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1997 J. Phys.: Condens. Matter 9 2593

(http://iopscience.iop.org/0953-8984/9/12/008)

J. Phys.: Condens. Matter **9** (1997) 2593–2605. Printed in the UK PII: S0953-8984(97)78416-9

Spin-fluctuation theory of FeSi

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Received 7 October 1996

Abstract. The temperature dependence of the magnetic susceptibility of FeSi is calculated over a wide temperature range on the basis of spin-fluctuation theory applied to a model of an exchange-enhanced semiconductor. We can reproduce the low-temperature activation-type behaviour of the susceptibility as well as the Curie–Weiss behaviour observed at high temperature. We also give an explanation for the behaviour of the observed wave-vector- and frequency-dependent neutron scattering intensities.

1. Introduction

The cubic compound FeSi with B20 crystal structure has long attracted lots of interest because of the anomalous temperature dependence of its various physical properties. The magnetic susceptibility, for instance, shows an activation-type temperature dependence at low temperature. After showing a broad maximum at around 500 K, it starts to decrease, showing a Curie–Weiss-like temperature dependence (Jaccarino *et al* 1967). From the band-structure calculations by Nakanishi *et al* (1980) for the series of 3d-transition-metal monosilicides, it was shown that FeSi is a narrow-gap semiconductor.

In the late 1970s we proposed (Takahashi and Moriya 1979) that the magnetic and thermal properties of FeSi can possibly be explained via the idea of temperature-induced magnetic moments proposed by Moriya (1978). The study was done in the course of trials to treat the spin fluctuations of general amplitude with the aim of extending the self-consistently renormalized (SCR) spin-fluctuation theory (for reviews, see Moriya 1985) towards a unified description of itinerant-electron magnetism (Moriya and Takahashi 1978). The negative mode–mode coupling effective at low temperature will induce a local moment with increasing temperature. At high temperature, the mode–mode coupling will change its sign, and it then suppresses the growth of the spin-fluctuation amplitude, leading to the observed Curie–Weiss-like temperature dependence of the magnetic susceptibility. Because of this non-linear mode–mode coupling, we cannot apply the SCR theory to this problem, which always gives constant positive mode–mode coupling.

Since then, experimental efforts have been made in order to detect the temperature-induced magnetic moments directly. But the efforts were either not successful in detecting any signals (Motoya *et al* 1980, Kohgi *et al* 1986, Oh *et al* 1987) or controversial (Ziebeck *et al* 1983) before 1987. Using a single crystal and the polarization technique, Shirane *et al* (1987) were able to detect the temperature-induced magnetic moment of FeSi, and Tajima *et al* (1988) revealed the wave-vector- and frequency-dependent neutron scattering cross section. We here summarize their main results: (1) they revealed the dominant ferromagnetic spin correlation as seen in the quasi-elastic peak of the scattering intensity

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around the origin of the wave-vector space; and (2) the integrated intensities with respect to the frequency, however, show little wave-vector dependence.

Interest has recently been renewed in FeSi since it was suggested that it may be classified as a typical Kondo insulator among transition metal compounds (Aeppli and Fisk 1992). This amounts to saying that the observed energy gap may result not from its electronic band structure, but as the result of the many-body electron correlations. Experimental efforts have been made to check this possibility (Schlesinger *et al* 1993, Ohta *et al* 1994, Saitoh *et al* 1995). Optical conductivity measurements by Schlesinger *et al* (1993) suggested that the charge response of FeSi is different from that of ordinary semiconductors. However, it was shown that the behaviour of the optical conductivity is mostly accounted for in terms of the band structure of this compound (Saitoh *et al* 1995). The temperature dependence of the magnetic susceptibility at low temperature was also found to be consistent with the energy gap structure derived from the band-structure calculations (Jarlborg 1995). Therefore it is more probable that FeSi is not a Kondo insulator, and its energy gap comes from the single-particle band structure of this compound. The problem is therefore that of how to reproduce the Curie–Weiss temperature dependence of the magnetic susceptibility at high temperature, and how to describe its temperature dependence over a wide temperature range in a unified framework.

The purpose of the present paper is twofold. The first aim is to give a theoretical description of the observed behaviours in neutron scattering experiments. The second is to discuss the temperature dependence of the magnetic susceptibility on the basis of the observed spin-fluctuation spectrum. From our point of view, the magnetic properties of various materials should be interpreted on the basis of the nature of the spin fluctuations contained in them. When we published our paper, no detailed information was available concerning the nature of the spin-fluctuation spectrum of FeSi. We did not even know that the ferromagnetic correlation is actually dominant.

