PHYSICAL REVIEW B VOLUME 56, NUMBER 13 1 OCTOBER 1997-I

Itinerant-electron theory of metamagnetism

Y. Nishiyama and S. Hirooka
Department of Electrical and Electronics Engineering, Kagoshima University, Kagoshima 890, Japan

(Received 4 June 1997)

Itinerant-electron theory of metamagnetism taking into account the effect of spin fluctuations is proposed on the Hubbard model within the static approximation in the functional integral formalism. It is possible to treat the metamagnetism of the system with large amplitudes of the moment and spin fluctuation. Effects of spin fluctuations and Stoner excitations on the temperature-dependence of paramagnetic susceptibility and the metamagnetic transition under external magnetic field are investigated numerically on a simple model (electronic) density of states. It is shown that both effects are quantitatively important in metamagnetism. [S0163-1829(97)00138-0]

Today, the generation of ultrahigh magnetic fields over 100 T in a laboratory has become practical and they have been revealing interesting magnetic properties of materials. Particularly, the itinerant-electron metamagnetic transition (MT) induced by an external magnetic field in d-electron systems is one of the most interesting phenomena and it has been investigated both from the theoretical and experimental points of view. Experimentally, it is well known that a typical metamagnetic transition is observed in various Co-based Laves phase compounds such as YCo_2 and $LuCo_2$.[1,2] Moreover, these materials are strongly exchange-enhanced paramagnets and show a broad maximum in the temperature dependence of paramagnetic susceptibility at room temperature. Theoretically, Wohlfarth and Rhodes[3] have pointed out on the phenomenological Landau theory that the MT occurs if there exists a maximum in the paramagnetic susceptibility. Shimizu[4] has obtained a condition for the appearance of the MT. However, Landau expansion coefficients estimated on the Stoner model are weakly dependent on temperature. So, it seems that their theories do not well describe the finite temperature behavior of the metamagnetic materials.

Recently, Yamada[5] has developed the itinerant-electron metamagnetism at finite temperature, by taking into account the effect of spin fluctuations on the phenomenological Landau-Ginzburg theory. He has shown that the paramagnetic susceptibility always shows a maximum in its temperature dependence when the MT from the paramagnetic to the ferromagnetic state is induced by the external magnetic field at low temperature. And he has obtained a condition among the Landau coefficients for the appearance of the first-order transition in the temperature dependence of the spontaneous magnetization. These results are worthy in explaining the universal properties that are observed in the magnetic materials showing the MT. However, there are some restrictions in the application of his phenomenological method to the realistic system. First, the Landau-Ginzburg free-energy functional is expanded in the powers of the sixth order of the magnetization density. So, his model is inadequate in the case of large amplitudes of the magnetization density. Second, the estimation of the Landau expansion coefficients on the band calculation is not so easy.

Previously, one of authors presented a method that takes account of the spin-fluctuation effect on itinerant-electron ferromagnets in the functional-integral formalism and in the Gaussian approximation.[6,7] It corresponds to the improved theory of Hertz and Klenin.[8] In this work, the Hubbard model is used as before and we will apply this method to the metamagnetism within the static approximation. When the static approximation is applied to previous results,[7] obtained results are summarized as follows. Transverse and longitudinal susceptibilities are

$$\chi_{-+}(\mathbf{q})=\frac{\hat{\chi}_{-+}(\mathbf{q})}{1-U\hat{\chi}_{-+}(\mathbf{q})}, \tag{1}$$

$$\chi_{z}(\mathbf{q})=\frac{\hat{\chi}_{z}(\mathbf{q})}{1-U\hat{\chi}_{z}(\mathbf{q})}, \tag{2}$$

and

$$\hat{\chi}_{-+}(\mathbf{q})=\frac{1}{2}\left\langle\left(\frac{\delta\eta_t}{\eta}\right)^2\chi_{0z}(\mathbf{q})+\left[1+\left(\frac{\eta_z}{\eta}\right)^2\right]\chi_{0-+}(\mathbf{q})\right\rangle_{\delta\eta}, \tag{3}$$

