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The influence of spin fluctuations on the temperature dependence of the magnetization in $ZrZn_2$

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Abstract. We consider the influence of spin fluctuations on the temperature dependence of the magnetization in an itinerant magnet. We calculate the temperature dependence of the magnetization, paying particular attention to the effects of the differing temperature dependences of the transverse $\langle m_\perp^2 \rangle$ and longitudinal $\langle m_\parallel^2 \rangle$ magnetic fluctuations. We find that over a wide range in temperature the magnetization obeys the mixed law $M^2/M_0^2 \sim 1 - \frac{4}{5}(T/T_1)^{4/3} - \frac{1}{5}(T/T_u)^2$. We use this result to explain the observed temperature dependence of the magnetization in the weak itinerant magnet $ZrZn_2$.

1. Introduction

There has been, and still is, great interest in understanding the temperature dependence of the spontaneous magnetization and magnetic susceptibility observed in weakly ferromagnetic metals [1–4]. Recent descriptions [2, 3] based on conventional interacting electron theory, but taking into account the temperature dependence of both transverse and longitudinal fluctuations in the local magnetization, appear to be very successful in describing these phenomena. There are, however, several aspects of this approach which in our opinion have not yet been fully explored. In particular the difference in the behaviour of the transverse $\langle m_\perp^2 \rangle$ and longitudinal $\langle m_\parallel^2 \rangle$ magnetic fluctuations in the ferromagnetic state has not been fully addressed. In this paper we present results of an approach based on electron liquid theory [5] which enables us to determine the dependence of magnetic fluctuations in a self-consistent way. This approach leads a differential equation for the coefficients of the free energy as a function of magnetization M and enables us to calculate the spontaneous magnetization, M, as a function of temperature. In particular if M_0 is the magnetization at zero temperature, we find that over a wide range in temperature, T, the spontaneous magnetization obeys the mixed law $M^2/M_0^2 \sim 1 - \frac{4}{5}(T/T_1)^{4/3} - \frac{1}{5}(T/T_u)^2$. We use this result to describe the temperature dependence of the spontaneous magnetization found in the weak itinerant ferromagnet $ZrZn_2$ [6, 7].

We begin by writing the free energy per unit volume F/V in the form [4]

$$F(T, V, M)/V = F_0/V + \tfrac{1}{2}AM^2 + \tfrac{1}{4}\gamma_u M^4 \tag{1}$$

where F_0 is the free energy when $M = 0$ and A and γ_u are given by

$$A = a + 2\gamma_u(1 + a/\gamma_u M^2)\langle m_\perp^2 \rangle + 3\gamma_u(1 + a/3\gamma_u M^2)\langle m_\parallel^2 \rangle \tag{2}$$

$$\gamma_u = G/12\rho\chi_p^2 E_F^2. \tag{3}$$

In equation (3)

$$G = [3(\rho'/\rho)^2 - \rho''/\rho]E_F^2 \tag{4}$$

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is a dimensionless quantity characterizing the shape of the density of states, ρ, around the Fermi level, E_F, and χ_p is the single-particle Pauli susceptibility given by $\chi_p = 2\rho\mu_B^2$. The first term in equation (2) can be expressed as

$$a = \mathcal{B}_+ \tfrac{5}{3}\chi_p \qquad (5)$$

where $\mathcal{B}_+$ is the Fermi liquid parameter determining the magnetic state of the system. In the case of $ZrZn_2$, $\mathcal{B}_+$ is very small and negative (~ -0.01) corresponding to $ZrZn_2$ being a weak ferromagnet. The quantity $\mathcal{B}_+$ and the Fermi energy E_F set the energy scales for the physical quantities of interest and, in particular, determine the temperature scales which will be used to determine the various regions of different magnetization behaviour.

The thermal variance of the local magnetization $\langle m_\nu^2\rangle$ introduced in equation (2) is formally given by

$$\langle m_\nu^2\rangle = 4\hbar \int \frac{\mathrm{d}\boldsymbol{q}\,\mathrm{d}\omega}{(2\pi)^4} n(\omega)\, \mathrm{Im}\, \chi_\nu(\boldsymbol{q}, \omega) \qquad (6)$$

where $n(\omega) = [\exp(\hbar\omega/k_BT) - 1]^{-1}$ is the Bose factor and $\chi_\nu(\boldsymbol{q}, \omega)$ is the generalized dynamical wave-vector dependent susceptibility. In these, the subscript ν is either $\perp$ or $\parallel$ depending on whether the magnetization fluctuations are perpendicular or parallel to the average magnetization M.

