Field dependences of magnetization and specific-heat coefficient in a nearly magnetic system: Liquid ^{3}He and strong Pauli paramagnets

M. T. Béal-Monod and E. Daniel*
Laboratoire de Physique des Solides, associé au Centre National de la Recherche Scientifique, Université de Paris-Sud, 91400 Orsay, France

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We analytically derive the first-order magnetic field dependences of the magnetization M and of the coefficient γ of the specific heat, in a nearly magnetic paramagnet, at low temperature.

The contribution of spin fluctuations, "paramagnons," to the temperature dependence of the spin susceptibility of a nearly magnetic Fermi liquid was calculated long ago,[1] for a parabolic band, with no adjustable parameter, in terms of the Stoner enhancement S given by experiments. The agreement with the data on normal liquid ^{3}He was very good for all available values of S. An extension was recently made[2] to account for a more general band structure. Polarized liquid ^{3}He as well as strongly exchange-enhanced metallic paramagnets, involving strong spin-spin interaction I among fermions, have received renewed interest due to recent measurements in high magnetic fields[3]: deviations of the magnetization $M(T,H)$ from linearity with the field H,H dependence of the coefficient of the specific heat ($\gamma=[C(T,H)/T]_{T=0}$). As recalled in Ref. 3, due to quantum effects,[4] the critical exponents at the ferromagnetic transition at 0 K, should assume their mean-field values. The critical spin fluctuations will not affect the $T=0$ powers of the Stoner enhancement S. It was thus expected[3] that the H dependence of M would qualitatively be the same at 0 K, as that of the Stoner-type result.[5] But at finite T, paramagnons modify by one power of S the zero-field T dependence of the susceptibility,[1] compared to the Stoner result.

It is the purpose of the present paper to provide analytical formulas, at finite H and T, for $M(T,H)$ and $\gamma(H)$, taking paramagnons into account. We give the result for a parabolic band but it could be generalized for a given band structure. Other authors[6] recently computed such field dependences, but their results were given under the form of complicated integrals difficult to compare with our present results; we note though that in Ref. 6 H always appears through the combination SH while here it rather involves $S^{3/2}H$. This last combination, we believe, ought to be the correct one due to the above-mentioned quantum effects imposing the critical exponent in $H\sim M^{\delta}$ to assume its mean-field value $\delta=3$.

We start with the same paramagnon formalism as that in Ref. 1 (hereafter referred to as BMMF) with the same notations, although M is used for the total magnetization, and to simplify we suppose that the number of particles per unit volume $N/V=1$, which does not affect the final result:

$$F(T,M)=G_0(T,B)+I(1-M^2)/4+\Delta F(T,B)-MB \quad . \tag{1}$$

B is defined by $(\partial F/\partial B)_{T,M}=0$; besides, $H=\partial F/\partial M$. These two equations yield

$$M=[-\chi_{\text{Pauli}}B+(\partial\Delta F/\partial B)]_{-B=H+IM/2} \quad . \tag{2}$$

ΔF is given by the same diagrams as in BMMF:

$$\Delta F=(T/2)\sum_{\vec{q},\omega}\{\ln(1-I^2\chi^{0++}\chi^{0--})+I^2\chi^{0++}\chi^{0--}+\ln(1-I\chi^{0+-})+I\chi^{0+-}+\ln(1-I\chi^{0-+})+I\chi^{0-+}\} \quad , \tag{3}$$

in terms of the dynamic susceptibilities in the absence of interactions,

$$\chi^{0\alpha,\beta}=-\sum_{\vec{p}}\{(f^{\alpha}_{\vec{p}}-f^{\beta}_{\vec{p}+\vec{q}})/[\omega+\xi^{\alpha}_{\vec{p}}-\xi^{\beta}_{\vec{p}+\vec{q}}+i\eta\,\mathrm{sgn}(\xi^{\beta}_{\vec{p}+\vec{q}}-\xi^{\alpha}_{\vec{p}})]\} \quad ;$$