The present paper differs from our former study in the following points. In our paper (Takahashi and Moriya 1979, Takahashi *et al* 1983), the non-linear effect of mode–mode coupling among various spin-fluctuation modes played the central role in determining the temperature dependence of the magnetic susceptibility. In the former treatment only the thermal components of spin fluctuations were taken into account, and no explicit mention of the temperature dependence of the quantum (or zero-point) components was made. Although the temperature dependence of the non-linear mode–mode coupling constant was taken into account, the explicit temperature dependence of the spin-fluctuation spectrum was ignored, although the temperature dependence of the average distribution of the spectrum was taken into account. From the nature of the static approximation, the calculated temperature dependence of the magnetic susceptibility at low temperature did not look like the observed behaviour, even if we included the dynamical effects.

In contrast, in the present paper we are starting from the idea that all of the magnetic properties are derived from the nature of the spin fluctuations. With the use of a kind of sum rule for the *total* spin-fluctuation amplitude, the temperature dependence of the magnetic susceptibility is derived. In this way, the zero-point spin-fluctuation components also play a significant role. For later convenience, we here define the above-mentioned spin-fluctuation components below. From the fluctuation-dissipation theorem of the linear response theory of the statistical mechanics, the squared local spin amplitude is expressed in terms of the dynamical magnetic susceptibility $\chi(q,\omega)$ in the following form:

$$\langle \boldsymbol{S}_i^2 \rangle = \frac{3}{2N_0^2} \sum_q \int_0^\infty \frac{\mathrm{d}\omega}{\pi} \coth(\beta\omega/2)\, \mathrm{Im}\, \chi(q,\omega). \tag{1}$$

That is

$$\langle \boldsymbol{S}_i^2 \rangle = \langle \boldsymbol{S}_i^2 \rangle_{\rm th} + \langle \boldsymbol{S}_i^2 \rangle_{\rm zp}. \tag{2}$$

The thermal and quantum (zero-point) amplitudes are then defined by

$$\langle \boldsymbol{S}_i^2 \rangle_{\rm th} = \frac{3}{N_0^2} \sum_q \int_0^\infty \frac{{\rm d}\omega}{\pi}\, n(\omega)\, {\rm Im}\, \chi(q,\omega) \qquad n(\omega) = [\exp(\beta\omega) - 1]^{-1} \tag{3}$$

$$\langle \boldsymbol{S}_i^2 \rangle_{\rm zp} = \frac{3}{2N_0^2} \sum_q \int_0^\infty \frac{{\rm d}\omega}{\pi}\, {\rm Im}\, \chi(q,\omega) \tag{4}$$

where $\boldsymbol{S}_i$ is the spin operator on some ith lattice site and β is the reciprocal of the absolute temperature. We use the units where $\hbar = 1$, throughout the paper. Note that the zero-point amplitude defined above does show temperature dependence through the change of the spin-fluctuation spectrum with temperature. The observed almost q-independent frequency-integrated neutron scattering cross section clearly suggests that the zero-point spin fluctuations also play an important role.

We have shown that the near temperature independence of the total spin-fluctuation amplitude gives rise to the same form of the equation as the SCR theory (Takahashi 1986, 1990). In addition, we were successful in deriving its interesting consequences (Takahashi 1992, 1994, 1995). The magnetic properties of weak ferromagnets are described by a smaller number of independent parameters characterizing the spin-fluctuation spectrum in q- and ω-space than those of the SCR theory, as was confirmed by subsequent experiments (Yoshimura *et al* 1988a, b, Shimizu *et al* 1990, Nakabayashi *et al* 1992). In the present treatment, because of the existence of the energy gap, a slight modification of our original idea will be needed. We have to take into account various extra temperature dependences inherent in the present problem. For instance, we have to take into account the temperature dependence of the total spin-fluctuation amplitude. The spin-fluctuation spectrum also depends on temperature.

The plan of the paper is as follows. In the next section we deal with the neutron scattering intensities on the basis of the simple semiconductor model. We calculate the temperature dependence of the dynamical magnetic susceptibility on the basis of the random-phase approximation. In the third section, we propose the sum rule for the total spin-fluctuation amplitude. The temperature dependence of the magnetic susceptibility is then derived with the use of the sum rule and on the basis of the spin-fluctuation spectrum derived in the second section. The final section is devoted to a discussion.

2. The spin-fluctuation spectrum of exchange-enhanced semiconductors

The aim of the present section is to give a theoretical explanation for the wave-vector and frequency dependence of neutron scattering intensities observed by Tajima *et al* (1988a, b). To begin with, let us evaluate the dynamical magnetic susceptibility of the system in the random-phase approximation (RPA). It is, in general, very difficult to do such calculations on the basis of the results of realistic band-structure calculations. In order to extract the underlying physics of the system, we are instead using a simple model system, and we make some further drastic simplifications. The Hamiltonian that we are concerned with here is the following single-band Hubbard Hamiltonian for conduction electrons:

$$H = \sum_{\boldsymbol{k}\sigma} (\varepsilon_{\boldsymbol{k}} - \mu) c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \tag{5}$$

where $c^{\dagger}_{k\sigma}$ ($c_{k\sigma}$) is the creation (annihilation) operator for conduction electrons with wave vector $\boldsymbol{k}$ and spin direction σ. The first term represents the single-particle kinetic energy of conduction electrons and the second one the intra-atomic Coulomb repulsion U among them on the same ith lattice site of Fe ions.

