Nuclear magnetic susceptibility of metals with magnetic impurities

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Abstract

We consider the contribution of magnetic impurities to the nuclear magnetic susceptibility $\chi$ and to the specific heat $C$ of a metal. The impurity contribution to the magnetic susceptibility has a $1/T^2$ behaviour, and the impurity contribution to the specific heat has a $1/T$ behaviour, both in an extended region of temperatures $T$. In the case of a dirty metal the RKKY interaction of nuclear spins and impurity spins is suppressed for low temperatures and the main contribution to $C$ and $\chi$ is given by their dipole-dipole interaction.

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The magnetic impurity contribution to the nuclear specific heat of a solid was established in the paper\textsuperscript{[1]}. In that paper the magnetic dipole-dipole interaction between the nuclear magnetic moments $\mu_n$ and the magnetic moments of impurities $\mu_{\text{imp}}$ was considered. This interaction has the form

$$V_{\text{d-d.}} = \frac{\mu_1 \mu_2 r^2 - 3(\mu_1 r)(\mu_2 r)}{r^5}. \quad (1)$$

The interaction (1) takes place both in metals and insulators. Experiments\textsuperscript{[2], [3], [4]} study the metal case. For metals the RKKY interaction is present in addition to (1),

$$V_{\text{RKKY}} = -\kappa \frac{\mu_n \mu_{\text{imp}}}{r^3} \cos(2k_F r)e^{-r/r_0}, \text{ for } k_F r \gg 1. \quad (2)$$
The characteristic length $r_0$ describes a suppression of the RKKY interaction by nonmagnetic impurities. It is of the order of the electron mean free path and is inversely proportional to the concentration of the nonmagnetic impurities. The value of the parameter $\kappa$ defines the relative weight of the RKKY interaction (2) related to the universal interaction (1), that does not depend on the impurity concentration. The parameter $\kappa$ can not be found precisely. It is known only, that $\kappa$ increases monotonously with an increase of the nuclear charge $Z$, leading to $\kappa \gg 1$ for $Z \gg 1$. It was shown in the paper [1], that at low temperatures the main part of the magnetic impurity contribution to the nuclear specific heat comes from nuclei situated at large distances from the magnetic impurity. Increasing the concentration of nonmagnetic impurities one can suppress the RKKY interaction and come to the situation, where only the interaction (1) is important. Then an interesting possibility appears to derive the value of the parameter $\kappa$ in (2) by comparing the values of $C$ and $\chi$ for clean and dirty metals.

For the sake of simplicity we consider the case, where the value of the nuclear spin is $S_n = 1/2$, which is valid for the compound $PtFe_x$ experimentally investigated in [2]. $^{195}Pt$ is the only stable isotope of platinum that has a nonzero spin, $S = 1/2$, $\mu = 0.6095\mu_N$. The naturally available $Pt$ is an isotopic mixture, containing about 34% of $^{195}Pt$. According to [2] the temperature of the nuclear spin spin-glass transition is definitely lower than $10^{-6}K$. The temperature of the impurity spin spin-glass transition is much higher, lying in the interval $T_{Fe} = (2.7 - 8.5)10^{-3}K$ for $Fe$ impurities. There exists hence an extended region of temperatures $T_{cn} << T << T_{Fe}$. In this region the impurity ($Fe$) spins are frozen and do not contribute to $C$ and $\chi$, contrary to the nuclear spins, which are uncorrelated. In the first approximation for the aforementioned temperature region we can put $T_{cn} = 0$ and $T_{Fe} = \infty$. Then the two systems, the nuclear spin system and the impurity spin system, considered separately, do not posses characteristic temperature scales. A characteristic temperature scale $\tau$ appears only after considering the interactions (1 2) between the two systems.

