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2000 J. Phys.: Condens. Matter 12 5669

(http://iopscience.iop.org/0953-8984/12/26/313)

J. Phys.: Condens. Matter **12** (2000) 5669–5679. Printed in the UK PII: S0953-8984(00)11112-9

Effects of zero-point spin fluctuation on the metamagnetism in YCo_2 and $LuCo_2$

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Received 19 January 2000

Abstract. Metamagnetism in YCo_2 and $LuCo_2$ is investigated in the spin-fluctuation theory, considering the contribution from both the zero-point and the thermal spin fluctuation. The metamagnetic transition at low temperature and the temperature dependence of the paramagnetic susceptibility at finite temperature are investigated in a self-consistent numerical calculation based on the spin-fluctuation theory and good agreements with experimental results have been obtained in a systematic way. It is shown that the effect of strong enhancement of the paramagnetic susceptibility and the electronic specific coefficient at low temperature can be explained satisfactorily by taking into consideration the zero-point spin fluctuation.

1. Introduction

The itinerant-electron metamagnetic transition induced by an external magnetic field in d-electron systems is a very interesting phenomenon and it has been investigated from both experimental and theoretical 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 $LuCo_2$ (Goto *et al* 1990, 1994, Sakakibara *et al* 1990). Moreover, these materials are strongly exchange-enhanced paramagnets and show a broad maximum in the temperature dependence of the paramagnetic susceptibility at room temperature. In particular, it is observed in the pseudo-binary compounds $Y(Co_{1-x}Al_x)_2$ that both the critical field B_c of the metamagnetic transition and the susceptibility-maximum temperature T_{max} decrease with increasing concentration of Al, and a ferromagnetic state is stabilized at a certain concentration. More recently, similar phenomena have been observed in $Lu(Co_{1-x}Gd_x)_2$ (Saito *et al* 1997).

Theoretically, the electronic structure and magnetism in cubic Laves phase transition metal compounds have been investigated systematically by Yamada and co-workers (Yamada 1988). Moreover, Yamada has discussed itinerant-electron metamagnetism at finite temperature within the phenomenological theory of spin fluctuations based on the Ginzburg–Landau free-energy functional and explained the universal properties observed in the magnetic materials showing the metamagnetic transition (Yamada 1993). It has been found that the itinerant-electron metamagnetism originates from the characteristic shape of the density of the electronic states (DOS) in these metamagnetic materials, which has a valley structure near the Fermi energy.

Incidentally, it has been believed that itinerant-electron metamagnetism at low temperature can be described within the Stoner theory, i.e. in the Hartree–Fock theory. In the conventional spin-fluctuation theory, only thermally excited spin fluctuations have been taken into consideration and the effects of zero-point spin fluctuation have been left out of consideration.

0953-8984/00/265669+11$30.00

As pointed out by Solontsov and Wagner (1994, 1995), it has become commonplace to say that they give rise to the temperature-independent effects which may be renormalized in the effective model parameters, e.g., in the exchange interaction constant. However, the strong enhancement of the paramagnetic susceptibility and the electronic specific heat coefficient at low temperature cannot be explained by the use of the effective value of the Coulomb interaction U in the Hartree–Fock theory, as discussed in the next section. In fact, they reach $\chi_p^{exp}/\chi_p^{band} \sim 26$ and $\gamma_e^{exp}/\gamma_e^{band} \sim 6$ for YCo_2, as obtained by the use of the DOS given by Tanaka and Harima (1998). Here, χ_p^{exp} and γ_e^{exp} are the experimental values of the paramagnetic susceptibility and the electronic specific heat coefficient of YCo_2 and χ_p^{band} and γ_e^{band} are the ones calculated from the DOS. Recently, the importance of the zero-point spin fluctuation has been pointed out (Solontsov and Wagner 1994, 1995, Lacroix *et al* 1996). Solontsov and Wagner have presented a generalized theory of the quantum Ginzburg–Landau approach to account for the large zero-point spin-fluctuation and spin-anharmonicity effects over a wide temperature range both below and far above the Curie temperature. Their theory has been applied to weak itinerant magnets such as MnSi and the itinerant-electron antiferromagnetism of $Y(Sc)Mn_2$ (Lacroix *et al* 1996).

Previously, we presented a spin-fluctuation theory of itinerant-electron metamagnetism (Nishiyama and Hirooka 1997) which is considered to be an extension of the theory of metamagnetism given by Yamada (1993) to the case with large amplitudes of the moment and spin fluctuation. However, the static approximation is used and so the zero-point spin fluctuation is not considered there. In this work the metamagnetism of YCo_2 and $LuCo_2$ at low and finite temperature will be investigated on the basis of the spin-fluctuation theory considering both the zero-point and the thermal spin fluctuation which is based on our previous work (Hirooka and Shimizu (1988a, b), hereafter referred as I). In the next section the Hartree–Fock theory is applied to these systems at zero temperature and it is shown that it does not yield good results in the investigation of the metamagnetic transition for these systems. In section 3, the work of I is extended to the multi-band system and a summary of our spin-fluctuation theory is given. Calculated results for the metamagnetism of YCo_2 and $LuCo_2$ at zero and finite temperature are given in the final section and a discussion is given in the remainder.

