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LETTER TO THE EDITOR

Theory of magnetization process of FeSi

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Abstract. We give a new theoretical explanation to the magnetization process of FeSi based on the spin fluctuation theory without assuming the negative mode–mode coupling mechanism previously proposed to explain the anomalous magnetic properties of this compound. It is shown that the numerical results based on this theory are qualitatively in good agreement with the recent magnetization measurement.

The nearly ferromagnetic narrow gap semiconductor FeSi has attracted a lot of interest because of the peculiar temperature dependence of its magnetic susceptibility, i.e. the activation-type increase at low temperature followed by the Curie–Weiss-like dependence at high temperature above 500 K. After an explanation by Jaccarino *et al* (1967) based on a simplified semiconductor model with an unrealistic zero band width for conduction and valence electrons, the temperature-induced local magnetic moment mechanism was proposed (Moriya 1978). Based on the unified picture of spin fluctuations with general amplitude (Moriya and Takahashi 1978), the idea was applied to explain the magnetic and thermal properties of FeSi (Takahashi and Moriya 1979, Takahashi *et al* 1983), which is free from assuming zero band widths for excited carriers. Later band structure calculations (Nakanishi *et al* 1980) showed that FeSi is actually a narrow gap semiconductor with the Fermi energy in the middle of the gap.

On the other hand, the self-consistent renormalization (SCR) theory of spin fluctuation has been quite successful in explaining various magnetic properties of weakly ferro- and antiferromagnetic materials, especially in deriving their Curie–Weiss temperature dependence of the magnetic susceptibilities (Moriya 1985, Lonzarich and Taillefer 1985). It takes into account the renormalization effect of the magnetic susceptibility due to the nonlinear mode–mode coupling among fluctuation modes with different wavevectors. The mode–mode coupling constant is assumed to be almost temperature independent and the spin fluctuation amplitude is assumed to remain small. Because of these restrictions, the SCR theory is not applicable to the case of FeSi. This is the reason why the unified theory of spin fluctuations has been invoked for FeSi in order to take into account the rapid increase of the spin fluctuation amplitude at lower temperature due to the negative mode–mode coupling mechanism, followed by the suppression of the amplitude at higher temperature due to the positive mode–mode coupling effect, which leads to the observed Curie–Weiss-like temperature dependence of the magnetic susceptibility.

After several experimental efforts, the temperature-induced magnetic moment was detected by inelastic neutron scattering measurements (Shirane *et al* 1987, Tajima *et al* 1988). A direct test of the above *negative* mode–mode coupling mechanism, on the other hand, was given by Miyajima (1982) by an Arrott plot analysis of the observed magnetization process, which showed that the fourth-order expansion coefficient of the free energy in the magnetization M actually becomes negative at low temperature, consistent with the negative mode coupling scheme. The same measurement, recently done by Koyama (1998), however, shows a quite different behaviour of the expansion coefficient. It is rather *positive* at low temperature, and its magnitude decreases rapidly with increasing temperature. Both measurements agree in their high-field behaviour. We now believe that the negative slope of the Arrott plot, assigned by Miyajima as the manifestation of the negative mode coupling for low magnetic field, may be the result of some extrinsic effects present in the narrow gap region due to some kind of sample imperfection. The purpose of the present note is to give a theoretical explanation to the above newly observed behaviour of the magnetization process of FeSi.

Recently we proposed a new theoretical explanation for the magnetic properties of FeSi (Takahashi 1997) with the aim of explaining the temperature dependence of the magnetic susceptibility based on the spin fluctuation spectrum consistent with the observed behaviour of neutron scattering intensities (Shirane *et al* 1987, Tajima *et al* 1988). It is based on the spin fluctuation theory (Takahashi 1986, 1990, Takahashi and Sakai 1995), which includes an account of both the thermal and quantum spin fluctuations, and whose consequences have been supported by later experimental investigations (Yoshimura *et al* 1988a, b, Shimizu *et al* 1990, Nakabayashi *et al* 1992). The present study is a straightforward extension of our previous study to obtain the magnetization process. In the following the magnetic susceptibility χ is represented in units of $(g\mu_B)^2$ (where g is the gyromagnetic ratio). We also introduce the magnetization σ per magnetic atom and the magnetic field h in units of energy by