To find the energy scale $\tau$, let us first consider an effective magnetic field $H_n$ acting on a nuclear spin. This field is a sum of an external magnetic field $H_0$ and an internal magnetic
field $H_1(r_n)$.

$$H_n = H_0 + H_1(r_n) \quad (3)$$

The free energy $f_n$ of a single nucleus with a $S = 1/2$ spin in the magnetic field $H_n$ is

$$f_n = -T \ln[2 \cosh \frac{\mu_n H_n}{2T}]. \quad (4)$$

The magnetic moment of the nucleus $m_n$ and the contribution of the nucleus to the heat capacitance $C_n$ are then given by a differentiation of $f_n$

$$m_n = -\frac{\partial f_n}{\partial H_n} = \frac{\mu_n H_n}{2} \tanh \frac{\mu_n H_n}{2T}, \quad (5)$$

$$C_n = -\beta^2 \frac{\partial^2 f_n}{\partial \beta^2} = \left(\frac{\mu_n H_n}{2T}\right)^2 \frac{1}{\cosh^2 \frac{\mu_n H_n}{2T}}, \quad \beta = 1/T.$$

Performing a sum of $C_n$ and $m_n$ over all nuclei one finds the magnetisation moment $M$ and the heat capacity $C$ per unit volume. It is then convenient to separate out the contribution of a clean metal.

$$M = M_0 + \frac{1}{V} \sum_n (m_n - m_0), \quad (6)$$

$$C = C_0 + \frac{1}{V} \sum_n (C_n - C_{n0})$$

The values of $m_0$ and $C_{n0}$ are defined by the formula (5) taking $H_n = H_0$.

For a low concentration of magnetic impurities their influence regions do not overlap, and one can consider a single impurity placed at the origin of coordinates. A contribution to $C$ and $\chi$ comes from a region near the magnetic impurity, which is much larger than the lattice spacing, so the summations can be replaced by integrations with respect to $d\mathbf{r}$. The magnetic impurity spins are in a spin-glass state and have random directions with respect to the external magnetic field, therefore one needs to average the resulting expressions with respect to the direction of the magnetic impurity spin.

Let us consider the absolute values of $M$ and $M_0$

$$M = M \frac{H_0}{H_0}, \quad M_0 = M_0 \frac{H_0}{H_0}. \quad (7)$$
Then
\[ M = M_0 + \frac{\mu_n}{4} n_{\text{imp}} n_n \int_{-1}^{1} dx \int d\mathbf{r} \left[ \frac{\mathbf{H}_0 \mathbf{H}_1}{H_n H_0} \tanh \frac{\mu_n H_n}{2 T} - \tanh \frac{\mu_n H_0}{2 T} \right], \tag{8} \]
\[ C = C_0 + \frac{1}{2} n_n n_{\text{imp}} \int_{-1}^{1} dx \int d\mathbf{r} \left[ \frac{(\mu_n H_n)^2}{2 T} \frac{1}{\cosh^2 \frac{\mu_n H_n}{2 T}} - \left( \frac{\mu_n H_0}{2 T} \right)^2 \frac{1}{\cosh^2 \frac{\mu_n H_0}{2 T}} \right]. \]

Here \( x = \cos \theta \), \( \theta \) is the angle between \( \mathbf{H}_0 \) and \( \mathbf{H}_1 \) in (3). Differentiating \( M \) with respect to \( H_0 \) (7) and taking \( H_0 = 0 \), one finds the impurity contribution to the nuclear magnetic susceptibility at zero magnetic field, a characteristic usually measured experimentally.

\[ \chi = \chi_0 [1 - J(T)], \quad \chi_0 = \frac{\mu_n^2 n_n}{4 T}, \tag{9} \]
\[ J(T) = n_{\text{imp}} \int d\mathbf{r} \left[ \frac{1}{3} \tanh^2 \frac{\mu_n H_1}{2 T} \right] + \frac{2}{3} \left( 1 - \frac{2 T}{\mu_n H_1} \tanh \frac{\mu_n H_1}{2 T} \right) \]

Let us first consider the case where the magnetic dipole-dipole interaction (1) plays the major role and the RKKY interaction can be neglected. In this case the expressions for the temperature dependence of the specific heat \( C \) were derived in (1). For the magnetic susceptibility \( \chi \) an integration with respect to \( d\mathbf{r} \) in (9) gives after some mathematics the value of \( J \), which we denote as \( J_1 \)

\[ J_1(T) = \frac{\theta_1}{T}, \tag{10} \]
\[ \theta_1 = \frac{2 \pi}{3} \mu_{\text{imp}} \mu_n n_{\text{imp}} [1 + \frac{1}{2 \sqrt{3}} \ln(2 + \sqrt{3})] I_1, \]
\[ I_1 = \int_0^\infty dz \frac{1}{z^2} \left[ \frac{1}{3} \tanh^2 z + \frac{2}{3} (1 - \frac{\tanh z}{z}) \right] = 1.137. \]