For a realistic description of the system, we would have to take into account the band degeneracy of conduction electrons as well as the hole band contributions. We neglect all of these effects, although the latter effect is implicitly taken into account by considering that the chemical potential μ always stays within the gap region below the bottom of the conduction band by the finite gap Δ, i.e.,

$$\Delta = \varepsilon_0 - \mu.$$

We assume that the temperature dependence of the gap energy Δ is neglected. The inter-band contributions to the dynamical susceptibility are also neglected. The neglected contributions taken together may give rise to some slight quantitative corrections to our final results, but we assume that the qualitative features will remain unchanged by these effects.

In the RPA the dynamical magnetic susceptibility $\chi(q,\omega)$ in units of $(g\mu_B)^2$ is expressed as

$$\chi(q,\omega) = \frac{\chi_0(q,\omega)}{1 - I\chi_0(q,\omega)} \tag{6}$$

where $\chi_0(q,\omega)$ is the non-interacting susceptibility given by

$$\chi_0(q,\omega) = \sum_{\boldsymbol{k}} \frac{f(\varepsilon_{\boldsymbol{k}+\boldsymbol{q}}) - f(\varepsilon_{\boldsymbol{k}})}{\omega + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}. \tag{7}$$

$I = U/N_0$ (N_0: the number of magnetic sites of the crystal), and $f(\varepsilon)$ is the Fermi distribution function. In order to evaluate the above expression, we assume a free-electron-like dispersion relation for $\varepsilon_{\boldsymbol{k}}$ in (5) with an effective mass m^*, i.e.,

$$\varepsilon_{\boldsymbol{k}} = \varepsilon_0 + \frac{k^2}{2m^*}.$$

In the following numerical calculations, all of the energies are measured in units of the gap energy Δ and the wave-vectors in units of k_0, defined by

$$k_0 = \sqrt{2m^*\Delta}.$$

We thus introduce the reduced dimensionless temperature t, the frequency w, and the wave-vector p:

$$t = k_B T/\Delta \qquad w = \omega/\Delta \qquad p = q/k_0.$$

The summation over spherical-angle degrees of freedom of (7) is easily performed, and gives the following form of the non-interacting dynamical susceptibility. The calculation is essentially the same as that of the free-electron-gas model. The difference is that we cannot replace the Fermi distribution function with the step function in the present case and we have to deal with its temperature dependence explicitly:

$$\chi_0(q,\omega) = \frac{3N_0}{2\,\Delta x_0^3} F(p,w) \tag{8}$$

$$F(p, w+\mathrm{i}0) = \frac{1}{2p}\int_0^{x_0} x\ \mathrm{d}x\ f(x)\left\{\ln\left(\frac{p_+ - x}{p_+ + x}\right) - \ln\left(\frac{p_- - x}{p_- + x}\right)\right\}$$
$$= F'(p,w) + \mathrm{i}F''(p,w) \tag{9}$$

$$p_\sigma = -(w + \mathrm{i}0 + \sigma p^2)/2p$$

where $x_0 = k_{BZ}/k_0$ with k_{BZ}, the effective zone-boundary wave-vector, given by the expression $(6\pi^2 N_0/V)^{1/3}$ (V: the volume of the crystal), and the Fermi distribution function $f(x)$ is given by

$$f(x) = 1/[\exp((x^2+1)/t) + 1].$$

The real and the imaginary parts F' and F'' in (9) are, respectively, expressed in the following integral forms:

$$F'(p,w) = \frac{1}{2p}\int_0^{x_0} x\ \mathrm{d}x\ f(x)\left\{\ln\left|\frac{x-p_+}{x-p_+}\right| - \ln\left|\frac{x-p_-}{x-p_-}\right|\right\} \tag{10}$$

$$F''(p,w) = \frac{\pi}{2p}\left\{\int_0^{\min(x_0,p_-)} x\ \mathrm{d}x\ f(x) - \int_0^{\min(x_0,p_+)} x\ \mathrm{d}x\ f(x)\right\}. \tag{11}$$

In the small-q, ω region, the imaginary part is easily integrated, giving

$$F''(p,w) = \frac{\pi t}{4p}\{\ln(1+\exp[-(1+p^2/4+w^2/4p^2 - w/2)/t]) - \ln(1+\exp[-(1+p^2/4+w^2/4p^2 + w/2)/t])\}. \tag{12}$$

It is easy to see that $F''(p,w)$ is given by the following w-linear form when w is very small:

$$F''(p,w) = \frac{\pi w}{4p}\frac{1}{\exp[(1+p^2/4)/t]+1}. \tag{13}$$

We have to perform numerical integration to obtain the real part of $F'(p,w)$ for a given value of p and w.