The applied magnetic field, B, is obtained by differentiating equation (1) with respect to M so that

$$B = (1/V)\,\partial F(T, V, M)/\partial M = AM + \tfrac{1}{2}M^2(\partial A/\partial M)_{T,V} + \gamma_u M^3. \qquad (7)$$

The parallel and perpendicular static susceptibilities are given by $\chi_\perp^{-1} = B/M$ and $\chi_\parallel^{-1} = \partial B/\partial M$ and so from equation (7)

$$\chi_\perp^{-1} = A + \tfrac{1}{2}M\,\partial A/\partial M + \gamma_u M^2 \qquad (8)$$

$$\chi_\parallel^{-1} = A + 2M\,\partial A/\partial M + \tfrac{1}{2}M^2\,\partial^2 A/\partial M^2 + 3\gamma_u M^2. \qquad (9)$$

These equations indicate the inherent self-consistency needed in the treatment of the magnetization problem, for in calculating A we need $\langle m_\nu^2\rangle$ which is related to χ_ν^{-1} and thus to A and its derivatives. One is therefore faced with a highly non-linear differential equation for A and hence M. Early attempts at solving this or understanding the general equation of state given by equation (7) were based on the Stoner model and gave estimates for A which were too small [8, 9]. Attempts were made to refine the theory by including transverse fluctuations [10], longitudinal fluctuations [3] and a temperature dependent cut-off wave vector for the thermally excited modes [3]. Unfortunately in [3] the assumption that the magnetization dependence of χ^{-1} could be approximated by

$$\chi_\parallel^{-1} = \partial B/\partial M = A + (1 + 2\eta)\gamma_u M^2 \qquad (10)$$

in which the parameter η was chosen to be either 1, 0 or M^2/M_0^2, was used in order to reduce the computation effort. The choice $\eta = 1$ corresponds to ignoring the $\partial A/\partial M$ terms and was found to lead to a first-order transition [3]. The choice $\eta = 0$ does however lead to a second-order transition but corresponds to treating longitudinal fluctuations in the same way as transverse fluctuations. The third choice, $\eta = M^2/M_0^2$, is an interpolation between the first two, and while numerically giving satisfactory results [3] is neither derived nor self-consistent. In this paper we use the results developed in [4] where no assumptions about the M dependence of $\chi_\parallel^{-1}$ were made. Apart from the Curie temperature T_c this approach leads to two new characteristic temperature scales, $T_\perp$ and

$T_\parallel$, associated with the fluctuations perpendicular to and along the direction of the average magnetization. As stated before, all these temperature scales can, however, be expressed in terms of the Fermi energy and the basic Fermi liquid parameter $\mathfrak{B}_+$.

2. Magnetization versus temperature

In this section we calculate the fluctuations in local magnetization, leading to the definition of the three characteristic temperatures $T_\parallel$, $T_\perp$ and T_c—the last being the Curie temperature while the others characterize fluctuations perpendicular and parallel to the average magnetization $\boldsymbol{M}$, respectively. To calculate the $\langle m_\nu^2\rangle$ we use the definition in equation (6) with the following approximation for the generalized susceptibility in the paramagnon regime

$$\chi_\nu^{-1}(\boldsymbol{q},\omega) = \chi_\nu^{-1}(\boldsymbol{q})[1 - \mathrm{i}\omega/\Gamma_\nu(\boldsymbol{q})] \tag{11}$$

with a quadratic dispersion

$$\chi_\nu^{-1}(\boldsymbol{q}) = \chi_\nu^{-1} + C_{0\nu}q^2 \tag{12}$$

and a linear damping

$$\Gamma_\nu(\boldsymbol{q}) = \gamma_0 q \chi_\nu^{-1}(\boldsymbol{q}). \tag{13}$$

