Mem. Fac. Sci., Shimane Univ.,
17, pp. 39–45 Dec. 20, 1983

Spin Fluctuations in Weak Itinerant Ferromagnet Sc_3In*

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(Received September 3, 1983)

The magnetic field dependences of the magnetization, electrical resistivity and specific heat of a weak itinerant ferromagnet Sc_3In are discussed in particular with the self-consistent renormalization theory of spin fluctuations. The experimental results of these different physical quantities are consistently explained by making use of the same parameters within the framework of this theory.

§ 1. Introduction

The itinerant electron ferromagnetism has been one of the most attractive subjects in magetism for a decade. Owing to recent theoretical and experimental investigations on physical properties of ferromagnetic metals and alloys, the problem has been focused on the spin density fluctuations of correlated itinerant electrons.[1] Moriya and Kawabata have developed a theory which takes account of the coupling between the different modes of spin fluctuations in a self-consistent way — the self-consistent renormalization (SCR) theory.[2]

It is well known that Sc_3In is one of the best candidates of a weak itinerant electron ferromagnet containing no magnetic constituent elements. This compound has been studied by us in detail by using calorimetrical, electrical and nuclear magnetic resonance methods.[3–5] Furthermore, the importance of the effect of a magnetic field on spin fluctuations was first pointed out. These experimental studies on various physical properties have confirmed step by step the predictions of the SCR theory.

In this paper we will reexamine the applicability of the SCR theory by applying this theory to the magnetic field dependences of the magnetization, electrical resistivity and specific heat. And we will discuss whether the experimental results of these different physical quantities are explained consistently by making use of the same parameters within the framework of this theory.

§ 2. Extension of the SCR Theory in the Presence of an External Magnetic Field

The effect of an external magnetic field on spin fluctuations is considered within

* Part of this work was reported at the International Conference on Magnetism, Kyoto (1982); Y. Masuda: J. Magn. Magn. Mat. 31–34 (1983) 259.

the framework of the SCR theory. According to Moriya and Kawabata the correction to the Hartree-Fock free energy is expressed in terms of transverse dynamical susceptibility $\chi_{\rm MI}^{-+}(q, \omega)$ which is given as[2]

$$\chi_{\rm MI}^{-+}(q, \omega) = \frac{\chi_{\rm M0}^{-+}(q, \omega)}{1 - I\chi_{\rm M0}^{-+}(q, \omega) + \lambda_{\rm MI}(q, \omega)}, \tag{1}$$

where M is the magnetization, I the exchange interaction constant and $\lambda_{MI}(q, \omega)$ gives the correction to a random phase approximation. The requirement of self-consistency is that the static and long wave limit ($q \to 0$, $\omega \to 0$) of the transverse dynamical susceptibility $\chi_{\rm MI}^{-+}(0, 0)$ should coincide with the static transverse susceptibility χ which should be calculated from the free energy $F(M, T)$:

$$\begin{aligned} &1 - I\chi_{00}^{-+}(0, 0) + \lambda_{0I}(0, 0) = \chi_0/\chi && \text{for} \quad T > T_c, \\ &1 - I\chi_{\rm M0}^{-+}(0, 0) + \lambda_{MI}(0, 0) = 0 && \text{for} \quad T < T_c, \end{aligned} \tag{2}$$

where χ_0 is the susceptibility of the non-interacting electron system.

In the presence of an external magnetic field H, static transverse susceptibility is given as $\chi = M(H)/H$. So self-consistency condition should be replaced as

$$1 - I\chi_{\rm M0}^{-+}(0, 0) + \lambda_{\rm MI}(0, 0) = \frac{\chi_0}{\chi} = \frac{\chi_0}{M(H)/H}. \tag{3}$$

Neglecting the q- and ω-dependences of λ, eq. (1) becomes as

$$\chi_{\rm MI}^{-+}(q, \omega) = \frac{\chi_0}{2\alpha} \frac{f_{\rm M0}(q, \omega)}{(1+\delta)f_{\rm M0}(0, 0) - f_{\rm M0}(q, \omega)}, \tag{4}$$

where $\delta = \chi_0/\alpha\chi$, $\alpha = I\chi_0/2$ and $f_{\rm M0}(q, \omega) = 2\chi_{\rm M0}^{-+}(q, \omega)/\chi_0$ is the reduced transverse dynamical susceptibility of the non-interacting system under the given magnetization M. Using eq. (4) the correction to the Hartree-Fock free energy $F_{\rm HF}(M, T)$ is given as

$$\begin{aligned} \Delta F(M, T) &= F(M, T) - F_{\rm HF}(M, T) \\ &= -\frac{2}{\pi}\int_{-\infty}^{\infty} d\omega \, \mathrm{sgn}\, \omega \, n(|\omega|) H_1(\omega), \end{aligned} \tag{5}$$

$$H_1(\omega) = \frac{1}{2}\sum_q \left\{ \tan^{-1} \frac{f''_{\rm M0}(q, \omega)}{(1+\delta)f_{\rm M0}(0, 0) - f'_{\rm M0}(q, \omega)} - \alpha f''_{\rm M0}(q, \omega) \right\},$$

where $n(\omega)$ is the Bose factor and $f_{\rm M0}(q, \omega) = f'_{\rm M0}(q, \omega) + if''_{\rm M0}(q, \omega)$.