$$M = N_0\mu_B\sigma \qquad h = g\mu_B H$$

where N_0 is the number of magnetic atoms in the crystal. Then the magnetic susceptibility χ is given by

$$\chi/N_0 = \sigma/(2h).$$

According to Takahashi (1986) the magnetization process of FeSi is discussed based on the following sum rule (Takahashi 1997):

$$\begin{aligned}\langle \boldsymbol{S}_i^2\rangle &= \langle\{S_i^+, S_i^-\}/2 + (\delta S_i^z)^2\rangle + \sigma^2/4 \\ &\simeq \frac{3}{4}\langle n_{i\uparrow} + n_{i\downarrow}\rangle = \frac{3}{2N_0}\int_{-\infty}^{\infty} \mathrm{d}\varepsilon\, \rho(\varepsilon) f(\varepsilon) = 3n(T)/2 \end{aligned} \tag{1}$$

where $\rho(\varepsilon)$ is the density of states for conduction electrons. We are mainly concerned with the effect of excited conduction electrons. Other contributions from excited holes and inter-band effects are neglected for simplicity as in our previous study. The band degeneracy of conduction electrons is also neglected.

From the fluctuation–dissipation theorem of statistical mechanics, the spin fluctuation amplitudes are expressed in terms of the response of the system, i.e. in terms of the dynamical magnetic susceptibilities as follows:

$$\begin{aligned}\langle(\delta S_i^z)^2\rangle &= \frac{1}{N_0^2}\sum_{\boldsymbol{q}}\int_{-\infty}^{\infty}\frac{\mathrm{d}\omega}{2\pi}\coth(\beta\omega/2)\mathrm{Im}\chi^{zz}(\boldsymbol{q},\omega) \\ \langle\{S_i^+, S_i^-\}/2\rangle &= \langle(S_i^x)^2 + (S_i^y)^2\rangle = \frac{1}{N_0^2}\int_{-\infty}^{\infty}\frac{\mathrm{d}\omega}{2\pi}\coth(\beta\omega/2)\mathrm{Im}\chi^{-+}(\boldsymbol{q},\omega).\end{aligned} \tag{2}$$

We employ the following form of the dynamical magnetic susceptibility $\chi^{zz}(\boldsymbol{q},\omega)$, for instance, in the paramagnetic phase:

$$\chi(\boldsymbol{q},\omega) = \frac{F(q,\omega)}{1/\chi + 2(U/N_0)[1 - F(q,\omega)]}$$
$$F(q,\omega - \mathrm{i}0) = \chi_0(\boldsymbol{q},\omega - \mathrm{i}0)/\chi_0(0,0) = 1 - \lambda_q(\omega) + \mathrm{i}\gamma_q(\omega) \tag{3}$$

which has the same form as that obtained by the random phase approximation, but with the many-body correction in the uniform and static limit. The Hubbard model underlies the present treatment. The parameter U represents the intra-atomic Coulomb repulsion energy among the conduction electrons. As another simplification we assume a simplified free-electron-gas-like dispersion relation ε_k for conduction electrons in evaluating the non-interacting magnetic susceptibility $\chi_0(\boldsymbol{q},\omega)$. The Fermi level μ, which lies below the bottom of the conduction band by an energy gap Δ, is assumed to be temperature independent, i.e. $\varepsilon_0 - \mu = \Delta$.

