Spin-density fluctuation in paramagnets

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We study the effects of spin-density fluctuation in itinerant electron magnetism. Spin fluctuation is described in terms of an averaged electron-hole polarization bubble dressed with paramagnons in effective random potential. We approximate the polarization bubble by letting all internal paramagnon lines have zero momentum transfer and by taking the Gaussian average over the fluctuating local field. The resultant static susceptibilities calculated for paramagnetic systems agree with experimental data better than those from RPA which is the noninteracting limit of this theory. The high temperature susceptibilities of Ni and Co approximate the Curie-Weiss law for intermediate coupling. [S0163-1829(96)02725-7]

I. INTRODUCTION

The problem of itinerant electron magnetism (IEM) is of fundamental importance in understanding the collective effects in many-body systems. For instance, the localized magnetic moments cannot explain[1] (i) that the neutron-diffraction data showing atomic magnetic moments are located between atoms thus being identified with the mobile electrons, (ii) that the number of Bohr magnetons per atom is not integral and the bandwidths are the order of electron volts, and (iii) that the specific heat of some materials is much higher than what is calculated with localized moment theory. The mean-field approximation is not well suited for IEM, as shown in the fact that the Stoner theory of a molecular field cannot explain the Curie-Weiss law.[2] Among many studies on IEM, Wang, Evenson, and Schrieffer[3] (WES) substituted the two-body interaction with the Gaussian functional average of a fluctuating one-body potential following Hubbard[4] and Muhlsclegel.[4] They solved the one-site Anderson model for temperatures higher than the characteristic spin-fluctuation temperature. Murata and Doniach[5] (MD) formed the partition function with a classical functional integral over the magnetization field. Hertz and Klenin[6] (HK) calculated the self-energy of paramagnon (which is exchange-enhanced spin fluctuations) and the static susceptibility, using similar methods of WES and MD. Moriya and Kawabata[7] added corrections to the Hartree Fock energy in terms of dynamic susceptibility. Assuming a free-electron-like band, they derived the static uniform susceptibility being consistent with a Curie-Weiss law for weakly correlated itinerant magnets. Lonzarich and Taillefer[8] used the MD model to study the magnetic equation of state of weak ferromagnets. After all these efforts, the theory is still far from satisfactory in explaining the experimentally observed, temperature-dependent magnetic susceptibility of various materials.[9]

In this paper, we study the exchange-enhanced spin fluctuation to explicitly calculate the temperature-dependent susceptibility for the paramagnetic states of itinerant electron ferromagnets. Calculated spin susceptibilities are compared with experimental data of a strong ferromagnet nickel which has the least-localized spins among pure transition metals and another ferromagnet cobalt. Since fluctuations in both of these materials are highly localized in momentum space, we employ a similar procedure to that done by WES and HK, while using different approximations for averaged electron-hole polarization bubble and the susceptibilities.

II. SPIN FLUCTUATIONS AND PARAMAGNON INTERACTIONS

The Hubbard Hamiltonian of on-site Coulomb repulsion is

$$H_U = U\sum_i^N n_{i\uparrow}n_{i\downarrow} = \frac{U}{4}\sum_i [(n_{i\uparrow}+n_{i\downarrow})^2-(n_{i\uparrow}-n_{i\downarrow})^2]$$
$$= \frac{U}{4}\sum_i^N n_i^2 + H_I, \tag{1}$$

where N is the number of sites. We take the spin-dependent second part as the interaction Hamiltonian H_I. Due to spin-exchange interactions, spin density fluctuates throughout the medium, in addition to thermal fluctuation. This spin fluctuation is expressed[3,4,6] as $g\sigma\{\phi(\tau)-\phi(0)\}$ where g is the coupling between one spin and the effective magnetic field which arises due to the fluctuating spin density and given by $g^2 = Uk_BT$. Here, τ is the imaginary time and $\sigma=+1$ for up spin and -1 for down spin.