For weakly ferromagnetic metals, $f_{\rm M0}(q, \omega)$ can be expanded for small q and small $s = \omega/q$:[2]

$$\begin{aligned} f_{\rm M0}(q, \omega) = 1 &+ iC_1 s - Aq^2 - B_1 s^2 + \cdots + R_1\zeta\left[D_1 \frac{s}{q} + \cdots\right] \\ &- \frac{1}{2} R_1^2 \zeta^2 \left[F_1 + F_2\left(\frac{s}{q}\right)^2 + \cdots\right], \end{aligned} \tag{6}$$

where $\zeta=M/N$ is the reduced magnetization, $R_1=N/\chi_0\varepsilon_F$, N the total number of electrons, and ε_F is the Fermi energy. Other coefficients are determined for any given band structure near the Fermi surface. In the following, numerical calculations are made for the electron gas model, in which case the coefficients are given as

$$A=1/12,\quad B_1=1/4,\quad C_1=\pi/4,\quad D_1=1/2,\quad F_1=1/3,\quad F_2=1/2,\quad R_1=2/3. \tag{7}$$

§3. Results and Discussion

3.1 *Magnetization*

The experimental results of the magnetization for Sc_3In in magnetic fields of 0, 5 and 10 kOe are plotted in Fig. 1 as $\zeta=M/N$ vs T/T_c. The Curie temperature $T_c=5.5$ K is derived by extrapolating the experimental data in zero magnetic field to $M=0$. These results are compared with both the Stoner model based on the Hartree-Fock approximation and the SCR theory.

On the basis of the Stoner model, the magnetization as functions of temperature and magnetic field is given as[6]

$$\zeta^2=\zeta_0^2\{1-(T/T_c)^2\}+k(H)\zeta_0^2/\zeta, \tag{8}$$

where $k(H)=2\chi_0 H/N$ and ζ_0 is the reduced magnetization at $T=0$ and $H=0$.

Meanwhile, the magnetization based on the SCR theory is obtained from the free energy as $\partial F(M,T)/\partial M=0$:

$$\frac{\partial F_0(M,T)}{\partial M}-\frac{1}{2}IM-\mu_B H+\frac{\partial \Delta F(M,T)}{\partial M}=0, \tag{9}$$

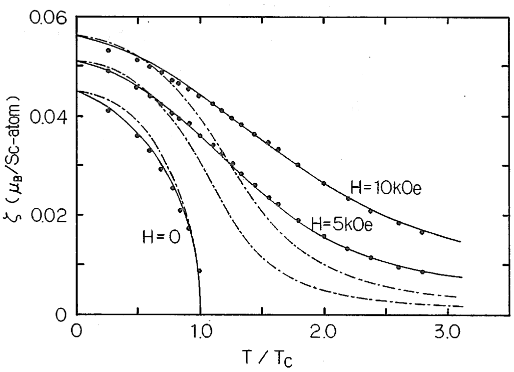

Fig. 1. Temperature dependence of the magnetization for Sc_3In under an external magnetic field. The solid lines are the calculated results based on the SCR theory and the dot-dashed lines the Stoner theory.

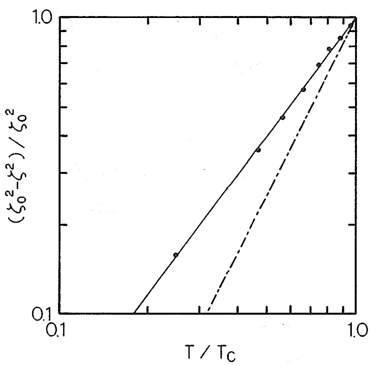

Fig. 2. Magnetization in zero magnetic field plotted as ln $\{(\zeta_0^2-\zeta^2)/\zeta_0^2\}$ vs ln (T/T_c). The slope of the solid line is 4/3 and that of the dot-dashed line is 2.

where $F_0(M, T)$ is the free energy for $I=0$ and the last term is the contribution of spin fluctuations stated in §2. For the electron gas model, eq. (9) is rewritten as

$$\frac{1}{\alpha}\left(1+\frac{2}{27}\zeta^2\right)=1+\frac{3}{2\alpha\zeta}\left(h(H)-\frac{1}{\varepsilon_F}\frac{\partial \Delta F(M, T)}{\partial M}\right), \tag{10}$$

where $h(H)=\mu_B H/\varepsilon_F$. In the above calculation which leads eq. (10), we used

$$\frac{1}{\varepsilon_F}\frac{\partial F_0(M, T)}{\partial M}=\frac{2}{3}\zeta\left(1+\frac{2}{27}\zeta^2+\cdots\right), \tag{11}$$

which is valid for small ζ.