In the presence of the uniform magnetization induced by the external field H, we have to take into account the anisotropy of the fluctuation amplitudes. The effect is included by assuming that the uniform and static limit of reciprocals of the perpendicular and longitudinal magnetic susceptibilities $\chi^{-+}(0,0)/2$ and $\chi^{zz}(0,0)$ are given by H/M and $\partial H/\partial M$, respectively, which in reduced units are given by

$$\delta = \frac{1}{U}\frac{h}{\sigma} = \delta_0 + F_1\sigma^2 + \cdots \tag{4}$$
$$\delta_z = \frac{1}{U}\frac{\partial h}{\partial \sigma} = \delta + \sigma\frac{\partial \delta}{\partial \sigma}.$$

The imaginary part of the dynamical magnetic susceptibility is then represented in the form:

$$U\,\mathrm{Im}\chi^{-+}(\boldsymbol{q},\omega)/2 = N_0\frac{\delta+1}{\delta+\lambda_q(\omega)}\frac{\gamma_q(\omega)\{\delta+\lambda_q(\omega)\}}{\{\delta+\lambda_q(\omega)\}^2+\gamma_q(\omega)^2}$$

for the transverse component. The longitudinal component is obtained by replacing δ by δ_z.

We have shown in our previous study that the frequency integrated intensity has a weak wavevector dependence around the origin $q = 0$ in accordance with experiments. We therefore evaluate the local fluctuation amplitude in terms of the dynamical susceptibility for some fixed wavevector $q = q_0$ without performing the wavevector summation as follows:

$$\langle\{S_i^+, S_i^-\}/2\rangle \simeq \frac{1}{N_0}\int_0^\infty \frac{\mathrm{d}\omega}{\pi}\coth(\beta\omega/2)\mathrm{Im}\chi(q_0,\omega). \tag{5}$$

Now by introducing the following function

$$G(\delta) = \frac{1}{\Delta}\int_0^\infty \mathrm{d}\omega\coth(\beta\omega/2)\frac{\delta+1}{\delta+\lambda_{q_0}(\omega)}\frac{\gamma_{q_0}(\omega)\{\delta+\lambda_{q_0}(\omega)\}}{\{\delta+\lambda_{q_0}(\omega)\}^2+\gamma_{q_0}^2(\omega)}$$

the sum rule is finally given by

$$\sigma^2/4 + \frac{\Delta}{\pi U}[G(\delta) + G(\delta_z)/2] \simeq 3n(T)/2. \tag{6}$$

In order to see the temperature dependence of the magnetization process, we have numerically solved (6) as a first-order differential equation of δ with respect to σ. In evaluating the temperature dependence of electron occupation, we employ the density of states curve as shown in the inset of figure 1. By analyzing σ versus h curves in the form of an Arrott plot, the fourth-order expansion coefficient F_1 of the free energy is evaluated for several temperatures. Because of the weak σ-dependence of the slope, the values are estimated in the limit of small σ. We show the result in figure 1. Because of various simplifications adopted in this study, a quantitative comparison with experiments is meaningless. However, the large positive value of the fourth-order free energy expansion coefficient as well as the significant suppression of its magnitude, of the order of 10^5, with increasing temperature are in agreement with the recent observation by Koyama (1998). This seems to further support our idea on the mechanism of the magnetization process of itinerant electron magnets. In contrast, the negative mode–mode coupling mechanism still relies, in a sense, on the single-particle excitation picture in deriving the temperature dependence of the magnetic susceptibility and the mode coupling constant. We can easily see this from the use of the local density of states in deriving the mode coupling effect (Takahashi and Moriya 1979). According to the present study, the magnetization process is determined by the suppression of the spin fluctuation amplitude by applying the external magnetic field. From the sum rule the reduced fluctuation amplitude is converted into the induced uniform magnetization, resulting in the magnetization process of the system. Therefore the underlying idea of the present study is quite different from our previous one.