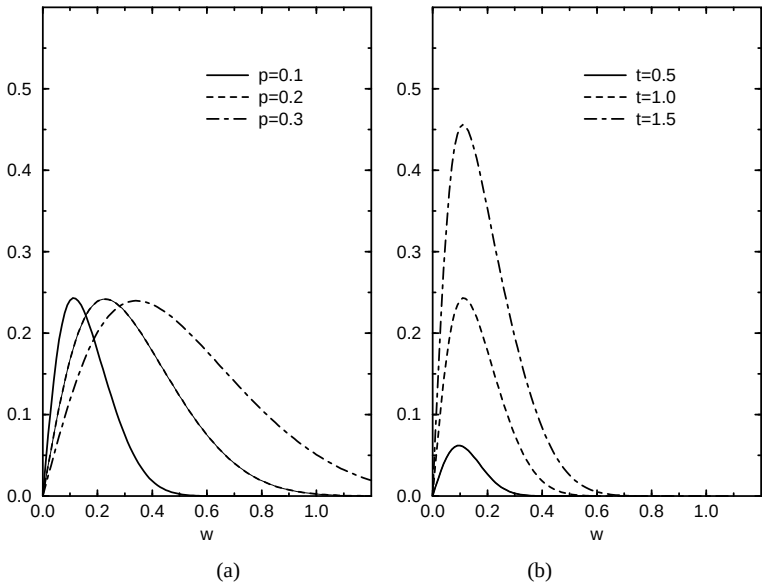

Figure 1. The frequency dependence of $I\ \mathrm{Im}\,\chi(q,\omega)$ for $p = 0.1$, 0.2, and 0.3 at $t = 1.0$ (a), and for $p = 0.1$ at $t = 0.5$, 1.0, and 1.5 (b).

On substituting (8) into (6), the imaginary part of $\chi(q,\omega)$ is represented by

$$\mathrm{Im}\,\chi(q,\omega) = \frac{1}{I\alpha(t)}\frac{\gamma_p(w)}{[1/\alpha(t) - 1 + \lambda_p(w)]^2 + \gamma_p^2(w)} \tag{14}$$

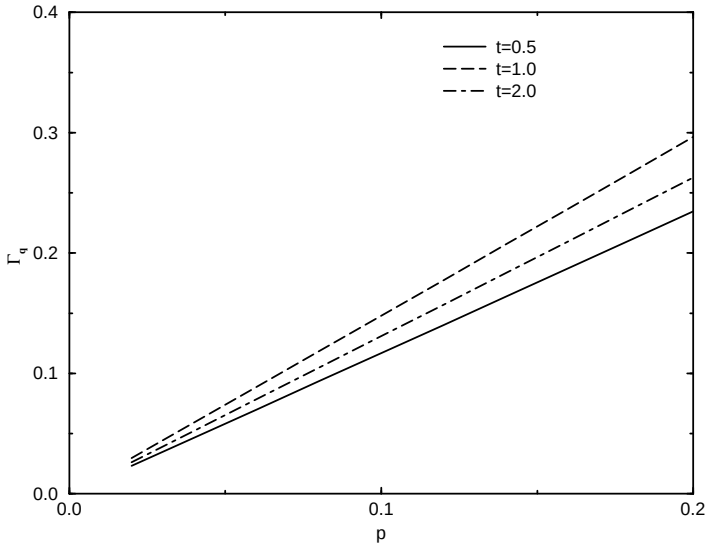

Figure 2. The wave-vector dependence of the damping constant (half-width) Γ_q at $t = 0.5$, 1.0, and 2.0.

where we define α_0, $\alpha(t)$, $\lambda_p(w)$, and $\gamma_p(w)$ by

$$\alpha_0 = \frac{3N_0}{2\,\Delta x_0^3} I \qquad \alpha(t) = \alpha_0 F'(0,0)$$
$$\lambda_p(w) = 1 - F'(p,w)/F'(0,0)$$
$$\gamma_p(w) = F''(p,w)/F'(0,0).$$

We show in figure 1 the w-dependence of $I\,\mathrm{Im}\,\chi(q,\omega)$ for several values of p at $t = 1.0$ (a) and for several values of t for a fixed wavenumber $p = 0.1$. We assume $\alpha_0 = 1$ throughout this paper. As shown in (13), because $\gamma_p(w)$ is proportional to w for small w, the w-dependence of the imaginary part is usually analysed in terms of the following Lorentzian form:

$$\mathrm{Im}\,\chi(q,\omega) \propto \frac{\Gamma_q \omega}{\omega^2 + \Gamma_q^2}. \tag{15}$$

By comparing (14) and (15) above, the damping constant Γ_q is found to be

$$\Gamma_q(w) = \frac{w}{\gamma_p(w)}\left[1/\alpha(t) - 1 + \lambda_p(w)\right]$$
$$\Gamma_q = \Gamma_q(w)\big|_{w\to 0} = \frac{4pF'(0,0)}{\pi}\{1 + \exp[(1 + p^2/4)/t]\}(1/\alpha(t) - 1 + \lambda_p(0)). \tag{16}$$

