

FIG. 1. (Color online) Partial t_{2g} (black) and e_g (red dashed) densities of states obtained by $\vec{J}$-QMC (top panel) and HF-QMC (bottom panel) calculations within LDA + DMFT. The Fermi level is indicated by the vertical (gray) line at zero energy. Insets: imaginary parts of the corresponding self-energies. The lower inset also shows the imaginary parts of the t_{2g} (green squares) and e_g (blue stars) self-energies obtained by HF-QMC with $J = 0$.

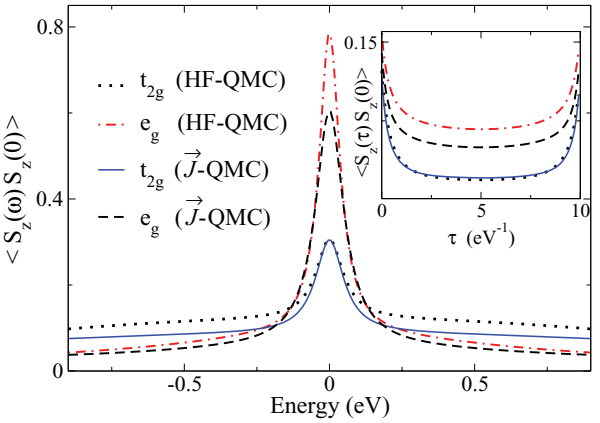

FIG. 2. (Color online) Spin-spin correlation functions on the real and imaginary energy (inset) axes calculated by the HF-QMC and $\vec{J}$-QMC within LDA + DMFT.

The self-energy for the t_{2g} orbitals remains Fermi-liquid-like in HF-QMC, but looks like non-Fermi-liquid in $\vec{J}$-QMC. However, the calculations at lower temperatures indicate that the coherence for the t_{2g} states is restored. Thus, the formation of local magnetic moments is accompanied by more incoherent e_g states and itinerant t_{2g} states.[11] The smaller magnitudes of self-energies in $\vec{J}$-QMC than in HF-QMC can indicate that the proposed method has a tendency to underestimate the strength of Coulomb correlations. In the lower inset we also present the imaginary parts of self-energies calculated by HF-QMC with the exchange parameter switched off, $J = 0$. One can clearly see that the self-energies tend to zero at low frequencies. This confirms again that the incoherent behavior is defined by the exchange interaction. We also note that at low frequencies the self-energies obtained by the $\vec{J}$-QMC method are close to those of the HF-QMC. This indicates that the physics near the Fermi level is dominated by the spin fluctuations, while the charge fluctuations play a minor role.

In Fig. 2 we show our results for the orbitally-resolved spin-spin correlation functions on the real and imaginary energy axes. In both approaches, the non-Fermi-liquid behavior of the e_g electrons yields a pronounced peak at zero energy of the real energy axis indicating the presence of local magnetic moments. The satisfactory agreement of the results obtained by the HF-QMC and $\vec{J}$-QMC methods suggests that the effect of the charge fluctuations, averaged within the $\vec{J}$-QMC approach, is minor.

To proceed further we compute the uniform magnetic susceptibility as a response to an external magnetic field. The temperature dependence of the inverse uniform magnetic susceptibility obtained by the LDA + DMFT shows a linear behavior at high temperatures (Fig. 3). This indicates the presence of local magnetic moments and corresponds to the Curie-Weiss law, $\chi^{-1} = 3(T - T_C)/\mu_{\text{eff}}^2$, where T_C is the Curie temperature and μ_{eff} is the effective local magnetic moment. The results of the least-square fit to the Curie-Weiss law are shown in Fig. 3 by straight lines. It is clearly seen that the HF-QMC method, limited to the Ising-type exchange interaction, overestimates the Curie temperature value almost

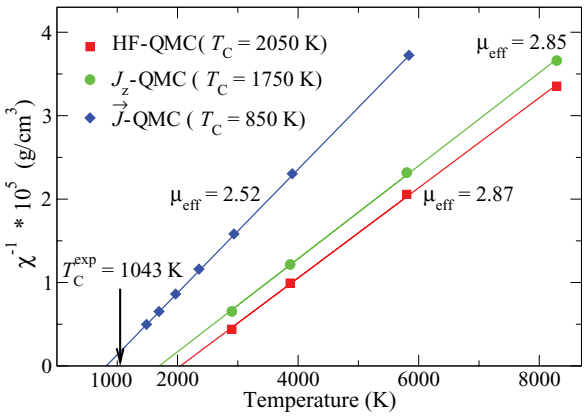

FIG. 3. (Color online) Temperature dependence of the inverse uniform magnetic susceptibility obtained by the LDA + DMFT. The straight lines depict the least-squares fit to the Curie-Weiss law. The experimental value of $T_C = 1043$ K is denoted by the (black) arrow. The experimental value of the local magnetic moment is $\mu_{\text{eff}}^{\text{exp}} = 3.13\mu_B$ (Ref. 23).

twice. The J_z-QMC approach, which has the Ising-type exchange interaction, gives a slightly smaller value of the T_C than the HF-QMC. This confirms the validity of the static approximation for the charge degrees of freedom. Taking into account the full rotationally invariant exchange interaction, our calculations result in a substantial decrease of the T_C value, which is now found to be in satisfactory agreement with experiment. These findings are compatible with the results of the recent two-band model studies.[12] In the case of α iron, the influence of the approximate treatment of the charge fluctuations on the Curie temperature appears to be much smaller than the inclusion of spin-flip terms to the Hamiltonian.

IV. CONCLUSIONS

We presented a generalization of the spin-fluctuation theory of magnetism which allows one to take into account the full rotational invariance of the exchange interaction. The approach is formulated in terms of the LDA + DMFT method, providing a systematic many-body treatment of the effect of spin-density fluctuations. We employed this technique to study the electronic and magnetic properties of α iron. Our results agree well with experiment and show that overestimation of the Curie temperature by the LDA + DMFT in the case of α iron is mostly related to the approximate (Ising-type) treatment of the exchange Coulomb interaction rather than to the single-site nature of the DMFT.

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