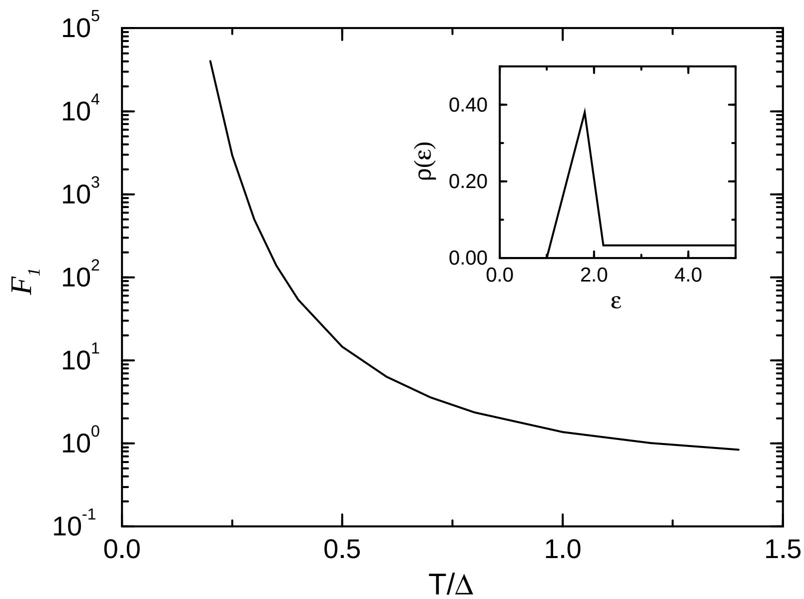

Figure 1. The temperature dependence of the fourth-order free energy expansion coefficient F_1 in arbitrary units as a function of kT/Δ for $U\chi_0(0,0)/N_0 = 2.0$ and $U/\Delta = 10.0$.

In order to check the Kondo insulator model for FeSi proposed by Aeppli and Fisk (1992), the pressure dependence of the gap energy was observed by measuring the pressure dependence of the magnetic susceptibility at low temperature below the susceptibility maximum (Koyama *et al* 1998). In their analysis they assume that each excited carrier has a $1/T$ contribution to the bulk magnetic susceptibility. In order to check their assumption we also show in figure 2 the ratio of the temperature dependence of the calculated magnetic susceptibility and the electron occupation number. As shown in the figure the ratio shows quite a good $1/T$ dependence at low temperature. Reasonable agreement of the size of the pressure dependence of the energy gap with that of the band calculation (Grechnev *et al*

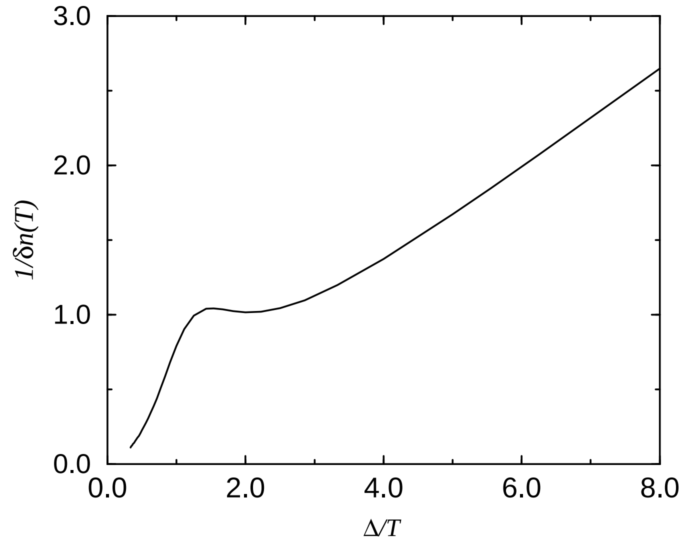

Figure 2. The temperature dependence of the reduced magnetic susceptibility $1/\delta$ per excited conduction electron, $n(T)$.

1994) indicates that the magnetism of FeSi is basically described by the itinerant electron model.

In conclusion, the anomalous magnetic properties of FeSi are explained based on the itinerant electron picture by taking into account the effects of exchange enhanced spin fluctuations. We explain both the temperature dependence of the magnetic susceptibility and the fourth-order expansion coefficient of the free energy within the unified theoretical framework of the spin fluctuation theory over a wide temperature range.

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