2. Difficulties in the Hartree–Fock theory

This section will be devoted to the exposition of some difficulties in the Hartree–Fock theory of metamagnetism of YCo_2 at low temperature. In this section and in the following sections it is assumed that only d electrons of a Co atom in YCo_2 contribute to the metamagnetism. The high-field magnetization $m^{Co}(h)$ is obtained from a self-consistent equation:

$$m^{Co}(h) = \int_{-\infty}^{\zeta_0} \left[\rho_+^{Co}(\omega) - \rho_-^{Co}(\omega)\right] \mathrm{d}\omega \tag{1}$$

where

$$\rho_\sigma^{Co}(\omega) = \rho^{Co}\left(\omega + \sigma h + \sigma \frac{U}{2} m^{Co}\right)$$

is the DOS of Co with $\sigma = +$ or $-$, h an external magnetic field and U the Coulomb interaction of d electrons in Co atoms. On the other hand, the paramagnetic susceptibility χ_p^{Co} at zero temperature is given in the Hartree–Fock theory as follows:

$$\chi_p^{Co} = \frac{2\rho^{Co}(\zeta_0)}{1 - U\rho^{Co}(\zeta_0)} \tag{2}$$

where ζ_0 is the Fermi energy. Now, many calculations of the electronic structure of YCo_2 have been carried out. In figure 1 the DOS calculated recently by Tanaka and Harima (1998) is shown. The DOS calculated by various methods are very similar to each other and characteristic as regards the shape—which consists of two sharp peaks mainly for d states of the Co atom—and the Fermi energy ζ_0 is in the valley, as shown in figure 1. However, the height of the DOS at the Fermi energy, the bandwidth and so on are quantitatively different. For example, we have $\rho^{Co}(\zeta_0) = 1.44\ eV^{-1}$/(atom spin) in the tight-binding method (Yamada 1988), $\rho^{Co}(\zeta_0) = 0.47\ eV^{-1}$/(atom spin) in the self-consistent APW method (Aoki and Yamada 1989) and $\rho^{Co}(\zeta_0) = 0.55\ eV^{-1}$/(atom spin) in the full-potential linear APW method (Tanaka and Harima 1998). In the following numerical calculation we will use the DOS given by Tanaka and Harima. The high-field magnetization and paramagnetic susceptibility at zero temperature for YCo_2 have been calculated from equations (1) and (2). In figure 2 the high-field magnetization is shown by a broken line in the case of $U = 1.084$ eV. It yields $B_c \simeq 70$ T which is nearly equal to an experimental value (Goto *et al* 1990, 1994). The experimental results for the magnetization are shown in figure 2 by a dotted line. In table 1 calculated

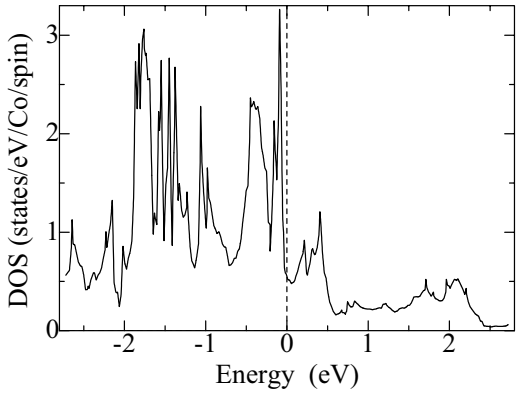

Figure 1. (a) The Co d component of the electronic density of states (DOS) of YCo_2 (Tanaka and Harima 1998). The vertical broken line shows the position of the Fermi energy.

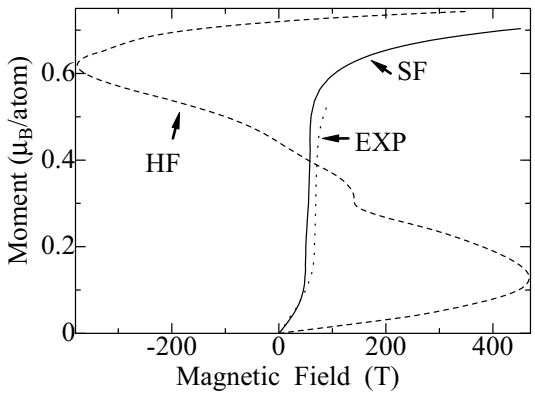

Figure 2. The high-field magnetization of YCo_2 at zero temperature: results calculated within the Hartree–Fock theory for $U = 1.084$ eV (broken line, HF) and in the spin-fluctuation theory for $U = 1.068$ eV and $\bar{q}_c = 0.491$ (solid line, SF); the dotted line (EXP) shows the experimental result (Goto *et al* 1990, 1994).

Table 1. Calculated paramagnetic susceptibilities χ_p^{Co} and the critical fields B_c of YCo_2 at zero temperature within the Hartree–Fock theory for various values of the Coulomb interaction U obtained by the use of the DOS given by Tanaka and Harima (1998).