At finite temperature, the exact partition function is given:[10]

$$Z = Z_0\left\langle T_\tau \exp\left(-\int_0^\beta H_I d\tau\right)\right\rangle. \tag{2}$$

Using the Hubbard-Stratonovich transformation,[11] we reduce the quadratic exponent from the H_I to a linear term. Then the partition function becomes $Z=Z_0Z_I$, where

$$Z_0 = \int \mathcal{D}\phi(\tau)\exp\left(-\sum_i \beta^{-1}\int_0^\beta \phi(\tau)^2 d\tau\right), \quad (3)$$

$$Z_I = \mathrm{Tr}\left\langle T_\tau \exp\int_0^\beta \{-H_0 + g\phi(\tau)[n_{i\uparrow}(\tau) - n_{i\downarrow}(\tau)]\} d\tau\right\rangle. \quad (4)$$

The fluctuating effective potential can be written[3,6,11] $V^\sigma \equiv g\sigma\phi(\tau)$ which is the Zeeman term. We multiply the potential V in Z_I by an arbitrary parameter λ and differentiate with respect to λ to obtain

$$\frac{\partial}{\partial\lambda}\ln Z_I = -\sum_\sigma \int_0^\beta d\tau V^\sigma \langle n_{i\sigma}\rangle, \quad (5)$$

where

$$-\langle n_{i\sigma}\rangle = -\frac{\langle T_\tau n_{i\sigma}\exp[-\lambda\int_0^\beta d\tau' V^\sigma n_{i\sigma}(\tau')]\rangle}{\langle T_\tau \exp[-\lambda\int_0^\beta d\tau' V^\sigma n_{i\sigma}(\tau')]\rangle}$$
$$= -\langle T_\tau n_{i\sigma}(\tau) n_{i\sigma}(0)\rangle = G_i^\sigma(\tau). \quad (6)$$

Also, the Green's function is expressed as a series in terms of the free part G^0 and the propagator of an interacting particle (electron):

$$G^\sigma(\tau,\tau') = G_\sigma^0(\tau,\tau') + \lambda\int G^0 V^\sigma G^\sigma(\tau,\tau') d\tau. \quad (7)$$

Thus,

$$\frac{\partial}{\partial\lambda}\ln Z_I = -\sum_\sigma \frac{\partial}{\partial\lambda}\langle[\mathrm{Tr}\ln(1-\lambda V^\sigma G^0)]\rangle. \quad (8)$$

Integrating the equation over λ, we get

$$Z_I = \exp\left(-\sum_\sigma \mathrm{Tr}\ln(1-\lambda V^\sigma G^0)\right). \quad (9)$$

Then the total partition function is reduced to

$$Z = \int \mathcal{D}\phi(\tau)\exp\left(-\sum_i \beta^{-1}\int_0^\beta \phi(\tau)^2 d\tau - \sum_\sigma \mathrm{Tr}\ln(1-V^\sigma G^0)\right), \quad (10)$$

where the second part describes the electron scattering by the effective potential V.

Now we represent the spin fluctuation by paramagnon which is the quanta of elementary excitations in magnets for temperatures higher than the phase transition temperature. That is, paramagnon propagates as it polarizes medium thus creating electron-hole pair bubbles. The mean-square fluctuation of Fourier-transformed field components (averaged over the auxiliary field) is defined

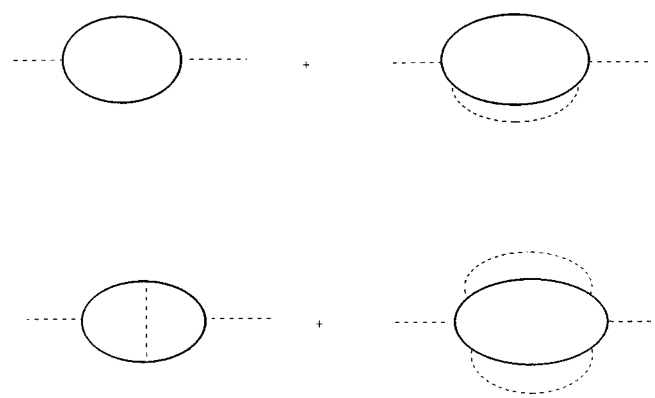

FIG. 1. Paramagnon self-energy diagrams shown to the fourth order in g. Solid lines are for electrons and holes, dotted lines outside the bubbles are the effective external field and the intermediate ones for paramagnon propagators.

$$\langle|\phi|^2\rangle = \frac{1}{Z}\int \prod_{q'm'} \frac{d\phi_{q'm'}}{\sqrt{\pi}}|\phi_{q'm'}|^2 \exp\left(-\sum_{q'm'}|\phi_{q'm'}|^2 + \sum_\sigma \mathrm{Tr}\ln(1-V^\sigma G^0)\right), \quad (11)$$

where q is the paramagnon wave vector or the momentum transfer.