The above analysis implicitly assumes that the w-dependence of $\Gamma_q(w)$ is very weak for small w. We found, however, we cannot neglect the w-dependence of $\Gamma_q(w)$ especially at low temperature. The value of Γ_q defined in (16) overestimates the actual half-width of the spectrum. In the present paper, we numerically evaluated the damping constant Γ_q from the spectral half-width of $\mathrm{Im}\,\chi(q,\omega)/\omega$. The value of Γ_q obtained in this way differs significantly from the one defined in (16) at low temperature. At high temperature, the difference tends to become very small. We show the q-dependence of the half-width in figure 2 for several temperatures t. All of the curves show good p-linear

dependence, indicating the Landau damping mechanism of the excited conduction electrons. The behaviour is also in good accordance with experiments except at low temperature ($T = 300$ K), where the observed Γ_q shows a steep increase according to the q^2-like behaviour. Note that we have to take into account the considerable experimental errors in deriving Γ_q due to the very weak neutron intensities for such low temperatures. We have to be very careful in drawing any definite conclusions from the above discrepancy between the theory and the experiments.

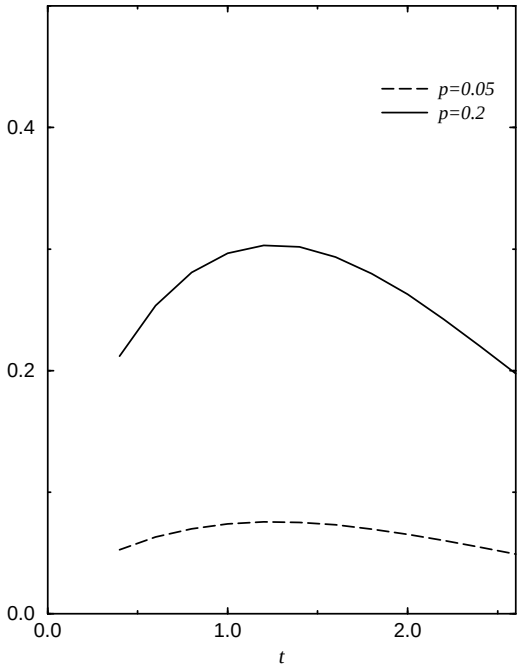

Figure 3. The t-dependence of the damping constant Γ_q for $p = 0.05$ and 0.2.

As for the t-dependence of the half-width, we plot in figure 3 calculated values of Γ_q against t for $p = 0.05$ and 0.2. We see the general tendency that Γ_q increases with t at low temperature. After showing a broad maximum, it starts to decrease with increasing temperature. If we discard the low-temperature part of our calculated results because of the lack of corresponding experimental data, the rest of the t-dependence of Γ_q for $p = 0.2$ seems to reproduce well the observed monotonically decreasing behaviour for moderate values of q. Though we can reproduce the increasing behaviour of Γ_q for small q at low temperature, we were not successful in deriving its monotonically increasing behaviour. It is, however, at least consistent with the observed very weak temperature dependence for small wave-vectors.

The neutron scattering cross section is proportional to the following function, $S(q, \omega)$:

$$S(q, \omega) = \frac{1}{1 - \exp(-\beta\omega)} \operatorname{Im} \chi(q, \omega) \tag{17}$$

or

$$S(q, \omega) = \frac{1}{1 - \exp(-w/t)} \frac{\gamma_p(w)/I\alpha(t)}{[1/\alpha(t) - 1 + \lambda_p(w)]^2 + \gamma_p^2(w)}. \tag{18}$$

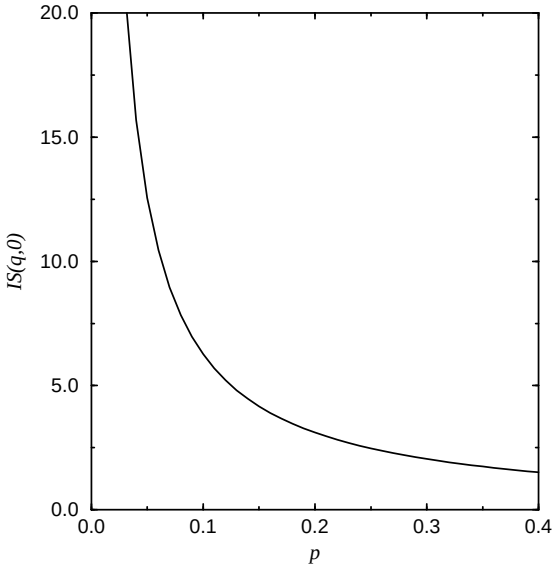

Figure 4. The calculated q-dependence of $IS(q, 0)$ for $t = 1.2$.