	U (eV)	B_c (T)	χ_p^{Co} (10^{-3} emu mol^{-1})
Metamagnetic transition	1.084	70.0[a]	0.178
Ferromagnetic instability	1.105 (U_c^{ET})	0.0	0.182
$\chi_p^{\mathrm{Co}} = \chi_p^{exp}$	1.744		1.83[a]
Stoner condition	1.818 (U_c^{ST})		∞

[a] Experimental values (Goto *et al* 1990, 1994).

paramagnetic susceptibilities χ_p^{Co} are shown for four values of U which are chosen as giving $B_c \simeq 70.0$ T, $B_c = 0.0$ T, $\chi_p^{\mathrm{Co}} = \chi_p^{exp}$ and $U\chi_p^{\mathrm{Co}} = 1$. Here, $\chi_p^{exp} = 1.83 \times 10^{-3}$ emu mol^{-1} is an experimental value and $U\chi_p^{\mathrm{Co}} = 1$ is the Stoner condition.

As is well known, the itinerant-electron metamagnetic materials can satisfy the extended condition of ferromagnetism given by Shimizu (1964, 1965) because of the valley structure in the DOS. This condition is given by $U \geqslant U_c^{ET}$ and $U_c^{ET} \leqslant U_c^{ST}$ where U_c^{ST} is determined by the Stoner condition. So, because $U \leqslant U_c^{ET}$ in the stable paramagnetic state, we have

$$\chi_p^{\mathrm{Co}} \leqslant \chi_p^{ET} = 2\rho^{\mathrm{Co}}(\zeta_0)/[1 - U_c^{ET}\rho^{\mathrm{Co}}(\zeta_0)].$$

Now, the use of the DOS given by Tanaka and Harima (1998) gives $U_c^{ET} = 1.105$ eV, $U_c^{ST} = 1.818$ eV and $\chi_p^{ET} = 0.182 \times 10^{-3}$ emu mol^{-1} as shown in table 1. Finally, we have $\chi_p^{\mathrm{Co}}/\chi_p^{band} \leqslant 2.6$, where $\chi_p^{band} = 2\rho^{\mathrm{Co}}(\zeta_0)$. This suggests to us that the strong enhancement of the paramagnetic susceptibility of YCo_2 cannot be explained by adjusting the effective value of U within the Hartree–Fock theory. As regards the metamagnetic transition, the calculated high-field magnetization curve shows too large a hysteresis compared with experimental results as shown in figure 2. In general, the shape of the magnetization curve seems to be too sensitive to the shape of the DOS near the Fermi energy in the Hartree–Fock theory.

3. Spin-fluctuation theory of itinerant-electron metamagnetism

In this section, results from I (Hirooka and Shimizu 1988a, b) are extended to the multi-band case and a summary of them will be given sufficient for the needs of the numerical calculation in this work. We assume as the multi-band Hubbard Hamiltonian the following form taking into consideration Hund coupling:

$$H = \sum_{l,m,i,j,\sigma} t_{ij}^{lm} a_{li\sigma}^{\dagger} a_{mj\sigma} - \frac{U}{4} \sum_i \left(\sum_l \vec{\sigma}_{li} \right)^2 \tag{3}$$

where $a_{li\sigma}^{\dagger}$ and $a_{mj\sigma}$ are creation and annihilation operators for electrons in the orbital state l, at lattice site i and in spin state σ ($\sigma = +$ or $-$). Moreover, t_{ij}^{lm} are hopping energies, U is the Coulomb interaction, $\vec{\sigma}_{li} = (\sigma_{li}^x, \sigma_{li}^y, \sigma_{li}^z)$, $\sigma_{li}^x = a_{li+}^{\dagger} a_{li-} + a_{li-}^{\dagger} a_{li+}$, $\sigma_{li}^y = -\mathrm{i}(a_{li+}^{\dagger} a_{li-} - a_{li-}^{\dagger} a_{li+})$ and $\sigma_{li}^z = a_{li+}^{\dagger} a_{li+} - a_{li-}^{\dagger} a_{li-}$. In the Hamiltonian equation (3), the spin fluctuations are considered exclusively and the charge fluctuations are ignored. Now, the interaction term in equation (3) is expressed in the harmonic form of the spin density. Thus, we can apply the Stratonovich–Hubbard transformation to it and use the functional integral formalism as in I. By following this procedure, we can get the desired form of the result for the multi-band system.

In short, the interaction term in equation (3) suggests to us that d electrons in the ith site feel magnetic fields

$$\vec{h}_i = \frac{U}{2}\sum_l \vec{\sigma}_{li}$$

caused by the other d electrons. In the functional integral formalism, corresponding to the operators $\vec{h}_i$, the magnetic fields $\vec{\eta}_i(u)$ for the c-number are introduced and the Hamiltonian equation (3) is transformed into a one-body Hamiltonian $H_f(u)$ as follows:

$$H_f(u) = \sum_{l,m,i,j,\sigma} t_{ij}^{lm} a_{li\sigma}^{\dagger}(u) a_{mj\sigma}(u) - \sum_i \vec{\eta}_i(u)\cdot\sum_l \vec{\sigma}_{li}(u) \tag{4}$$

where $A(u) = \mathrm{e}^{uH_0} A \mathrm{e}^{-uH_0}$ (A is an operator), u is the imaginary time and