α,β are spin indices, the f's are the Fermi functions expressed in terms of the fermions energies in atomic units $\xi^{\alpha,\beta}_{\vec{p}}=p^2/2-\mu+(\alpha,\beta)B$, μ is the chemical potential, B is in units of energy. All that is reproduced from BMMF. Now straightforward algebra gives

$$\mathrm{Re}\chi^{0++}=N(E_F)\left[\frac{h_-}{2}+\frac{1}{8\bar{q}}(h_-^2-x^2)\ln\frac{h_-+x}{h_--x}-\frac{1}{8\bar{q}}(h_-^2-y^2)\ln\frac{h_-+y}{h_--y}\right] \quad ,$$

$$\mathrm{Re}\chi^{0--}(h)=\mathrm{Re}\chi^{0++}(-h) \quad , \tag{4}$$

$$h_{\mp}=(1\mp h)^{1/2},\ h=B/E_F,\ x=\frac{\bar{\omega}}{4\bar{q}}+\bar{q},\ y=\frac{\bar{\omega}}{4\bar{q}}-\bar{q},\ \bar{\omega}=\frac{\omega}{E_F},\ \bar{q}=\frac{q}{2k_F}$$

$$\mathrm{Re}\chi^{0+-} = N(E_F)\left[\frac{h_+x_+ - h_-y_+}{4\bar{q}} + \frac{1}{8\bar{q}}(h_+^2 - x_+^2)\ln\frac{h_+ + x_+}{h_+ - x_+} - \frac{1}{8\bar{q}}(h_-^2 - y_+^2)\ln\frac{h_- + y_+}{h_- - y_+}\right] ,$$

$$\mathrm{Re}\chi^{0-+}(\bar{\omega},h) = \mathrm{Re}\chi^{0+-}(-\bar{\omega},h) = \mathrm{Re}\chi^{0+-}(\bar{\omega},-h) , \tag{5}$$

$$x_\pm = x \pm h/2\bar{q},\ y_\pm = y \pm h/2\bar{q} .$$

$N(E_F)$ is the density of states per spin at the Fermi level, k_F the Fermi momentum. In the region of interest, where $\mathrm{Im}\chi^{0\alpha\beta}$ is linear with $\bar{\omega}$, one has

$$\mathrm{Im}\chi^{0\alpha\beta} = N(E_F)(\pi/8)(\bar{\omega}/\bar{q})\lambda^{\alpha\beta} ,\quad \lambda^{\pm\pm} = \theta(h_\mp^2 - y^2)\theta(h_\mp^2 - x^2) ,\quad \lambda^{\pm\mp} = \theta(h_\mp^2 - y_\pm^2)\theta(h_\pm^2 - x_\pm^2) . \tag{6}$$

$\lambda^\mp$ is defined in a domain symmetric from the one of $\lambda^\pm$ with respect to the $\bar{q}$ axis. The above-defined domains played a key role in the further integrations over $\bar{q}$ and $\bar{\omega}$. Note that in the $(\bar{\omega},\bar{q})$ plan there are gaps for small $\bar{q}$ and $\bar{\omega}$ around the origin, for the transverse susceptibilities due to the presence of B. The $\bar{\omega}=0$, $\bar{q} < 1$, and $h < 1$ expansions are useful:

$$\mathrm{Re}\chi^{0+-}(\bar{\omega}=0) = \mathrm{Re}\chi^{0-+}(\bar{\omega}=0) \approx N(E_F)\left[1 - \frac{\bar{q}^2}{3} - \frac{h^2}{8}\left[\frac{1}{3} + \frac{\bar{q}^2}{5}\right] + \cdots\right] ,$$

$$(\mathrm{Re}\chi^{0++}\mathrm{Re}\chi^{0--})_{\bar{\omega}=0} \approx N^2(E_F)\left[1 - \frac{2\bar{q}^2}{3} - \frac{h^2}{2}\left[1 + \frac{2\bar{q}^2}{3}\right] + \cdots\right] , \tag{7}$$

$$\mathrm{Re}\chi^{0++}(\bar{\omega}=0) + \mathrm{Re}\chi^{0--}(\bar{\omega}=0) \approx 2N(E_F)\left[1 - \frac{\bar{q}^2}{3} - \frac{h^2}{8}(1+\bar{q}^2) + \cdots\right] .$$

We found it more convenient to calculate $[\Delta F(B) - \Delta F(0)]$ which makes immediately evident the finite terms in B. We find

$$\Delta F(T,B) - \Delta F(T,0) = \chi_{\mathrm{Pauli}}[\alpha(\bar{T},\bar{I})B^2/2 + \beta(\bar{T},\bar{I})B^4/(4E_F^2) + \cdots] \tag{8}$$

in appropriate units, where α and β are functions of $\bar{T} = T/T_F$ and of the dimensionless interaction $\bar{I} = IN(E_F)$, or the Stoner factor $S = (1-\bar{I})^{-1}$; in units of the square of the magnetic moment, χ_{Pauli} is $2N(E_F)$. Then (2) and (8) yield

$$M(T,H) = \chi_{\mathrm{Pauli}}H\left[\frac{1-\alpha}{1-\bar{I}+\alpha\bar{I}} - \beta\frac{H^2}{E_F^2}\frac{1}{(1-\bar{I}+\alpha\bar{I})^4} \cdots\right] . \tag{9}$$