$$\hat{\chi}_{z}(\mathbf{q})=\left\langle\left(\frac{\eta_z}{\eta}\right)^2\chi_{0z}(\mathbf{q})+\left(\frac{\delta\eta_t}{\eta}\right)^2\chi_{0-+}(\mathbf{q})\right\rangle_{\delta\eta}, \tag{4}$$

where $\eta^2=\delta\eta_t^2+\eta_z^2$, $\eta_z=h_z+\eta_0+\delta\eta_z$, and $\delta\eta_t^2=\delta\eta_x^2+\delta\eta_y^2$. $\delta\vec{\eta}=(\delta\eta_x,\delta\eta_y,\delta\eta_z)$ is a fluctuatiing field, h_z is an external magnetic field, and η_0 is a constant field induced by h_z. $\chi_{0-+}(\mathbf{q})$ and $\chi_{0z}(\mathbf{q})$ in Eqs. (3) and (4), which are transverse and longitudinal susceptibilities of noninteracting system under the field η, are defined as

$$\chi_{0\sigma\mu}(\mathbf{q})=\sum_{\mathbf{k}}\frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}\sigma})-f(\varepsilon_{\mathbf{k}\mu})}{\varepsilon_{\mathbf{k}\mu}-\varepsilon_{\mathbf{k}+\mathbf{q}\sigma}}, \tag{5}$$

$$\chi_{0z}(\mathbf{q})=\frac{2\chi_{0++}(\mathbf{q})\chi_{0--}(\mathbf{q})}{\chi_{0++}(\mathbf{q})+\chi_{0--}(\mathbf{q})}, \tag{6}$$

where σ, $\mu=+$ or $-$, $f(\varepsilon_{\mathbf{k}\sigma})=\{1+\exp[\beta(\varepsilon_{\mathbf{k}\sigma}-\zeta)]\}^{-1}$ and $\varepsilon_{\mathbf{k}\sigma}=\varepsilon_{\mathbf{k}}-\sigma\eta$. Averages $\langle\cdots\rangle_{\delta\eta}$ in Eqs. (3) and (4) are defined as $\langle A(\delta\eta)\rangle_{\delta\eta}$

$=\int_{-\infty}^{\infty}d(\delta\eta_z)\int_0^{\infty}d(\delta\eta_t)\delta\eta_t\rho(\delta\eta)A(\delta\eta)$ and the Gaussian distribution function $\rho(\delta\eta)$ for fluctuating field $\delta\eta$ is given as

$$\rho(\delta\eta)=\frac{1}{(2\pi)^{1/2}\sigma_z\sigma_t^2}\exp\{-[(\delta\eta_t/\sigma_z)^2+(\delta\eta_z/\sigma_z)^2]/2\}. \tag{7}$$

The variances of $\rho(\delta\eta)$ are

$$\sigma_t^2=\frac{U^2}{2\beta}\sum_{\mathbf{q}}\chi_{-+}(\mathbf{q}), \tag{8}$$

$$\sigma_z^2=\frac{U^2}{2\beta}\sum_{\mathbf{q}}\chi_z(\mathbf{q}), \tag{9}$$

where $\chi_{-+}(\mathbf{q})$ and $\chi_z(\mathbf{q})$ are given in Eqs. (1) and (2). Moreover, we must add to Eqs. (1)–(9) the condition for the determination of the Fermi energy ζ as $N_e=\langle\Sigma_{\mathbf{k}}\{f(\varepsilon_{\mathbf{k}+})+f(\varepsilon_{\mathbf{k}-})\}\rangle_{\delta\eta}$, where N_e the electron number.

When $h_z=0$ and so $\eta_0=0$ in the paramagnetic state, the system becomes isotropic. Therefore, we get $\hat{\chi}_p(\mathbf{q})\equiv\hat{\chi}_{-+}(\mathbf{q})=\hat{\chi}_z(\mathbf{q})$ from Eqs. (3) and (4) as

$$\hat{\chi}_p(\mathbf{q})=\tfrac{1}{3}\langle 2\chi_{0-+}(\mathbf{q})+\chi_{0z}(\mathbf{q})\rangle_{\delta\eta}. \tag{10}$$

Then, we have $\chi_p(\mathbf{q})\equiv\chi_{-+}(\mathbf{q})=\chi_z(\mathbf{q})$ and $\sigma_p^2\equiv\sigma_t^2=\sigma_z^2$ in Eqs. (1), (2), (8), and (9). The paramagnetic susceptibility is obtained by solving self-consistently these simplified equations.