$$H_0 = \sum_{l,m,i,j,\sigma} t_{ij}^{lm} a_{li\sigma}^{\dagger} a_{mj\sigma}.$$

The magnetic field $\vec{\eta}_i(u)$ consists of a constant part $\vec{\eta}_0$ and a fluctuating part $\delta\vec{\eta}_i(u)$ which obey a Gaussian distribution as follows:

$$\vec{\eta}_i(u) = \vec{\eta}_0 + \delta\vec{\eta}_i(u). \tag{5}$$

The constant field $\vec{\eta}_0$ is determined by

$$\eta_0 = \frac{U}{2}\langle\langle M_i^z(u)\rangle_T\rangle_{\delta\eta} \tag{6}$$

where $M_i^z(u) = \sum_l \sigma_{li}^z(u)$. Moreover, the symbol $\langle\cdots\rangle_T$ means the thermal average under the Hamiltonian $H_f(u)$ and $\langle\cdots\rangle_{\delta\eta}$ the average with respect to the Gaussian distribution for the fluctuating field $\delta\eta_i(u)$. The variances of the Gaussian distribution are determined from

$$\langle\delta\eta_i^{\alpha}(u)\,\delta\eta_j^{\alpha'}(u')\rangle = \frac{U^2}{4\beta}\chi_{\alpha\alpha'}^{ij}(u,u') \qquad \alpha,\alpha' = x,y,z \tag{7}$$

where

$$\chi_{\alpha\alpha'}^{ij}(u,u') = \beta\left\langle\left\{T[M_i^{\alpha}(u)M_j^{\alpha'}(u')]\right\}_c\right\rangle_{\delta\eta}$$

is a generalized susceptibility, β an inverse temperature, T a time-ordering operator and

$$\{T[A(u)B(u)]\}_c \equiv \langle T[A(u)B(u)]\rangle_T - \langle A(u)\rangle_T\langle B(u)\rangle_T.$$

Finally, the many-body problem described by the Hamiltonian in equation (3) is transformed to a one-body problem described by the Hamiltonian $H_f(u)$ in which each d electron moves in a constant field $\vec{\eta}_0$ and a random fluctuating field $\delta\vec{\eta}_i(u)$ caused self-consistently by themselves, from equations (6) and (7).

However, there are still more difficulties in realistic numerical calculations of these results and further approximations are required. Following Hertz and Klenin (1974, 1977) as in I, we adopt the local mean-field approximation for the variances in equation (7). Then, local correlations of the spin fluctuation in space and time, which dominate the variances in equation (7), are taken into consideration exclusively. They are given as $\langle\delta\eta_i^{\alpha}(u)\,\delta\eta_i^{\alpha'}(+u)\rangle = \chi_{\alpha\alpha'}^{ii}(u,u)$ and are not dependent on the site i and the imaginary time u because of the translational symmetry for the space and the time. Moreover, we can actually do slightly better than treating the correlations as purely local by taking $\delta\vec{\eta}_i(u)$ in equation (4) constant in time and uniform in space as $\delta\vec{\eta}$. This treatment is justified for slowly varying components of the spin fluctuation in space and time.

The results are summarized as follows. When each d electron moves in the magnetic field, $\vec{\eta} = (\delta\eta_t, \eta_0 + \delta\eta_z)$ is caused by the other d electrons, where η_0 is a uniform and static field and

$\delta\vec{\eta} = (\delta\eta_t, \delta\eta_z)$ are random magnetic fields generated by the spin fluctuation of d electrons. If an external field h exists, we have $\vec{\eta} = (\delta\eta_t, h + \eta_0 + \delta\eta_z)$. Here the subscripts t and z denote the transverse and the longitudinal components, respectively. The distribution of fluctuating fields is a Gaussian and it is given as

$$g(\delta\eta) \propto \exp\left\{-\frac{1}{2}\left[\left(\frac{\delta\eta_t}{\sigma_t}\right)^2 + \left(\frac{\delta\eta_z}{\sigma_z}\right)^2\right]\right\}. \tag{8}$$

By the use of the dynamical susceptibility $\chi_\alpha(\vec{q}, \omega_l)$ with the Matsubara frequencies $\omega_l = 2\pi l/\beta$, the variances, σ_t and σ_z, of the distribution $g(\delta\eta)$ of fluctuating fields are given as

$$\begin{aligned}\sigma_\alpha^2 &= \frac{U^2}{4\beta}\sum_{\vec{q},\omega_l}\chi_\alpha(\vec{q}, \omega_l) \\ &= \frac{U^2}{4\pi}\sum_{\vec{q}}\int_0^\infty \mathrm{Im}\,\chi_\alpha(\vec{q}, \omega)\,\mathrm{d}\omega + \frac{U^2}{2\pi}\sum_{\vec{q}}\int_0^\infty \frac{1}{\mathrm{e}^{\beta\omega} - 1}\,\mathrm{Im}\,\chi_\alpha(\vec{q}, \omega)\,\mathrm{d}\omega \\ &\equiv (\sigma_\alpha^0)^2 + (\sigma_\alpha^T)^2 \qquad \alpha = z, t\end{aligned} \tag{9}$$