Expanding further, for low $T \ll T_{\mathrm{sf}}$, and low $H \ll T_{\mathrm{sf}}/\sqrt{S}$, with the spin fluctuation temperature $T_{\mathrm{sf}} = (1-\bar{I})T_F$ (T_F the bare Fermi temperature), we obtain

$$M(T,H) = S\chi_{\mathrm{Pauli}}H\left[1 - \alpha_1 S^2\frac{T^2}{T_F^2} - \beta_0 S^3\frac{H^2}{T_F^2} + (\beta_1 + 4\alpha_1\beta_0)S^2\frac{T^2}{T_F^2}S^3\frac{H^2}{T_F^2} \cdots\right] . \tag{10}$$

On the other hand we find (for a parabolic band)

$$\beta_0 \approx \tfrac{1}{6},\quad \alpha_1 \approx \pi^2/6,\quad \beta_1 \approx 23\pi^2/24^2,\quad H \ll T , \tag{11}$$

$$\beta_0 = \tfrac{1}{6},\quad \alpha_1 \approx \pi^2/4,\quad \beta_1 \approx 27\pi^2/24^2,\quad T \ll H .$$

Several remarks arise at that point:

(i) As recalled above and detailed in Ref. 4, the fluctuations do not affect the $T=0$ behaviors: $\beta(T=0) \equiv \beta_0$ is the value computed in the Stoner-Wohlfarth theory[5]; $\alpha(T=0) \equiv \alpha_0$ is assumed to have been incorporated in the definition of $\bar{I}$ in (9) and is extracted from experiments. In contrast, the T dependences of α and β do diverge with S: The fluctuations greatly enhance the finite temperature dependence of M. For comparsion, the analog of (10) in the absence of fluctuations, as given in Ref. 5, would contain ST^2/T_F^2 instead of S^2T^2/T_F^2.

(ii) $(M/H)_{H=0}$ must be compared to the result $\chi(T,0)$ in BMMF. The $H \ll T$ limit of α in (11) is $\pi^2/6 = 4\pi^2/24$ to be compared with the result in BMMF, $3.2\ \pi^2/24 = [4-(8/\pi^2)]\pi^2/24$. The extra term $8/\pi^2$ arose from less-important contributions in the integrals that we have neglected here; nevertheless the coefficient in BMMF, $3.2\pi^2/24 \simeq 1.3$, was more accurate than (although very close to) the one we have here, $4\pi^2/24 \simeq 1.6$.

(iii) The first term in (9) appeared with such a form in the theory of Moriya and Kawabata.[7] As already pointed out elsewhere,[8] their formalism is identical to the one derived earlier in BMMF that we use here.

(iv) As emphasized in Refs. 3, 8, and 9, one notes the scaling in ST/T_F and the one in $S^{3/2}H/T_F = \sqrt{S}H/T_{sf}$ which is in agreement with the relation, at the ferromagnetic transition ($T=0,\bar{I}=1$), $H \sim M^{\delta-3}$, where one replaces M by $\chi H \sim SH$. Thus the results of Ref. 6, which do not contain the same scaling, appear doubtful to us.

(v) From (10), it is clear that the deviations of M from linearity with respect to H are less pronounced when T increases. To observe the nonlinear behavior, one would need higher fields at high temperatures than at low T. For a different band structure, when α_1, β_0, and β_1 would be all negative instead of positive here, then $M(T,H)/H$ would increase with H (instead of decreasing here), but less and less so when the temperature increases; this is in qualitative agreement with what is found in $TiBE_2$.[10]

(vi) From the Maxwell relation[1,3] $\partial M/\partial T = \partial S/\partial H$, we deduce $\partial^2 M/\partial T^2 = \partial\gamma/\partial H$, which thus yields with (10) (for a parabolic band)

$$\gamma(H)-\gamma(0) = -\chi_{\text{Pauli}}\alpha_1\left(\frac{S^{3/2}H}{T_F}\right)^2\left[1-\left(\frac{\beta_1}{2\alpha_1}+2\beta_0\right)\left(\frac{S^{3/2}H}{T_F}\right)^2\cdots\right] . \tag{12}$$