When the amplitude of fluctuations is small, the uniform paramagnetic susceptibility can be expanded in a Taylor's series in powers of σ_p^2 in Eqs. (1), (3), and (10) as

$$\frac{1}{\chi_p(0)}\simeq\left(\frac{1}{A_0}-U\right)+\frac{5}{2A_0^2}\left(\frac{A_1^2}{A_0}-\frac{A_2}{3}\right){\sigma_p}^2+O(\sigma_p^4), \tag{11}$$

where $A_n=\int d^nD(\varepsilon)/d\varepsilon^n[-\partial f(\varepsilon)/\partial\varepsilon]d\varepsilon$, $D(\varepsilon)$ is the density of states (DOS) and $f(\varepsilon)$, the Fermi distribution function. The first term in Eq. (11) is the inverse paramagnetic susceptibility on the Hartree-Fock approximation and so $1/A_0-U>0$. Higher-order terms $O(\sigma_p^4)$ will increase with a rise in temperature. A necessary condition for metamagnetic transion is given by $A_1^2/A_0-A_2/3<0$ on the Stoner theory.[4] Then, because of $\sigma_p^2\propto T$ (T is the temperature) from Eqs. (8) and (9), Eq. (11) decreases with a rise in temperature at low temperature and so the paramagnetic susceptibility $\chi_p(0)$ has a maximum in the temperature dependence. This is the same result as the one shown by Yamada.[5] We have $A_2\simeq[d^2D(\varepsilon)/d\varepsilon^2]_{\varepsilon=\zeta}$ at low temperature. Therefore, as well known, we expect the metamagnetism in the system with DOS curve of a positive curvature, namely, $[d^2D(\varepsilon)/d\varepsilon^2]_{\varepsilon=\zeta}>0$, at Fermi energy.

On the other hand, when $h_z\neq 0$, the magnetic moment under the magnetic field is usually determined by the condition of thermodynamic equilibrium, namely, by minimizing the free energy. It gives

$$h_z=\eta_0^*-(U/2)\hat{M}_z(\eta_0^*), \tag{12}$$

where $\eta_0^*=\eta_0+h_z$, $\hat{M}_z(\eta_0^*)=\langle M(\eta)\eta_z/\eta\rangle_{\delta\eta}$, and $M(\eta)=\Sigma_{\mathbf{k}}\{f(\varepsilon_{\mathbf{k}+})-f(\varepsilon_{\mathbf{k}-})\}$. However, it is well known that an

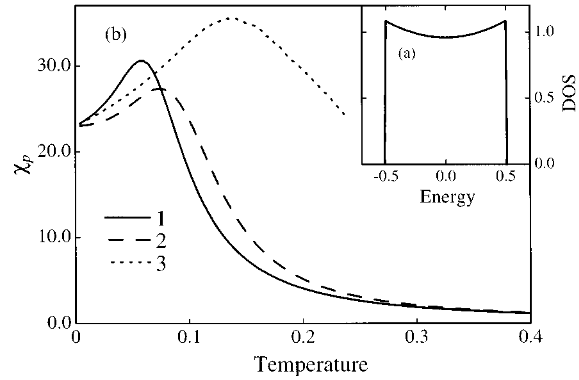

FIG. 1. (a) Inset, the electronic density of states (DOS) curve for $D_0=0.96$ and $\lambda=0.25$ in $D(\varepsilon)=D_0+\lambda\varepsilon^2$. (b) The paramagnetic susceptibility as a function of the temperature for $N_e=1.0$, $U=1.0$, and $N_{\rm sf}=0.22$. The solid curve (1) and the broken curve (2) are, respectively, the results for $N_{\rm sf}=0.22$ and $N_{\rm sf}=0.0$. The dotted curve (3) is the result neglecting the Stoner excitation for $N_{\rm sf}=0.22$.