We take the static limit $\omega_m = 0$ components only of the field ϕ_{qm}, for the thermal energy of $k_B T \gg k_B T_{\mathrm{SF}}$ where $k_B T_{\mathrm{SF}}$ is the characteristic energy[6,7] of spin fluctuation. Since $T_{\mathrm{SF}} \approx T_C$ in transition-metal ferromagnets, we work in the range $T > T_C$. While letting the intermediate paramagnon lines have zero momenta,[6] we keep the wave vector q on the paramagnon lines outside the bubbles (shown as dotted lines in Fig. 1). So, the paramagnons propagate (or spins fluctuate) in this ''static'' limit. The paramagnon self-energy diagrams[6,10] in Fig. 1 show the polarization bubble $B(q,i\omega_m)$ dressed by absorbing and reemitting paramagnons. We exclude the interactions between the emitted paramagnons in higher-order diagrams in the series. The pair bubble $B_2(q)$ is

$$\langle B_2(q,i\omega_m)\rangle \equiv \langle B_2(q)\rangle$$
$$= -\beta^{-1}\sum_{k,n}\langle G^\sigma(k,i\omega_n;\{\phi\}) \times G^\sigma(k+q,i\omega_{n+m};\{\phi\})\rangle. \quad (12)$$

From a generalized Hartree approximation of the diagram series and by requiring a self-consistent relation[6] between the polarization amplitude and the effective field, the mean-square fluctuation in Eq. (11) is determined:

$$\langle|\phi|^2\rangle = \frac{1}{2}\sum_q \frac{1}{1-U\langle B_2(q)\rangle}. \quad (13)$$

III. SPIN SUSCEPTIBILITY

The static susceptibility follows:

$$\chi(q,i\omega_m) = \frac{1}{U}(2\langle|\phi_{qm}|^2\rangle - 1) = \frac{\langle B_2(q)\rangle}{1-U\langle B_2(q)\rangle}. \quad (14)$$

Above χ has the general structure of the response function in its form except being written in terms of the *averaged* bubble $\langle B_2(q)\rangle$ which is an effective Lindhard function and the susceptibility at a noninteracting limit.

The Gaussian average of the polarization bubble is defined exactly:[3,6]

$$\langle B(q)\rangle=\int \frac{d\phi}{[2\pi\langle|\phi(q)|^2\rangle]^{1/2}} \times\exp[-\phi^2(q)/2\langle|\phi(q)|^2\rangle]B(q). \tag{15}$$

Up to this point, the general formalism followed the previous works by other authors as cited. Now we approximate the wave-vector-dependent pair bubble differently (from what was given in Ref. 6 by HK, for instance). Our purpose is to write the approximated average value of the pair bubble consistently from the definition of the Gaussian average given above and to calculate the temperature-dependent form of the spin susceptibility explicitly. We take

$$B(q)\approx B(\epsilon+g\phi)\approx B(\epsilon_F)+g\phi B'(\epsilon+g\phi)|_{\epsilon_F}+\cdots\approx B(\epsilon_F)$$

by expanding it around the Fermi energy ϵ_F and taking into account of the fact that the lowest-order bubble for noninteracting electron falls off very slowly from its maximum value at $q=0$ for $0<q<2k_F$. The ϵ_F is actually the chemical potential which is determined from fixing the electron number $n=\int f(\epsilon)\langle N(\epsilon)\rangle d\epsilon$. Then we take the averaged bubble at the long-wavelength limit as

$$\langle B_2(q)\rangle\approx\langle B_2(0)\rangle\exp[-\phi(q)^2/2\langle|\phi|^2\rangle], \tag{16}$$

where the exponential term comes from the integral in Eq. (15).

From the expression of the bubble as a product of electron Green's functions, we find the pair bubble as energy integrals by Poisson's summation formula,[6,11] including the thermal factor through the Fermi distribution function $f(\epsilon)$:

$$B_n(0)=\frac{1}{(n-1)!}\int d\epsilon N_0(\epsilon)f^{(n-1)}(\epsilon). \tag{17}$$

In order to determine temperature-dependent susceptibility, we note that the general structure of susceptibility functions (correlations) is an average of a product of density of states (at different times or at different temperatures, for instance).