We can then write for $\langle m_\nu^2\rangle$ [4]

$$\langle m_\nu^2\rangle = \frac{k_{\mathrm{F}}T}{\pi}\int_{q_{1\nu}}^{q_{2\nu}} q^2\,\mathrm{d}q\,\chi_\nu^{-1}(\boldsymbol{q})[1 + (3\hbar\gamma_0 q/\pi k_{\mathrm{F}}T)\chi_\nu^{-1}(\boldsymbol{q})]^{-1} \tag{14}$$

where $q_{1\nu} = q_{\mathrm{sw}}$ when $\nu = \perp$ and $q_{1\nu} = 0$ when $\nu = \parallel$, $\gamma_0 = 2\chi_{\mathrm{p}}v_{\mathrm{F}}/\pi$ and $C_{0\nu} \simeq C_0 = 1/12k_{\mathrm{F}}^2 v_{\mathrm{F}}$, v_{F} is the Fermi velocity and k_{F} is the Fermi wave number, β is the electron magnetic moment and $q_{\mathrm{sw}} = M/v_{\mathrm{F}}\hbar\beta\rho$ is the wave vector separating the spin wave and paramagnon regimes in the excitation spectrum. The upper cut-off $q_{2\nu}$ varies with temperature so as to take into account the growing number of thermally excited modes but, as shown in [4], can be effectively set to ∞ in the present calculation.

By introducing a characteristic correlation length

$$l_\nu \equiv (C_0\chi_\nu)^{1/2} \tag{15}$$

we can rewrite equation (14) according to

$$\langle m_\nu^2\rangle = \frac{k_{\mathrm{B}}T\chi_\nu}{l_\nu^3}P(\bar{q}_\nu, \delta_\nu/\alpha_\nu) \tag{16}$$

with $\bar{q}_\nu \equiv q_{1\nu}l_\nu$ and

$$\delta_\nu \equiv T_\nu/T \tag{17}$$

being the crucial parameter controlling the temperature dependence of the fluctuations. The characteristic temperature T_ν is defined as [4]

$$k_{\mathrm{F}}T_\nu \equiv 3\hbar\gamma_0 c_0\alpha_\nu/\pi l_\nu^3 \tag{18}$$

where $\alpha_\perp = \bar{q}_\nu^3$ and $\alpha_\parallel = 1$ due to the different cut-offs in the transverse and longitudinal cases. Note that since in general $l_\perp \neq l_\parallel$ it follows that $T_\perp \neq T_\parallel$. The function P introduced in equation (16) is given by

$$P(a,b) = \int_a^\infty \frac{\mathrm{d}x}{\pi^2}\frac{x^2}{(1+x^2)}\frac{1}{[1+bx(1+x^2)]} \tag{19}$$

and in the following discussion we shall make use of the limiting forms for $P(\bar{q}_\nu, \delta_\nu/\alpha_\nu)$

$$P(\bar{q}_\nu, \delta_\nu/\alpha_\nu) = (2\sqrt{3}/9\pi)\delta_\nu^{-1/3} \qquad \delta_\nu \ll 1 \tag{20}$$

$$P(\bar{q}_\nu, \delta_\nu/\alpha_\nu) = \frac{1}{2\pi^2\delta_\nu}\frac{1}{(1+\bar{q}_\nu^2)} \qquad \delta_\nu \gg 1 \tag{21}$$

leading to the different temperature dependences of $\langle m_\nu^2\rangle$ in the different regions. Introducing the dimensionless forms $\langle \bar{m}_\nu^2\rangle = \langle m_\nu^2\rangle M_0^{-2}$, $\bar{M} = M/M_0$ and

$$\bar{A} = A/a = 1 + 2\bar{M}^{-2}(1-\bar{M}^2)\langle \bar{m}_\perp^2\rangle + \bar{M}^{-2}(1-3\bar{M}^2)\langle \bar{m}_\parallel^2\rangle$$

where $M_0^2 = -a/\gamma_u$ we have:

$$\langle \bar{m}_\parallel^2\rangle = \begin{cases} \frac{1}{5}(T/T_c)^{4/3} & T_\parallel \ll T \\ (T/T_u)^2 |a/\chi_\parallel^{-1}| & T \leqslant T_\parallel \end{cases} \tag{22}$$

where $|\chi_\parallel^{-1}/a| = \bar{A} - 3\bar{M}^2 + 2\bar{M}A' + \frac{1}{2}\bar{M}^2\bar{A}''$, and

$$\langle \bar{m}_\perp^2\rangle = \begin{cases} \frac{1}{5}(T/T_c)^{4/3} & T_\perp \ll T \\ 2(T/T_u)^2/G\bar{M}^2 & T \leqslant T_\perp . \end{cases} \tag{23}$$