where the first term σ_α^0 denotes the zero-point spin fluctuation and the second term σ_α^T denotes the thermal spin fluctuation. The dynamical susceptibilities $\chi_z(\vec{q}, \omega_l)$ and $\chi_t(\vec{q}, \omega_l) = [\chi_{-+}(\vec{q}, \omega_l) + \chi_{+-}(\vec{q}, \omega_l)]/2$ are given as

$$\chi_s(\vec{q}, \omega) = \frac{2\hat{\chi}_s(\vec{q}, \omega)}{1 - U\hat{\chi}_s(\vec{q}, \omega)} \qquad s = z, +-, -+ \tag{10}$$

where $\hat{\chi}_z(\vec{q}, \omega)$, $\hat{\chi}_{+-}(\vec{q}, \omega)$ and $\hat{\chi}_{-+}(\vec{q}, \omega)$ are obtained by averaging the longitudinal and the transverse dynamical susceptibilities of a non-interacting electron system in the presence of the magnetic field $\vec{\eta}$ with respect to the distribution $g(\delta\eta)$. The expressions for them are given in I and their uniform and static components are written in the form

$$\begin{aligned}\hat{\chi}_z &\equiv \hat{\chi}_z(0, 0) = \left\langle \chi_{0z}(\eta)\cos^2\theta + \chi_{0t}(\eta)\sin^2\theta \right\rangle_{\delta\eta} \\ \hat{\chi}_t &\equiv \hat{\chi}_{+-}(0, 0) = \hat{\chi}_{-+}(0, 0) = \frac{1}{2}\left\langle \chi_{0z}(\eta)\sin^2\theta + \chi_{0t}(\eta)(1 + \cos^2\theta)\right\rangle_{\delta\eta}\end{aligned} \tag{11}$$

where $\cos\theta = \eta_z/\eta$, $\sin\theta = \delta\eta_t/\eta$, $\eta = (\delta\eta_t^2 + \eta_z^2)^{1/2}$, $\eta_z = h + \eta_0 + \delta\eta_z$ and the symbol $\langle\cdots\rangle_{\delta\eta}$ denotes the average over the Gaussian distribution $g(\delta\eta)$ in equation (8). Moreover, $\chi_{0z}(\eta)$ and $\chi_{0t}(\eta)$ in equation (11) are given as

$$\chi_{0z}(\eta) = \frac{2\chi_{0+}(\eta)\chi_{0-}(\eta)}{\chi_{0+}(\eta) + \chi_{0-}(\eta)}$$

$$\chi_{0t}(\eta) = \frac{m(\eta)}{2\eta}$$

where

$$\chi_{0\sigma}(\eta) = -\int_{-\infty}^{\infty}\frac{\partial f(\omega)}{\partial\omega}\rho(\omega - \sigma\eta)\,\mathrm{d}\omega \qquad \sigma = +, -$$

$$m(\eta) = \int_{-\infty}^{\infty} f(\omega)[\rho(\omega + \eta) - \rho(\omega - \eta)]\,\mathrm{d}\omega.$$

$f(\omega) = [\exp\{\beta(\omega - \zeta) + 1]^{-1}$ is the Fermi distribution function and $\rho(\omega)$ the DOS. The constant molecular field η_0, which is induced by the external magnetic field h, is determined self-consistently by

$$\eta_0 = \frac{U}{2}\langle m(\eta)\cos\theta\rangle_{\delta\eta} \tag{12}$$

which is obtained from the condition of thermodynamic equilibrium, where $M_z(h) = \langle m(\eta)\cos\theta\rangle_{\delta\eta}$ is the magnetization under the external magnetic field h. By putting $h = 0$ and so $\eta_0 = 0$ in equations (10) and (11), the paramagnetic susceptibility is obtained as

$$\begin{aligned} \chi_p &= \frac{2\hat{\chi}_p}{1 - U\hat{\chi}_p} \\ \hat{\chi}_p &= \frac{1}{3}\langle 2\chi_{0t}(\delta\eta) + \chi_{0z}(\delta\eta)\rangle_{\delta\eta}\,. \end{aligned} \tag{13}$$

A mass enhancement factor γ_{ehs} is defined by

$$\gamma_{ehs} = \frac{\langle\rho\rangle}{\rho(\zeta_0)} \tag{14}$$

where

$$\langle\rho\rangle \equiv \frac{1}{2}\langle\rho(\zeta - \delta\eta) + \rho(\zeta + \delta\eta)\rangle_{\delta\eta} \tag{15}$$

and $\rho(\zeta_0)$ is the value of the DOS at the Fermi energy ζ_0. Equations (8)–(15) form a self-consistent set of equations for use in the numerical calculation.