As pointed out in Ref. 3, the lowest field dependence of $\gamma(H)-\gamma(0)$, i.e., the first term in (12), follows from the curvature of $\chi(T,H=0)$ at $T=0$. However, at higher fields, or strong enough values of $[\beta_1/(2\alpha_1)+2\beta_0]$, the H^4 term in (12) being of opposite sign, may counterbalance the first one. This seems to be the case in $TiBe_2$ (Ref. 11) where, at $H=7$ T, $\gamma(H) \leq \gamma(0)$, while $\chi(T,H=0)$ first increases with T; however, it was pointed out to us[12] that at 7 T, $\chi(T,H)$ decreases rather than increases when T increases. Therefore the predicted values given in Ref. 3 for the relative variation of $\gamma(H)$ at 7 T was erroneous since it corresponded to the extrapolation of the first term in (12) to a field value (7 T) where $\chi(T,H)-\chi(0,H)$ does not have the same sign that $\chi(T,H\simeq 0)-\chi(0,H\simeq 0)$; in that case, the 2nd term in (12), but also higher-order ones (not computed here), ought to be checked. In contrast, for Pd, where M remains linear in H up to about 35 T (see Ref. 39 in Ref. 3), the H^2 term in (10) and the H^4 term in $\gamma(H)$ must be very tiny and, from $\chi(T,0)$ increasing with T, one expected[3] a small increase of $\gamma(H)$ with H; some experiments (Ref. 34 of Ref. 3) found a strong decrease while others[13] did observe recently a small increase.

(vii) A discussion was given elsewhere[8] concerning the T variation of the nuclear relaxation rate T_1^{-1} in strongly enhanced paramagnets, recovering the proportionality to $T\chi(T)$ already proposed by Moriya and Ueda[14] but also pointing out a scaling in T/T_{sf} below and above T_{sf}. If $\chi(T,H)$ replaces $\chi(T,0)$ in the finite field dependence of T_1^{-1}, then one can write at low $T \ll T_{sf}$ and $H \ll T_{sf}/\sqrt{S}$, $T_1^{-1} \propto T[M(T,H)/H]$, with $M(T,H)$ given by (10). This is qualitatively analogous to the result derived in Ref. 15 but with different numerical coefficients. We note however that Ref. 15 used a supposedly general expansion for small $\bar{q}$ and $\bar{\omega}/\bar{q}$ for the transverse susceptibility χ^{0-+}, which appears incomplete to us; such terms as $h/\bar{q}^2$ [that we have in (5)] are missing, for instance. The respective ratios of the four small quantities $\bar{q}$, $\bar{\omega}/\bar{q}$, $\bar{T}$, and h must be handled with special care since the domain of integration in the $(\bar{\omega},\bar{q})$ plan is very tricky as can be seen from (6).

Our main result (10), with positive coefficients (11) can apply to two physical cases: liquid ^3He, where there is no band structure and (11) applies as such, and UAl_2 which behaves the same way (Ref. 7 of Ref. 3), but whose band structure has not been calculated so far. Recent experiments on this last material[16] exhibit a variation of M vs H in qualitative agreement with our result: M deviates from linearity with H around 15 T at low T but remains linear at much higher fields at high T, for which we expect that one would need higher fields to observe nonlinearity. A quantitative comparison is difficult though, since our result only applies for $T \ll T_{sf}$, and for $H \ll T_{sf}/(S\beta_0)^{1/2}$, conditions which are not all fulfilled in the experiments. One remark: for this compound where $S \sim 4$, $(S\beta_0)^{1/2} \simeq 1$ so that the characteristic field equal to $T_{sf}/(S\beta_0)^{1/2}$ is practically equal to T_{sf}; but for higher values of S, for instance in $TiBe_2$ ($S \sim 65$), the two values will be different. For liquid ^3He, we expect deviations from linearity for M vs H to be of order 2% at 100 kG at 15 mK, at melting pressure where $S \sim 20$; but in confined geometries, if S can be as large as 60,[17] then the corresponding effects are expected to be larger, ~25%. The characteristic field where nonlinearity is expected to occur is estimated to be about 90 T at melting pressure for the bulk, but may be reached below ~18 T in confined geometry if an S value of ~60 can be achieved.

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*Permanent address: Université Louis Pasteur, 4 rue Blaise Pascal, F-6700 Strasbourg, France.

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