Note that in the limit $T \gg T_\nu$, $\langle \bar{m}_\nu^2\rangle$ is independent of ν. In equations (22) and (23) we have introduced the temperature scales

$$k_B T_u/E_F = (6z)^{1/2}\mathcal{B}_+ \qquad z = \rho^2/k_F^3(\rho'' - 3\rho'^2/\rho)E_F \tag{24}$$

$$k_B T_\perp/E_F = (1/\pi^2)(6|\mathcal{B}_+|\tfrac{1}{3}G)^{3/2} \tag{25}$$

$$k_B T_c/E_F = (\pi^2\mathcal{B}_+/5\alpha_1)^{3/4}(1/3\pi^{3/4})z^{3/4} \qquad \alpha_1 = 2\pi^{1/3}/3^{11/6} \simeq 1.228. \tag{26}$$

The longitudinal temperature $T_\parallel$ is the solution of

$$1 - (T_\parallel/T_c)^{4/3} - (T_\parallel/T_0)^{2/3} = 0 \tag{27}$$

where $T_0 = (24\mathcal{B}_+)^{3/2}E_F/\pi^2$. From equation (27) $T_\parallel \simeq T_c$ if $T_0 \gg T_c$. To ensure both that the region $T_\perp < T < T_\parallel$ is wide and $T_\parallel \to T_c$ we require

$$T_\perp \ll T_c \ll T_0. \tag{28}$$

This condition is in fact a restriction on the value of $\mathcal{B}_+$ and for a spherical Fermi surface it is equivalent to requiring $7\times10^{-4} < |\mathcal{B}_+| < 112\times10^{-4}$ while for the 'lens-shaped' Fermi surface discussed in [4] it is $6\times10^{-4} < |\mathcal{B}_+| < 126\times10^{-4}$. As is shown in [4] one can also find similar limits for the case of $ZrZn_2$. While in the spherical and 'lens-shaped' cases the left and right bounds in equation (28) differ by factors of 16 and 21 respectively, in the case of $ZrZn_2$ the difference was found to be 10^4. For $ZrZn_2$ the region in which $T_\perp < T < T_\parallel \leqslant T_c$ can therefore be quite wide.

We now express the magnetization as a function of T using the magnetic equation of state given in equation (7) and the results presented above for $\langle \bar{m}_\nu^2\rangle$. To do this we divide the temperature range into three intervals given below.

2.1. $T < T_\perp$

Since $\langle \bar{m}_\nu^2\rangle \sim T^2$ we can go to the limit of very low temperatures, which means that we can approximate A by [4]

$$\bar{A} = 1 + f(M)(T/T_u)^2 \tag{29}$$

where

$$3f(\bar{M}) = 5\bar{M}^{-2} + 4\bar{M}^{-4} \tag{30}$$

so that

$$\bar{A} = 1 - \tfrac{1}{3}(5\bar{M}^{-2} + 4\bar{M}^{-4})(T/T_u)^2. \tag{31}$$

With this, it follows that the solution of equation (2) for the equilibrium value of the magnetization is

$$\bar{M}^2 \simeq 1 - (T/T_u)^2 + \bar{B} \tag{32}$$

where $\bar{B} \equiv B\gamma_u^{1/2}|a|^{-3/2}$.