As a result the impurity contribution to \( \chi \) is proportional to \( 1/T^2 \) (9, 10). The magnetic susceptibility of a clean metal has one more \( 1/T^2 \) correction, related to the spin-spin interaction among nuclei. The expansion of \( \chi \) in powers of \( 1/T \) has then the form

\[ \chi = \frac{\mu_n^2 n_n}{4 T} (1 - \frac{\theta}{T}), \quad \theta = \theta_n + \theta_1. \tag{11} \]

One can roughly estimate the value of \( \theta_n \) as \( \theta_n \sim \mu_n^2 n_n \). A relation of \( \theta_1 \) to \( \theta_n \) contains a small parameter \( x_{\text{imp}} = \frac{n_{\text{imp}}}{n_n} \), but also a large parameter \( \frac{\mu_{\text{imp}}}{\mu_n} \sim 10^4 \). Therefore, already for a small concentration of magnetic impurities \( x_{\text{imp}} \sim 10^{-4} \), the impurity part makes the main contribution to \( \theta \) in (11).
Let us consider now another limiting case, where the RKKY interaction is much stronger than the magnetic dipole interaction. For the RKKY interaction (2) using the expression for the impurity field \( H_1(r) \)

\[
H_1(r) = \kappa \frac{\mu_{\text{imp}}}{r^3} \cos(2k_F r)e^{-r/r_0}, \tag{12}
\]

and performing the substitution of the integration variable in (8,9) \( r \rightarrow z \), where

\[
z = \frac{\mu_{\text{imp}}\mu_n\kappa}{2Tr_0^3}, \tag{13}
\]

one gets

\[
\chi(T) = \chi_0[1 - J_2(T)], \tag{14}
\]

\[
J_2(T) = \frac{\theta_2(T)}{T}, \quad \theta_2(T) = \frac{2\pi}{3} \mu_{\text{imp}}\mu_n\kappa n_{\text{imp}} I_2(T),
\]

\[
I_2(T) = \int_0^\infty \frac{dz}{z^2} \left[ \frac{1}{3} \tanh^2 y + \frac{2}{3} \left( 1 - \frac{\tanh y}{y} \right) \right],
\]

where

\[
y = y(z) = z \cos\left(\frac{z_0}{z}\right)^{1/3} \exp\left[-\left(\frac{z_1}{z}\right)^{1/3}\right]. \tag{15}\]

The parameters \( z_0, z_1 \) are related to the two characteristic temperature scales

\[
z_0 = \frac{T_0}{T}, \quad T_0 = 12\pi^2 n_e \mu_n \mu_{\text{imp}} \kappa,
\]

\[
z_1 = \frac{T_1}{T}, \quad T_1 = \frac{\mu_{\text{imp}}\mu_n\kappa}{2r_0^3}.
\]

Here \( n_e \) is the electronic density \( n_e = \frac{k_F^3}{3\pi^2} \).

In analogy to (8) one obtains the impurity contribution to the specific heat at zero external magnetic field

\[
C - C_0 = \frac{2\pi}{3} \frac{n_{\text{imp}}\mu_{\text{imp}}n_n\mu_n\kappa}{T} I_3(T), \tag{17}
\]

\[
I_3(T) = \int_0^\infty \frac{dz}{z^2} \frac{y^2}{\cosh^2 y}.
\]