First we approximate the derivative of Fermi function $f^{(n-1)}(\epsilon)$ with a Gaussian density of states:[6]

$$N(\epsilon)=\frac{1}{\sqrt{2\pi(w^2+\pi^2/3\beta^2)}}\exp\left[-\epsilon^2/2\left(w^2+\frac{\pi^2}{3\beta^2}\right)\right], \tag{18}$$

where the factor $\pi^2/3\beta^2$ is from the second moment of the derivative of Fermi function and the width $w=g\sqrt{\langle\phi^2\rangle}$ which is from considering what follows . According to the random-phase approximation which is the noninteracting limit of this theory, we have ferromagnetic instability when $U\langle N(\epsilon)\rangle>1$. In the intermediate-coupling region where $UN(\epsilon)\geqslant 1$, we take the average $\langle N(\epsilon)\rangle$ over the energy width $\langle\phi^2\rangle\sqrt{Uk_BT}$. Next, we integrate after taking the product of the band density of states $N_0(\epsilon)=(2\pi w^2)^{-1/2}e^{-\epsilon^2/2w^2}$ with the above Gaussian density of states N. Then, we equate $B_n(0)=1/(n-1)!N^{(n-2)}(\epsilon_F)$ for the effective density of states at finite temperature.[6] That is, we have the average pair bubble $\langle B_2(0)\rangle=\langle N(\epsilon_F)\rangle$.

Thus, we arrive at

$$\langle B_2(0)\rangle=\int\frac{d\phi}{2\pi\langle\phi^2\rangle\sqrt{(g^2\langle\phi^2\rangle+\pi^2/3\beta^2)}} \times\exp\left[-\frac{\phi^2(g^2\langle\phi^2\rangle+\pi^2/3\beta^2)+\epsilon_F^2\langle\phi^2\rangle}{2(g^2\langle\phi^2\rangle+\pi^2/3\beta^2)\langle\phi^2\rangle}\right] \tag{19}$$

$$\approx\frac{1}{4\pi\sqrt{g^2+\pi^2/3\beta^2}} \times\exp\left[-\frac{1}{4\pi g^2(g^2+\pi^2/3\beta^2)}\right], \tag{20}$$

where the ϕ integral is over the range of $g\sqrt{\langle\phi^2\rangle}, 0.5\leqslant\langle\phi^2\rangle\geqslant 1.5$ and the chemical potential near the top of the band is $\epsilon_F=1/2\pi g^2\langle\phi^2\rangle\approx 1/2\pi Uk_BT$.

Finally, the inverse susceptibility is computed

$$\chi^{-1}=\frac{1-U\langle B_2(0)\rangle}{\langle B_2(0)\rangle}\approx\sqrt{\pi k_BT[U+(\pi^2/3)k_BT]}-U. \tag{21}$$

We choose eV units since the Coulomb interaction U is usually given in eV. Above χ^{-1} is not identical to $\chi^{-1}=C^{-1}(T-T_C)$ in its algebraic form. But, the actual curve of this, plotted as a function of temperature, is very much linear over most of the temperature range of interest. As a matter of fact, many experimental data of inverse susceptibilities of many paramagnets differ from the simple straight line of the $C^{-1}(T-T_C)$ type.

IV. RESULTS AND CONCLUSION

We calculate the spin susceptibility of nickel which is a representative of the face-centered-cubic iron group and a pure transition metal whose itinerant ferromagnetism calls for clear understanding based on an improved theory beyond the Stoner model. The inverse susceptibility of Ni as a function of temperature plotted in Fig. 2 approximates the Curie-Weiss law very well. The solid line from our calculation is for the Coulomb interaction U of 0.23 eV fitting the experimental data[12] most closely while the effective U value estimated for the $3d^84f^2$ configuration is between 0.11 eV (according to Herring[13]) and 0.5 eV (according to Gunnarson in Wohlfarth,[13] based on the spin-density functional formalism). This is a much better agreement than the qualitative nature of previous calculations[6] that formulated a similar microscopic theory. The paramagnetic temperature Θ for the Curie-Weiss-like line in Fig. 2 is 600 K, while the experimentally determined Curie temperature is $\Theta=654$ K. Since the theory is meant to apply to the paramagnetic states for higher temperatures, the minor discrepancy in Θ should not concern us. Actually, experimental data for many paramag-