2.2. $T_{\parallel} < T < T_c$

In this interval $\langle m_{\perp}^2 \rangle = \langle m_{\parallel}^2 \rangle = \frac{1}{5}(T/T_c)^2$ which means that $\bar{A} = 1 - (T/T_c)^2(1 - \frac{2}{5}\bar{M}^2)$ and the solution of equation (2) is now

$$\bar{M}^2 \simeq 1 - (T/T_c)^{4/3} + \bar{B}. \qquad (33)$$

2.3. $T_{\perp} < T < T_{\parallel}$

In this interval we have in general to solve the full problem but as shown in [4] this can be approximated by

$$\bar{A} \simeq 1 - \tfrac{1}{4}(T/T_u)^2 - \tfrac{3}{5}(T/T_c)^{4/3}[1 - (1/\bar{M}^2)]. \qquad (34)$$

The temperature dependence of the magnetization is therefore

$$\bar{M}^2 \simeq 1 - \tfrac{1}{4}(T/T_u)^2 - \tfrac{3}{5}(T/T_c)^{4/3} + \bar{B} \qquad (35)$$

giving a mixed temperature dependence law for the magnetization.

3. Results and discussion

In this section we use equations (32), (33) and (34) to describe the temperature dependence of the spontaneous magnetization found in $ZrZn_2$ [6, 7]. To do this we have first to estimate $T_{\perp}$, $T_{\parallel}$ and T_u. From equations (24), (25) and (26) these can be obtained from $\mathcal{B}_+$, G and E_F which in turn can be estimated from a comparison of our theoretical expressions for T_c, M_0 and χ_p with the values found experimentally in [6]. In particular we use $T_c = 27$ K, $1/a\rho_0 = 0.98 \times 10^{-6}$ m^3 kg and $M_0/\rho_0 = 3.2$ A m^2/kg where ρ_0 is the density of $ZrZn_2$ (7.29 g cm^{-3}) and we take the value E_F to be 1 Ryd. According to [4] we find that $G^{-1} = 0.05$ and from equation (26) $|\mathcal{B}_+| = 6 \times 10^{-3}$. The values of the temperatures $T_{\perp}$ and T_u are determined by equations (24) and (25) and we find $T_{\perp} = 1.5$ K and $T_u = 20.5$ K. To obtain $T_{\parallel}$ we solved equation (27) numerically to obtain $T_{\parallel} = 25$ K. Using these, the predicted temperature dependence of the magnetization in

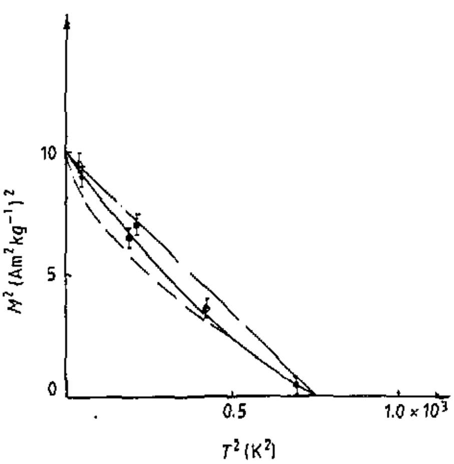

Figure 1. This figure gives the magnetization dependence (35), which is denoted by the full curve, as a function of T^2. The dependence $1 - (T/T_c)^2$ is shown as a chain curve. The broken curve represents the dependence $M^2 = 1 - (T/T)^{4/3}$, which arises when the longitudinal fluctuations T^2 are not taken into account. The experimental points are taken from [6].

$ZrZn_2$ is therefore

$$\tilde{M}^2(T) = \begin{cases} 1 - (T/20.5)^2 & 0\,\mathrm{K} < T < 1.5\,\mathrm{K} \\ 1 - \frac{1}{4}(T/20.5)^2 - \frac{3}{5}(T/27)^{4/3} & 1.5\,\mathrm{K} < T < 25\,\mathrm{K} \\ 1 - (T/27)^{4/3} & 25\,\mathrm{K} < T < 27\,\mathrm{K} \end{cases} \qquad (36)$$

with the mixed temperature law holding over most of the temperature range below T_c.

In figure 1 the experimental temperature dependence of the spontaneous magnetization for $ZrZn_2$ from [6] is compared with equation (35). Also plotted are curves corresponding to $M(T)^2 = 1 - (T/T_c)^2$ and $M(T)^2 = 1 - (T/T_c)^{4/3}$. Of these curves, equation (35) clearly provides the best fit to experimental data.

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