Before proceeding to the next section, we briefly refer to the effects of the zero-point spin fluctuation on the paramagnetic susceptibility and the electronic specific heat coefficient at low temperature. The paramagnetic susceptibility χ_p in equation (13) and the mass enhancement factor γ_{ehs} in equation (14) can be expanded in a Taylor's series in powers of σ_0^2 at $T = 0$ as

$$\begin{aligned} \frac{1}{\chi_p} &\simeq \frac{1}{2}\left(\frac{1}{\rho_0} - U\right) - \frac{5}{12{\rho_0}^2}\left[\rho_0^{(2)} - \frac{3(\rho_0^{(1)})^2}{\rho_0}\right]{\sigma_0}^2 \\ \gamma_{ehs} &\simeq 1 + \frac{1}{2\rho_0}\left[\rho_0^{(2)} - \frac{(\rho_0^{(1)})^2}{4\rho_0}\right]{\sigma_0}^2 \end{aligned} \tag{16}$$

where $\rho_0^{(n)} = [\mathrm{d}^n\rho(\omega)/\mathrm{d}\omega^n]_{\omega=\zeta_0}$ and ζ_0 is the Fermi energy for $\sigma_0 = 0$. The first terms of equations (16) correspond to results within the Hartree–Fock theory. In general the second terms of equation (16) are, respectively, negative and positive for the systems having a valley structure in the DOS near the Fermi energy such as YCo_2. So, it is found that the zero-point spin fluctuation enhances the paramagnetic susceptibility and the electronic specific heat coefficient at low temperature in these systems.

4. Calculated results and discussion

It is considered that the metamagnetism in YCo_2 and $LuCo_2$ is dominated by the spin magnetism of Co atoms; thus in the following numerical calculation the contributions from Y atoms and Lu atoms to the metamagnetism are neglected for simplicity and so only the spin magnetism of Co atoms is taken into consideration. Now, it is very difficult in reality to get $\hat{\chi}_s(\vec{q},\omega)$ in equation (10) concretely and also to perform a numerical estimation of the integral in equation (9). So, we approximate $\hat{\chi}_s(\vec{q},\omega)$ in equation (10) as

$$\begin{aligned} &\hat{\chi}_s(\vec{q},\omega) \equiv \hat{\chi}_s(0,0)F_s(\vec{q},\omega) \\ &F_s(\vec{q},\omega) = \frac{\hat{\chi}_s(\vec{q},\omega)}{\hat{\chi}_s(0,0)} \approx \frac{\chi_s^f(\vec{q},\omega)}{\chi_s^f(0,0)} \qquad s = z, +-, -+ \end{aligned} \tag{17}$$

where $\chi_z^f(\vec{q},\omega)$, $\chi_{+-}^f(\vec{q},\omega)$ and $\chi_{-+}^f(\vec{q},\omega)$ are dynamical susceptibilities of a non-interacting free-electron gas under the magnetic field $\eta_0 + h$ at $T = 0$ and so they are given analytically by

the use of the Lindhard function. Under the approximation in equation (17), the effects of the spin fluctuation are included through $\hat{\chi}_s(0,0)$ defined in equation (11), and $F_s(\vec{q},\omega)$, which is estimated by the use of the dynamical susceptibilities of the free-electron gas, expresses the dependencies on the wave vector $\vec{q}$ and the frequency ω of the dynamical susceptibilities. As discussed in the previous section, we have adopted the local mean square approximation (Hertz and Klenin 1974, 1977) where the long-wavelength and the low-frequency components of the spin fluctuation are taken into consideration exclusively and the short-wavelength and the high-frequency components are cut off. The substitution of the dynamical susceptibilities of a free-electron gas is considered to be appropriate in this long-wavelength and low-frequency region.

Thus, on introducing the cut-off wavenumber q_c in the integral in equation (9) with respect to $\vec{q}$, the variances $(\sigma_\alpha)^2$ in equation (9) are approximated as

$$(\sigma_\alpha)^2 = \frac{3}{\pi} N_e U^2 \hat{\chi}_\alpha \int_0^{\bar{q}_c} \bar{q}^2 \, \mathrm{d}\bar{q} \int_0^\infty \left(\frac{1}{\mathrm{e}^{\beta\omega} - 1} + \frac{1}{2} \right) \mathrm{Im}\, \Gamma_\alpha(\vec{q},\omega) \, \mathrm{d}\omega \tag{18}$$

where $\alpha = z, t$ and

$$\Gamma_z(\vec{q},\omega) = \frac{F_z(\vec{q},\omega)}{1 - U\hat{\chi}_z F_z(\vec{q},\omega)}$$

$$\Gamma_t(\vec{q},\omega) = \frac{1}{2}\left[\frac{F_{+-}(\vec{q},\omega)}{1 - U\hat{\chi}_t F_{+-}(\vec{q},\omega)} + \frac{F_{-+}(\vec{q},\omega)}{1 - U\hat{\chi}_t F_{-+}(\vec{q},\omega)} \right].$$

Here, $\hat{\chi}_z$ and $\hat{\chi}_t$ are defined in equation (11), $\bar{q}_c = q_c/k_f$ (k_f is the Fermi wavenumber of a free-electron gas) and N_e is the number of free electrons per atom and per spin. The parameters required for the numerical calculation are the number N_e and the Fermi energy ζ_f of the free electrons. The hole description is employed in the numerical estimation of equation (18). So, N_e is equal to the number of d holes which is given as $N_e \simeq 1.35$/(atom spin) (Tanaka and Harima 1998). The Fermi energy ζ_f is determined from the relation $\zeta_f = \frac{3}{2} N_e / \langle \rho^{\mathrm{Co}} \rangle$ for the free electron, where $\langle \rho^{\mathrm{Co}} \rangle$ is the averaged partial DOS for a Co atom in YCo_2 as given by equation (15). Finally, we have two adjustable parameters, namely, the Coulomb interaction U and the cut-off wavenumber $\bar{q}_c$, in the numerical calculation. The adjustable parameters U and $\bar{q}_c$ were made as the calculated values of the paramagnetic susceptibility and the critical field B_c of the metamagnetic transition of YCo_2 at low temperature were made as close as possible to the experimental ones. $U = 1.068$ eV and $\bar{q}_c = q_c/k_f = 0.491$ were chosen as a result. The value of $U = 1.068$ eV is somewhat large compared to the value $U = 0.7$ eV obtained by photoemission spectroscopy (PES) and inverse PES experiments (Duo *et al* 1994).