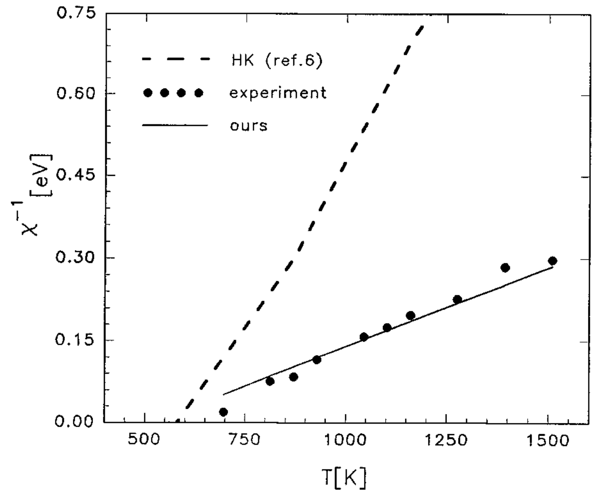

FIG. 2. Inverse susceptibility of Ni. The experimental data are from Zornberg (Ref. 12).

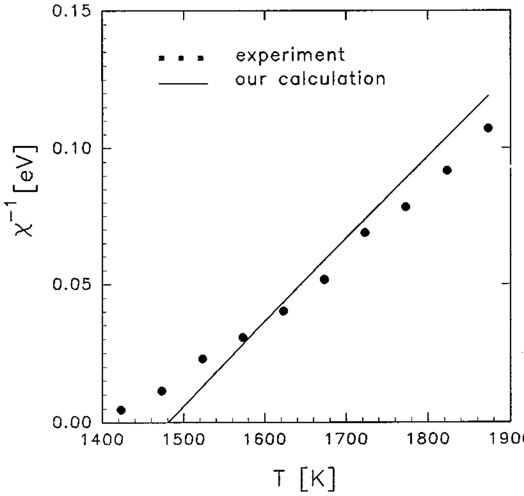

FIG. 3. Inverse susceptibility of beta Co. The experimental data are from Wijn (Ref. 12).

nets show nonlinear behaviors near the critical points. The random-phase-approximation (RPA) result for similar range of U values falls between the dotted line (HK) and the solid line which is our calculation, in Fig. 2. In Fig. 3, we have the inverse susceptibility of beta cobalt (fcc) which is another pure transition-metal ferromagnet. The paramagnetic Curie temperature Θ is known to be between 1403 and 1423 K and the effective $U\approx0.9$ eV according to Gunnarson in Wohlfarth.[13] Our calculated fitting line for the experimental data is with $U=0.62$ eV, which is rather close, considering the semiquantitative nature of these estimates. The Θ from our calculation is between 1400 and 1480 K (the range coming from the difference in the slopes of fitting lines).

Both results show the improvements from the RPA calculation which is a partial sum of ring diagrams valid for the limit of no coupling between one spin (of one electron) and the rest of spins in the medium. The susceptibility expression we have is in terms of the (electron-hole) pair bubble averaged over a fluctuating effective field which represents the spin fluctuation. Though the paramagnetic susceptibility as a function of temperature has been studied before by some authors,[6,13] it has not been written out explicitly in a nonphenomenological version of the exchange-enhanced spin-fluctuation theory. Also, we used a different approximation of the averaged pair bubble from that of HK. Unlike some studies in which arbitrary momentum cutoffs were introduced, our procedure does not require such parameters, since we evaluated for the static limit.

As for low-density systems, we may employ the ladder approximation as well. The criterion for low density[10] would be $k_F R \ll 1$ where R is the range of repulsive force. Since the particle contribution is much greater than the hole contribution then, the ladder diagrams (having the least number hole lines) would be the dominant ones. As a microscopically well-founded calculation, the agreements of our calculation and the experimental data indicate that the spin fluctuations in paramagnetic systems are the dominant mechanism for the Curie-Weiss behavior. Comparisons with other contributions such as electron-phonon interactions are left for future study.

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