Here the parameter \( y = y(z) \) is defined in (15). Let us now give an analysis of the expressions for \( \chi \) (14) and \( C \) (17) in different limiting cases.
In simple metals like Pt, the electron density $n_e$ is equal to the density of nuclei and is of order of $a^{-3}$, where $a$ is the lattice spacing. For the clean case (no nonmagnetic impurities) the mean free path $r_0 \gg a$. Then from the formulas we find that there exists an extended region of $T$, $T_1 \ll T \ll T_0$, such that one can assume $z_1 = 0$, $z_0 = \infty$. This corresponds to $I_2 = 0.723$ and $I_3 = 0.636$. We see that in this region the values of $I_2$ and $I_3$ do not depend on $T$, leading to $C \sim 1/T$ and $\chi \sim 1/T^2$. The oscillating behaviour of the RKKY interaction does not show itself in this region, the oscillations of $V_{RKKY}$ are averaged out. The graphs of $I_2(z_0)$, $I_3(z_0)$ for $z_1 = 0$ are shown in the Fig. 1a, 1b. One can see from the Fig. 1a, 1b that the values of $I_2(z_0)$, $I_3(z_0)$ oscillate only by about two times in the whole region $0 < z_0 < \infty$. The asymptotic value is approached slowly, since $I_2$ and $I_3$ depend effectively on $z_1^1/3$. Note that for the nuclear specific heat of PtFe, the $C \sim 1/T$ law extends through four orders of magnitude of $T$.

For a dirty metal there exists a temperature region $T_{cn} \ll T \ll T_1 < T_0$, where on one hand the nuclear spins are uncorrelated, and on the other hand one needs to account for the suppression of the RKKY interaction related to the finite electron mean free path $r_0$. In this region of temperatures one has $z_0 = \infty$ and $z_1 \gg 1$ and the parameters $I_2$, $I_3$ have the asymptotics

$$I_2 \simeq 0.09 \frac{1}{z_1} \ln^3 z_1, \quad I_3 \simeq 0.25 \frac{1}{z_1} \ln^2 z_1. \quad (18)$$

Therefore, for $T \ll T_1$, a saturation of the RKKY contribution to the impurity part of $C$ and $\chi$ occurs.

$$C - C_0 \sim \frac{\kappa}{T_1} \ln^2 \frac{T_1}{T}, \quad \chi - \chi_0 \sim \frac{\kappa}{TT_1} \ln^3 \frac{T_1}{T}. \quad (19)$$

The graphs of $I_2, I_3$ as functions of $z_1$ for $z_0 = \infty$ are given in the Fig. 2a, 2b.

Let us now go further down in temperatures. For a dirty metal, when the temperature is lowered, a situation occurs, where the saturated RKKY contribution to $C$ and $\chi$, which is proportional to the large parameter $\kappa$, becomes lower, than the dipole-dipole interaction, which is proportional to $1/T$. This happens for $T \ll \frac{T_0}{\kappa}$. Increasing
the concentration of nonmagnetic impurities one can narrow the temperature window $T_1 \ll T \ll T_0$, where the strong RKKY interaction plays role, and extract the contribution of the magnetic dipole-dipole interaction $\mathcal{I}$. Therefore, taking into account that $\kappa \gg 1$, we await a very strong dependence of the impurity part of $C$ and $\chi$ on the concentration of the nonmagnetic impurities in the metal.

As a conclusion, there exist two distinct temperature regions, where the law $C - C_0 \sim 1/T$ is fulfilled. In the first region, $T_1 \ll T \ll T_0$, the impurity contribution to $C$ is related to the RKKY interaction. In the second region, $T_{cn} \ll T \ll \frac{T_1}{\kappa}$, the main contribution to $C(T)$ is given by the magnetic dipole-dipole interaction $\mathcal{I}$. In the interval of temperatures $\frac{T_1}{\kappa} < T < T_1$ the impurity contribution to $C - C_0$ has only a weak logarithmic temperature dependence. Measuring the impurity contribution to $C$ and $\chi$ one can extract the parameter $\kappa$ of the RKKY interaction $\mathcal{I}$.

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FIGURE CAPTIONS

Fig. 1a: $I_2$ as a function of $z_0$ for $z_1 = 0$.

Fig. 1b: $I_3$ as a function of $z_0$ for $z_1 = 0$.

Fig. 2a: $I_2$ as a function of $z_1$ for $z_0 = \infty$.

Fig. 2b: $I_3$ as a function of $z_1$ for $z_0 = \infty$. 
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Fig. 1a, Dyugaev et al.
Fig. 1b, Dyugaev et al.
Fig. 2a, Dyugaev et al.
Fig. 2b, Dyugaev et al.