The magnetization at $T = 0$ of YCo_2 has been calculated from equation (9) and is shown by a solid line in figure 2 together with the experimental results (Goto *et al* 1990, 1994) and the ones calculated within the Hartree–Fock theory given in section 2. This gives the critical field of the metamagnetic transition $B_c \sim 54$ T. The paramagnetic susceptibility at $T = 0$ has been calculated as $\chi_p = 1.70 \times 10^{-3}$ emu mol^{-1} from equation (13); this gives the enhancement of the paramagnetic susceptibility $\chi_p/\chi_p^{band} \simeq 24.1$ where $\chi_p^{band} = 2\rho^{\mathrm{Co}}(\zeta_0)$. The mass enhancement factor in equation (14) is obtained as $\gamma_{ehs} \simeq 2.63$ which is less than half of the experimental value $\gamma_{ehs}^{exp} \simeq 5.9$ (Tanaka and Harima 1998). The disagreement with the experimental result will be lessened to some extent by considering the mass enhancement originating from the electron–phonon interaction. Tanaka and Harima have obtained $\gamma_{ehs} \simeq 5.9$ from the self-energy of Co d electrons calculated in the second-order perturbation of U by the use of $U = 1.8$ eV. However, it is noted that such a large value of U makes the paramagnetic state of YCo_2 unstable as discussed in section 2. It is seen that the zero-point spin fluctuation strongly enhances the paramagnetic susceptibility and the electronic specific heat coefficient at low temperature and makes the hysteresis of the magnetization curve calculated within the

Hartree–Fock theory, which is too large compared with the experimental one, remarkably small. Figure 3 gives the amplitudes of the spin fluctuations for the magnetic field—where a broken line, a dotted line and a solid line denote, respectively, the longitudinal component $(\delta m_0^z)^2$, the transverse component $(\delta m_0^t)^2$ and their sum $\delta m_0^2 = 2(\delta m_0^t)^2 + (\delta m_0^z)^2$—which are defined as $\sigma_z^2 = (U/2)(\delta m_0^z)^2$ and $\sigma_t^2 = (U/2)(\delta m_0^t)^2$. The longitudinal spin fluctuations are properly suppressed under the stronger external magnetic field as seen from figure 3. The temperature dependence of the paramagnetic susceptibility of YCo_2 has been calculated from equation (13) and the result is shown in figure 4 together with the experimental ones (Goto *et al* 1990, 1994). It gives $T_{max} \simeq 275$ K for YCo_2. Here, T_{max} is defined as the temperature at which a paramagnetic susceptibility shows a maximum in the temperature dependence. Figure 5 gives the amplitudes of the spin fluctuations for the temperature and a broken line, a dotted line and a solid line denote, respectively, the zero-point component δm_0^2, the thermal component δm_T^2 and their sum δm^2. It is clearly seen from figure 5 that the strong enhancement of the paramagnetic susceptibility at $T \sim 0$ is caused by the zero-point spin fluctuation and that a broad maximum in the temperature dependence of the paramagnetic susceptibility at

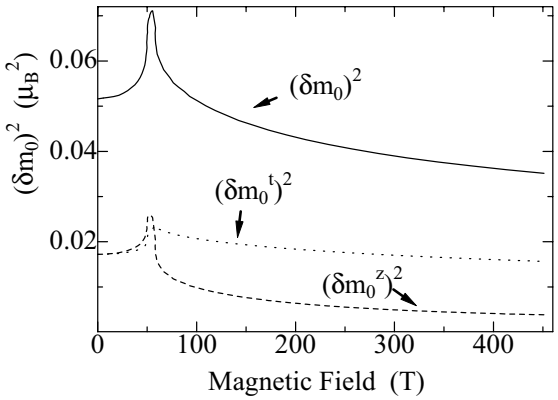

Figure 3. Magnetic field dependences of the amplitudes of the zero-point spin fluctuation (solid line), the longitudinal component (broken line) and the transverse component (dotted line) at zero temperature for $U = 1.068$ eV and $\bar{q}_c = 0.491$.

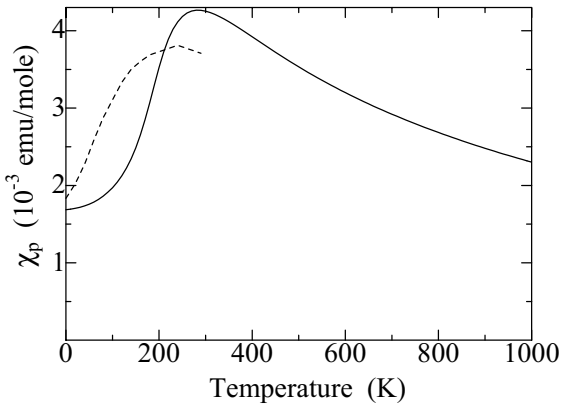

Figure 4. Temperature dependences of the paramagnetic susceptibility for $U = 1.068$ eV and $\bar{q}_c = 0.491$ (solid line). The broken line shows the experimental result (Goto *et al* 1990, 1994).