In the case of zero magnetic field, eq. (8) for the Stoner model is reduced to

$$\zeta^2=\zeta_0^2\{1-(T/T_c)^2\}. \tag{12}$$

On the other hand, eq. (10) for the SCR theory in the case of $H=0$ is approximated in the fairly wide temperature region as

$$\zeta^2=\zeta_0^2\{1-(T/T_c)^{4/3}\}. \tag{13}$$

The zero field data are plotted in Fig. 2 as $\ln\{(\zeta_0^2-\zeta^2)/\zeta_0^2\}$ vs $\ln(T/T_c)$, where $\zeta_0=0.045\ \mu_B$/Sc-atom is determined by extrapolating the data to $T=0$. As shown in this figure, the experimental results exhibit a very good fit with eq. (13).

The whole numerical calculations of eqs. (8) and (10) were performed. The dot-dashed lines in Fig. 1 represent the numerical results of eq. (8) derived by making use of the Stoner model. The solid lines represent those of eq. (10) with eqs. (5)–(7) obtained by the SCR theory. Here, the value of α is determined as $1-\alpha=1.5\times10^{-4}$ from eq. (10) in the case of $T=0$ and $H=0$. The value of $k(H=5\text{ kOe})$ in eq. (8) or $h(H=5\text{ kOe})$ in eq. (10) is determined so that the calculated value of ζ at $T=0$ coincides with the experimental value 0.052 μ_B/Sc-atom, which is derived by extrapolating the data in the field of 5 kOe to $T=0$. The value of $k(H=10\text{ kOe})$ or $h(H=10\text{ kOe})$ is chosen twice as large as that of $k(H=5\text{ kOe})$ or $h(H=5\text{ kOe})$, respectively. As shown in Fig. 1, the experimental results can be represented remarkably well by the numerical results of the SCR theory, and cannot by the Stoner model. This fact clearly shows the appropriateness of the SCR theory.

3.2 *Electrical resistivity*

The experimental results of the longitudinal electrical resistivity for Sc_3In in magnetic fields of 0, 1, 2.5, 5 and 10 kOe are shown in Fig. 3. According to the SCR theory, the electrical resistivity due to spin fluctuations has the following properties: (1) the T^2-dependence at low temperature and the coefficient of T^2 being large; (2) the $T^{5/3}$-dependence near T_c both above and below and the change of slope at T_c being small. As shown in Fig. 3 and an inset in this figure, the experimental results have

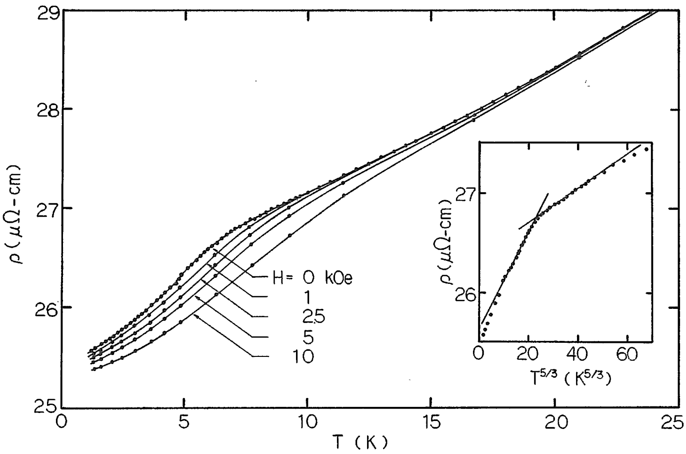

Fig. 3. Temperature dependence of the electrical resistivity for Sc_3In under an external magnetic field. In an inset, electrical resistivity in zero magnetic field plotted against $T^{5/3}$.

just these properties. The coefficient of T^2-dependent resistivity below about 3 K is 0.048 $\mu\Omega$-cm/K^2, which is three orders of magnitude larger than those of typical ferromagnets.