inconvenient first-order transition occurs when we use the Gaussian approximation in the functional-integral formalism. So, in order to avoid these defects we have proposed another method making use of the rotational symmetry of the system for the determination of the spontaneous magnetization.[9,7] On the same consideration, we introduce the equation of determination of the magnetic moment under the external magnetic field h_z as follows:

$$h_z=\eta_0^*\{1-U\hat{\chi}_{-+}(0)\}, \tag{13}$$

where $\hat{\chi}_{-+}(0)$ is given by putting as $\mathbf{q}=0$ in Eq. (3) and the magnetic moment $\hat{M}_z(\eta_0^*)$ is obtained from $\hat{M}_z(\eta_0^*)=\langle M(\eta)\eta_z/\eta\rangle_{\delta\eta}$ and $M(\eta)=\Sigma_{\mathbf{k}}\{f(\varepsilon_{\mathbf{k}+})-f(\varepsilon_{\mathbf{k}-})\}$. When h_z is zero, Eqs. (12) and (13) become, respectively, $\eta_0=(U/2)\hat{M}(\eta_0)$ and $U\hat{\chi}_{-+}(0)=1$ as the equations of determination of the spontaneous magnetization in the ferromagnetic state. If neglecting fluctuations, both equations become $h_z=\eta_0^*-(U/2)M(\eta_0^*)$, which coincides with the result of the Hartree-Fock approximation. Although there are few little numerical differences between Eqs. (12) and (13) in the paramagnetic state in comparison with the ferromagnetic state, we employ Eq. (13). Thus, we can get the magnetization under external magnetic field by calculating self-consistently Eqs. (1)–(9).

Now, the evaluation of the integral by the wave vector $\mathbf{q}$ in Eqs. (8) and (9) is generally difficult. So, following Hertz and Klenin[8] we assume the q dependence of susceptibilities in the summation of the Eqs. (8) and (9) as $\hat{\chi}_{-+}(\mathbf{q})=\hat{\chi}_{-+}(0)\{1-(q/q_c)^2\}$ and $\hat{\chi}_z(\mathbf{q})=\hat{\chi}_z(0)\{1-(q/q_c)^2\}$ for $q\leqslant q_c$ and $\hat{\chi}_{-+}(\mathbf{q})=\hat{\chi}_z(\mathbf{q})=0$ for $q\geqslant q_c$. Here, q_c is the cutoff wave number and it is related to the freedom of spin fluctuations per atom as $N_{\rm sf}=a^3q_c^3/(6\pi^2)$ and $N_{\rm sf}<1$, where the a^3 is the atomic volume. The q_c or $N_{\rm sf}$ is one of the important parameters in our work. Under this approximation the evaluation of the integral by the wave vector $\mathbf{q}$ in the Eqs. (8) and (9) are carried out analytically. After all, we

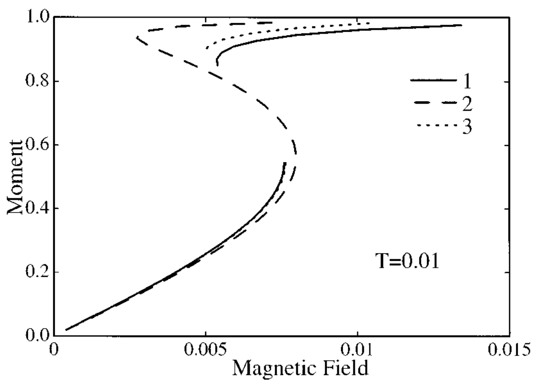

FIG. 2. The moment as a function of the external magnetic field for the temperature $T=0.01$. The three kinds of curves correspond to the ones in Fig. 1.