We show in figure 4 the calculated wave-vector dependence of $IS(q, 0)$ for $t = 1.2$. We see the diverging behaviour of $S(q, 0)$ around the origin, indicating the ferromagnetic correlation. We also show, in figure 5, the integrated intensity as a function of the wave-vector p for several temperatures (a), and the wave-vector dependence of each component of the thermal and the zero-point spin-fluctuation amplitudes (b). All of the curves for total amplitudes are almost q-independent, while their components show complementary q-dependences, keeping their sums almost q-independent.

To conclude, most of the qualitative behaviours of the observed $S(q, \omega)$ in q, ω-space seem to be relatively well accounted for by the present simple RPA treatment based on the semiconductor model. If we take into account the precise temperature dependence of the magnetic susceptibility going beyond the RPA as will be discussed below, the agreements are further improved. Slight qualitative discrepancies exist between the present theory and experiments as regards the q-dependence of Γ_q at low temperature, and its t-dependence for small q.

3. Magnetic properties of FeSi

In the preceding section, we showed that the qualitative features of the neutron scattering cross section in q, ω-space are well explained in the framework of the RPA. The approximation, however, is equivalent to evaluating the temperature dependence of the magnetic susceptibility in terms of the single-particle excitations through the electronic density-of-states curve. At low temperature such calculations based on the band-structure calculation are found to be in good agreement with experiments (Ohta *et al* 1994, Jarlborg 1995). The problem of this section is to derive the Curie–Weiss-like temperature dependence at high temperature.

Our strategy is very simple. We first note the following relation, which holds for the

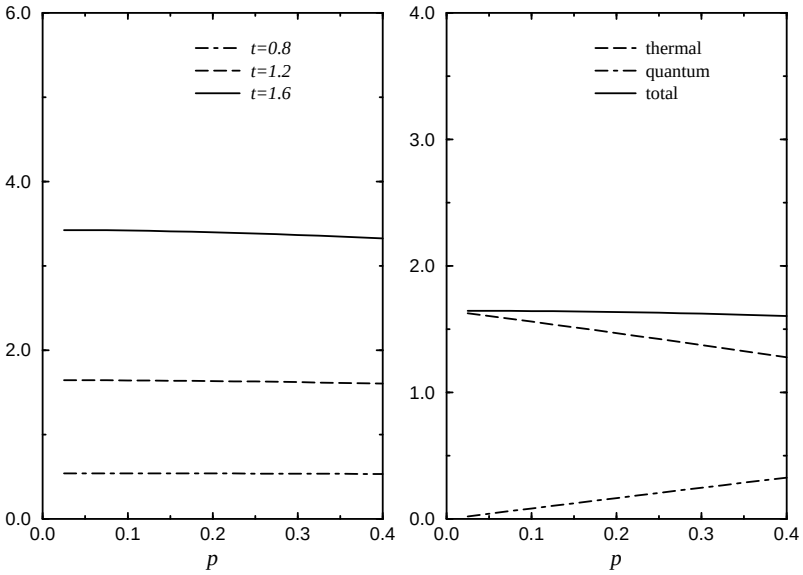

Figure 5. (a) The q-dependence of the integrated neutron intensity for $t = 0.8$, 1.2, and 1.6. (b) The q-dependence of the total spin-fluctuation amplitude (solid line), and each of its component thermal (dashed line) and quantum zero-point (chain line) amplitudes.

spin operators on each ith lattice site:

$$\frac{1}{2}\langle\{S_i^+, S_i^-\}\rangle = \left\langle \frac{1}{2}(n_{i\uparrow} + n_{i\downarrow}) - n_{i\uparrow}n_{i\downarrow} \right\rangle \tag{19}$$

where $\{A, B\}$ is the anti-commutation relation between operators A and B, and $\langle\cdots\rangle$ represents the canonical thermal average. The second term of the right-hand side of (19) can be neglected because it is very small when the electron occupations are small, and even if they are finite it is also neglected because of the on-site Coulomb repulsion between spin-up and spin-down electrons. In the present case, it is neglected mainly for the first reason given above, as will be shown below in figure 6. It follows that the right-hand side of (19) is simply determined by the thermal average of the occupation numbers of the conduction electrons, and it is thus evaluated in terms of the single-particle Green's function $G_{k\sigma}$:

$$\langle n_i\rangle = \sum_\sigma \langle n_{i\sigma}\rangle = \frac{1}{\pi N_0}\sum_{k\sigma}\int_{-\infty}^{\infty} \mathrm{d}\varepsilon \; f(\varepsilon)\,\mathrm{Im}\,G_{k\sigma}(\varepsilon) = \int_{-\infty}^{\infty}\mathrm{d}\varepsilon \; f(\varepsilon)\rho(\varepsilon) \tag{20}$$

$$\rho(\varepsilon) = \frac{1}{\pi N_0}\sum_{k\sigma}\mathrm{Im}\,G_{k\sigma}(\varepsilon) \tag{21}$$

where $\rho(\varepsilon)$ is the local density of states per magnetic ion. The quasi-particle pole of the Green's function $G_{k\sigma}$ is affected by the effects of a shift and a lifetime broadening because of various scattering processes. These effects are, however, smeared out by the process of the integration over the whole Brillouin zone, and the local density $\rho(\varepsilon)$ defined above will be well approximated by the density of states obtained by band-structure calculations. The local density is less affected by the low-lying magnetic excitations, and its temperature dependence is neglected.