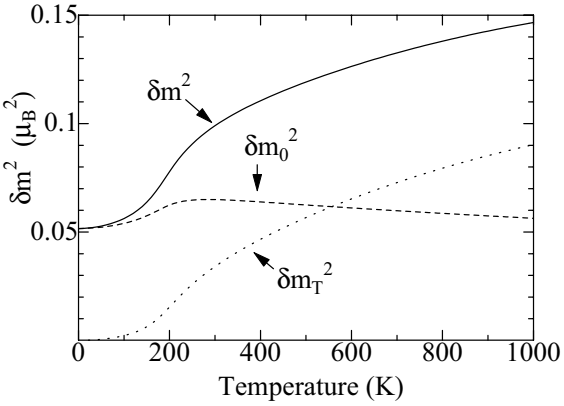

Figure 5. Temperature dependences for the amplitudes of the total spin fluctuation (solid line), the zero-point component (broken line) and the thermal component (dotted line) for $U = 1.068$ eV and $\bar{q}_c = 0.491$.

$T \sim T_{max}$ is caused mainly by the thermal spin fluctuation. At the same time, the amplitude of the zero-point spin fluctuation becomes dependent on the temperature as a result of the coupling to the thermal spin fluctuation.

We have carried out similar calculations for $LuCo_2$. A calculated DOS for $LuCo_2$ has been given by Tanaka and Harima (1998) and it resembles that of YCo_2. The adjustable parameters are chosen as $\bar{q}_c = 0.503$ and $U = 1.037$ eV, which are close to those of YCo_2. Calculated results are shown in table 2 together with those for YCo_2.

Table 2. Calculated results for physical quantities in the metamagnetism of YCo_2 for $\bar{q}_c = 0.491$ and $LuCo_2$ for $\bar{q}_c = 0.503$.

		U (eV)	B_c (T)	δm_0 (μ_B/atom)	γ_{ehs}	$\chi_p(T=0)$ (10^{-3} emu mol^{-1})	T_{max} (K)	$\chi_p(T=T_{max})$ (10^{-3} emu mol^{-1})
YCo_2	(theory)	1.068	53.6	0.23	2.63	1.70	275	4.26
	(experiment)		70.0		5.93	1.83	240	3.81
$LuCo_2$	(theory)	1.037	62.1	0.23	2.97	1.31	378	3.28
	(experiment)		73.6		4.67	1.41		

Now, it is conventional for the effect of the zero-point fluctuation to be regarded as renormalized with respect to the effective value of the Coulomb interaction U. However, it has been shown in this work that the zero-point spin fluctuation seriously changes the single-electron spectrum and so the DOS rather than the effective value of the Coulomb interaction. Recently, the importance of the zero-point spin fluctuation has been pointed out in various works (Solontsov and Wagner 1994, 1995, Lacroix *et al* 1996). Solontsov and Wagner showed that the quantum dynamical effects of the zero-point spin fluctuation give rise to the strong spin-anharmonicity effects neglected in the conventional spin-fluctuation theory of weak itinerant-electron magnetism. They questioned the common view that the zero-point spin fluctuation gives rise to temperature-independent effects which may be included in the effective model parameters, e.g., in the exchange interaction constant. A local magnetic moment at zero temperature is one of the physical quantities which measure the importance of the zero-point spin fluctuation. Solontsov and Wagner estimated the zero-temperature values for the local magnetic moments of weak itinerant magnets, e.g., $\delta m_0 = 0.85\mu_B$/atom (μ_B is the Bohr

magneton) for MnSi, which may be compared with polarized neutron scattering data (Ziebeck *et al* 1986). In this work, $\delta m_0 \sim 0.23\mu_B$/Co has been obtained for the values of the local magnetic moments at $T = 0$ for YCo_2 and $LuCo_2$. It is considered that itinerant-electron metamagnets such as YCo_2 and $LuCo_2$, where the paramagnetic susceptibility is strongly exchange enhanced, are typical systems as well as weak itinerant-electron magnets, for which the zero-point spin fluctuation plays important roles in the magnetism.

In conclusion, we have investigated the metamagnetism of YCo_2 and $LuCo_2$ by means of a self-consistent calculation based on the spin-fluctuation theory including the contribution of both the zero-point spin fluctuation and the thermal spin fluctuation, and have given a systematic explanation for the experimental results on itinerant-electron metamagnets. The itinerant-electron metamagnetism in these systems originates from the characteristic shape, namely the valley structure, of the DOS around the Fermi energy. Owing to this characteristic shape of the DOS, the zero-point spin fluctuation and the thermal spin fluctuation cause, respectively, the strong enhancements of the paramagnetic susceptibility and the electronic specific heat coefficient at low temperature and a broad maximum in the temperature dependence of the paramagnetic susceptibility at finite temperature.

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