A rather large negative magnetoresistance seems to reflect that the electrical resistivity due to spin fluctuations is reduced strongly by an external magnetic field which suppresses spin fluctuations. According to the theoretical treatment of the magnetoresistance based on the SCR theory, the magnetic field dependence of the coefficient of T^2-term $B(H)$ is given as[7]

$$\frac{B(H)}{B(0)}=\left[\frac{1}{2}\left\{\frac{1}{\sqrt{\tilde{L}(z^2-1)}}\left(\frac{\pi}{2}-\tan^{-1}\frac{z}{\sqrt{\tilde{L}(z^2-1)}}\right)+\frac{z}{z^4+L(z^2-1)}\right\}\right.$$
$$\left.+\frac{\pi}{8}\frac{1}{\sqrt{\tilde{L}(3z^2-1)}}\right]\left[1+\frac{\pi}{8}\frac{1}{\sqrt{2\tilde{L}}}\right]^{-1}, \tag{14}$$

where $z=\zeta/\zeta_0$ and $\tilde{L}$ is determined from the given band structure. By making use of the value of $h(H)$ determined from the magnetization measurement and $\tilde{L}=2$ for the electron gas model, the calculation of eq. (14) was performed. Here, the relation between $h(H)$ and z is given by eq. (10). The calculated result is depicted by the solid line in Fig. 4, which describes well the tendency of the experimental data. Provided $\tilde{L}=2.5$ instead of 2, the calculated result shown by the dashed line in this figure becomes a very good fit with the experimental data. This value of $\tilde{L}$ is to be compared with $\tilde{L}=1.7$, which was obtained from the nuclear magnetic resonance measurement for powdered Sc_3In.[4] The value of z of powder sample is more or less larger than that of bulk sample, which is probably due to the difference of sample preparation procedures between them. In spite of this experimental ambiguity, the agreement seems to be satisfactory.

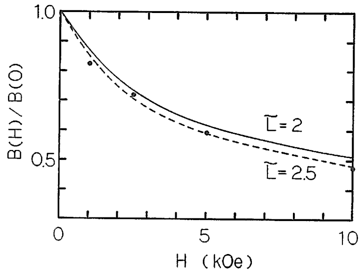

Fig. 4. The coefficient of the T^2-term of the low temperature resistivity as a function of magnetic field. The solid line is the calculated results based on the SCR theory for $\tilde{L}=2$ and the dashed line for $\tilde{L}=2.5$.

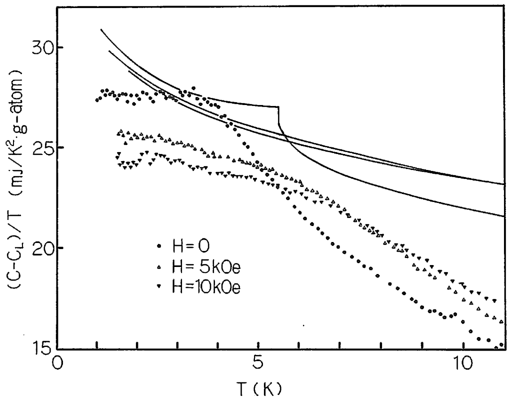

Fig. 5. Temperature dependence of the electronic specific heat for Sc_3In under an external magnetic field. The solid lines are the calculated results based on the SCR theory.

3.3 *Specific heat*

The experimental results of the electronic specific heat, the difference between the total specific heat C and the lattice one C_L, in the magnetic fields of 0, 5 and 10 kOe are shown in Fig. 5. As can be seen from the figure, the effect of an external magnetic field reduces the zero field specific heat in the lower temperature region ($T \lesssim T_c$) and enhances it in the higher temperature region ($T > T_c$).

The magnetic field dependence of the specific heat based on the SCR theory can be calculated from the free energy stated in §2 as follows:[5]

$$C(T) = C_{HF}(T) + \Delta C(T),$$

$$\Delta C(T) = -T\left\{\frac{\partial^2 \Delta F(M, T)}{\partial T^2} + \frac{\partial^2 \Delta F(M, T)}{\partial M \partial T}\frac{\mathrm{d}M}{\mathrm{d}T}\right\}, \tag{15}$$

where $C_{HF}(T)$ is the Hartree-Fock contribution and $\Delta C(T)$ the correction due to spin fluctuations. The numerical calculations of eq. (15) with eqs. (5)–(7) were performed by making use of the values of two parameters, α and $h(H)$, determined from the magnetization measurement and the numerical results of the temperature dependence of ζ. The results are shown in Fig. 5 by the solid lines. Both the zero field data and the deviation from them by applying the magnetic field are in qualitative agreement with the calculation. The fit seems to be satisfactory in view of the case of an electron gas model instead of a realistic band structure. It is noticeable that the behavior of the specific heat in magnetic field crossing the zero field data is caused by the correction term to the Hartree-Fock one.

In summary, we compared the experimental results of the magnetization, electrical resistivity and specific heat for Sc_3In with the numerical calculations based on the SCR theory extended to the case where an external magnetic field is present. These different physical quantities measured were satisfactorily explained by making use of the same parameters, α and $h(H)$, the values of which were determined from the magnetization at $T=0$. This fact implies that spin fluctuations play a predominantly important part in various thermodynamical properties, and that the magnetic field affects them significantly, which is quite well understood within the framework of the SCR theory.

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