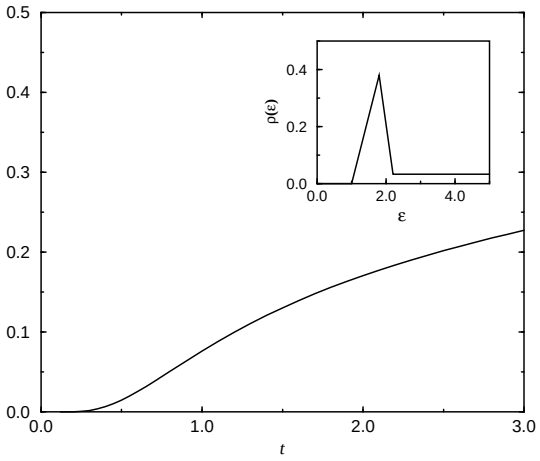

Figure 6. The temperature dependence of the occupation number $\langle n_i \rangle$. The model density of states is shown in the inset.

In the metallic system with a finite density of states at the Fermi energy, the temperature dependence of the Fermi distribution function is neglected. This is why we assumed the total spin-fluctuation amplitude to be temperature independent in our treatment of weakly ferromagnetic materials. In the present case, however, we have to take into account the temperature dependence of $\langle n_i \rangle$ through that of the Fermi distribution function $f(\varepsilon)$. The point is that its temperature dependence is determined independently of low-lying magnetic excitations. Therefore (19) serves as an external condition obeyed by the spin-fluctuation spectrum. Now with the use of (1), the sum rule (19) can be expressed in terms of the imaginary part of the dynamical magnetic susceptibility as follows:

$$\frac{1}{N_0^2}\sum_q \int_0^\infty \frac{\mathrm{d}\omega}{\pi}\ \coth(\beta\omega/2)\,\mathrm{Im}\,\chi(q,\omega) = \int_{-\infty}^{\infty} \mathrm{d}\varepsilon\ f(\varepsilon)\rho(\varepsilon). \tag{22}$$

Note that the dynamical susceptibility $\chi(q,\omega)$ in the RPA is not consistent with the requirement that its static and uniform limit has to coincide with the exact magnetic susceptibility. In the RPA, the limit is given by the value, $I[1/\alpha(t)-1]$, obtained from the Hartree–Fock approximation. In order to recover this condition, we assume that the form of the imaginary part of $\chi(q,\omega)$ in (14) is given by

$$\mathrm{Im}\,\chi(q,\omega) = \frac{\delta+1}{I[\delta+\lambda_p(w)]}\,\frac{\gamma_p(w)[\delta+\lambda_p(w)]}{[\delta+\lambda_p(w)]^2+\gamma_p^2(w)} \tag{23}$$

where we define the reciprocal magnetic susceptibility δ by

$$\delta = \frac{1}{I\chi} = \frac{1}{\alpha(t)} - 1 + \Delta\delta. \tag{24}$$

The last term $\Delta\delta$ in (24) represents the many-body correction to the Hartree–Fock approximation.

Equation (19) is now regarded as the external constraint for δ. We can regard δ as a free disposable parameter and at each temperature determine its magnitude from the above condition. We can thus derive the temperature dependence of the magnetic susceptibility over a wide temperature range. At low temperature, for example, because δ is very large, $\mathrm{Im}\,\chi(q,\omega)$ is well approximated by $\gamma_p(w)/I\delta$. Then from the condition (19), it is easy to see

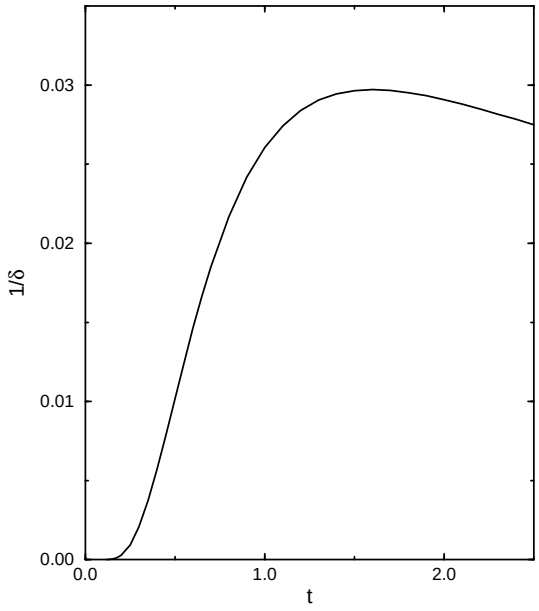

Figure 7. The calculated temperature dependence of the reduced magnetic susceptibility, $1/\delta$.

that (22) is well satisfied by the Hartree–Fock solution and the correction $\Delta\delta$ is negligible. The condition becomes effective only at high temperatures where the susceptibility increases and in the presence of the enhanced spin-fluctuation amplitudes.