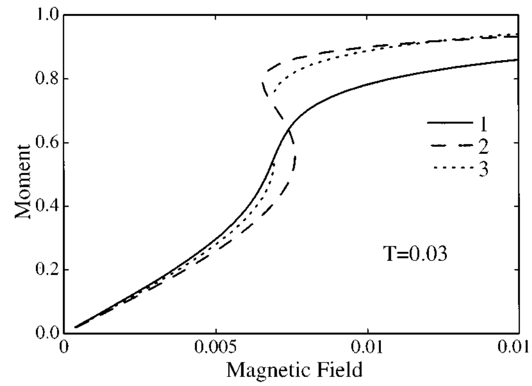

FIG. 3. The moment as a function of the external magnetic field for $T=0.03$. Three kind of curves correspond to the ones in Fig. 1.

need the Coulomb integral U, the DOS, the electron number N_e, and the freedom of spin fluctuations N_{sf} in our numerical calculation.

As an example, in order to investigate our method, we will show calculated results for a simple model DOS, $D(\varepsilon)=D_0+\lambda\varepsilon^2$, which is shown in Fig. 1(a). We expect metamagnetic behaviors of the system when $\lambda>0$. In the following calculation, temperature, magnetic field, susceptibilities, the DOS, and U are normalized by the bandwidth and so they are dimensionless. Figure 1(b) denotes calculated results of the temperature dependence of paramagnetic susceptibilities when $N_e=1.0$ per atom, $U=1.0$, $D_0=0.96$, and $\lambda=0.25$, and each results show a maximum in the temperature dependence. The solid curve and the broken curve give, respectively, the result for $N_{\mathrm{sf}}=0.22$ and the one for Hartree-Fock approximation ($N_{\mathrm{sf}}=0$). In a dotted curve, in order to investigate only the effect of spin fluctuations, Stoner excitation is neglected and so the Fermi distribution function is treated as a step function in Eqs. (5) and (6). This case corresponds to neglecting the temperature dependence of Landau expansion coefficients in Yamada's work.[5] In a solid curve both effects of Stoner excitation and spin fluctuations are considered. Figures 2 and 3 give calculated results of the magnetization under external magnetic field for the same values of parameters as above when the temperature $T=0.01$ and 0.03. They show the clear MT. Three kinds of curves correspond, respectively, to the ones in Fig. 1. It is noted that solid curves and dotted curves are broken off halfway along the curves. It is because these regions are thermodynamically unstable, where $\sigma_t^2<0$ and $\sigma_z^2<0$ in Eq. (7), and so Eqs. (1)–(9) and (13) do not have self-consistent solutions. From the calculated results it is found that the temperature of the susceptibility-maximum is shifted to low temperature and the MT becomes gradual with a rise in temperature due to effects of spin fluctuations as compared to Hartree-Fock approximation. At the same time, it should be stressed that both effects of Stoner excitation and spin fluctuations are equally important in temperature dependence of the paramagnetic susceptibility and in metamagnetic transition of the magnetization process.

In conclusion, it was found that our method can describe the overall behavior of metamagnetism for an arbitary amplitude of the magnetization with the knowledge of the DOS. This method was also used to investigate the contributions of the Stoner excitation and spin fluctuations by choosing values of the Coulomb integral U and the freedom of spin fluctuations N_{sf} using experimental results. The extension of our theory to multiple band systems and the comparison with experimental results of realistic itinerant metamagnetic systems, for example, Co-based Laves phase compounds such as YCo_2 and $LuCo_2$, will be carried out in future work.

[1] T. Goto, H. Aruga Katori, K. Fukamichi, H. Mitamura, K. Fukamichi, and K. Murata, J. Appl. Phys. **76**, 6682 (1994).

[2] T. Sakakibara, T. Goto, K. Yoshimura, and K. Fukamichi, J. Phys.: Condens. Matter **2**, 3381 (1990).

[3] E. P. Wohlfarth and P. Rhodes, Philos. Mag. **7**, 1817 (1962).

[4] M. Shimizu, J. Phys. (Paris) **43**, 155 (1982).

[5] H. Yamada, Phys. Rev. B **47**, 11 211 (1993).

[6] S. Hirooka and M. Shimizu, J. Phys. F **18**, 2223 (1988).

[7] S. Hirooka and M. Shimizu, J. Phys. F **18**, 2237 (1988).

[8] J. A. Hertz and M. A. Klenin, Phys. Rev. B **10**, 1084 (1974); Physica B **91**, 49 (1977).

[9] S. Hirooka and M. Shimizu, J. Phys. F **18**, L127 (1988).