In order to make the actual calculation more tractable, we utilize the fact that the frequency-integrated neutron intensity is almost q-independent. This means that, in place of (22), we can determine δ from the following condition for some single wave-vector $q = q_0$:

$$\frac{1}{N_0}\int_0^\infty \frac{\mathrm{d}\omega}{\pi}\ \coth(\beta\omega/2)\,\mathrm{Im}\,\chi(q_0,\omega) = \int_{-\infty}^{\infty}\mathrm{d}\varepsilon\ f(\varepsilon)\rho(\varepsilon). \tag{25}$$

We solved (25) numerically for δ. In the actual calculations, we employed the model density of states for $\rho(\varepsilon)$ shown in the inset of figure 6, and assumed $q_0 = 0.1$. The temperature dependence of $\langle n_i\rangle$ is shown in figure 6, and the calculated temperature dependence of the susceptibility in figure 7. From figure 7 we see that the temperature dependence of χ in FeSi is well reproduced in the present treatment. In order to produce the Curie–Weiss behaviour of the magnetic susceptibility at high temperature, the electron occupation has to show a tendency towards slight saturation. The behaviour was actually observed in neutron scattering measurements by Tajima *et al* (1988). The peak structure of the density-of-states curve around the band edges is also responsible for the Curie–Weiss behaviour.

4. Discussion

In the present paper we have succeeded in explaining the temperature dependence of the magnetic susceptibility of FeSi over a wide temperature range. We could show both the exponential rise of the magnetic susceptibility at low temperature and the Curie–Weiss-like decrease at high temperature at the same time in a unified treatment. The underlying physics of the origin of the Curie–Weiss behaviour is quite similar to the one that we

proposed previously (Takahashi 1986). It comes from the so-called sum rule for the squared local spin-fluctuation amplitude. We found that the peculiar temperature dependence of the magnetic susceptibility of this compound results from the temperature dependence of the spin-fluctuation spectrum, which reflects the gap structure of the electronic states and that of the electron occupation number originating from the strong peak structure of the electronic density of states around the edges of the energy gap.

In the present treatment we are led to the concept of temperature-induced moments, but the meaning is different from that in our previous work where only the thermal component of the spin fluctuations was considered. In the present case we refer instead to the induced *total* spin-fluctuation amplitude, whose temperature dependence is independently determined by the single-particle excitation of the charge degrees of freedom.

All of the experimental results obtained recently, as well as numerical band-structure calculations, suggest that the magnetic properties of FeSi are at low temperature well reproduced. In the present paper, we are successful in deriving the Curie–Weiss-like temperature dependence of the magnetic susceptibility. All of these points suggest that the anomalous magnetic properties of FeSi are basically realized in terms of the band model if a proper account of the effects of exchange-enhanced spin fluctuations is taken. It is reported (Hunt *et al* 1994, Degiorgi *et al* 1994) that FeSi is not a simple semiconductor at low temperature. We suppose that properties observed there are not intrinsic, and can be ascribed to some extrinsic effects—due to impurity levels in the narrow-gap region, for instance.

We show that the neutron scattering intensities are qualitatively well explained by our simple semiconductor model, except some slight discrepancies in limited temperature and wave-vector regions. In the present simplified treatment, we have neglected various contributions. For instance, the inter-band contribution to the magnetic susceptibility was neglected. As a semiconductor model we assume a simple free-electron-like dispersion relation. The effects of band degeneracy are also ignored. All of these effects may slightly alter the present results, though the qualitative features of results will remain the same. At the same time, these effects may provide the answer to the remaining problems mentioned above.

Before concluding this study, we would like to make a brief comment on our present method. In the present treatment, we make use of a kind of sum rule in deriving the temperature dependence of the magnetic susceptibility. It is thus free from any kinds of approximation method. What we need is the form of the spin-fluctuation spectrum. On the other hand, in our former treatment, we had to rely on the static approximation in order to derive the non-linear mode–mode coupling among the various spin-fluctuation modes. We have already mentioned that the conventional SCR theory was unable to treat the present problem in its original form. It is also very interesting to note that the sum rule plays a dominant role in determining the temperature dependence of the magnetic susceptibilities for quite a wide class of the magnetic materials, ranging from insulator magnets to weak itinerant-electron magnets.

Acknowledgments

The author would like to thank Professor K Tajima for valuable discussions, and Miss M Shiraishi for a careful reading of the manuscript.

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