

Field dependence of the properties of strongly exchange enhanced paramagnets. LuCo₂ and TiBe₂

M. T. Béal-Monod

*Laboratoire de Physique des Solides, Université de Paris-Sud,
91405-Orsay, France*

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Recent field dependences of the specific heat, the magnetization, and the magnetoresistivity of strongly enhanced paramagnets are discussed using existing theoretical results and phenomenological conjectures.

This paper is motivated by the recently increased interest in enhanced Pauli paramagnetism, which appears to be a common feature among materials *a priori* as different as: liquid ³He,¹ or Pd,² Ce compounds like Ce(In, Sn)₃,³ CeAl₃,⁴ some actinides or actinide intermetallic compounds like UAl₂,⁵ but also TiBe₂ (Ref. 6) and LuCo₂.⁷ This list is obviously not exhaustive. All these materials experimentally exhibit low-temperature behaviors characteristic of strongly exchange enhanced paramagnets or nearly magnetic Fermi liquids: a constant value of the zero-temperature susceptibility strongly enhanced compared to the Pauli value, a specific heat varying linearly with the temperature with a strong value of the coefficient, etc. The purpose of this paper concerns more specifically a discussion of the recently measured⁷ field dependences of the specific heat of LuCo₂ and of the magnetization⁶ of TiBe₂, of particular interest since there exist very few experimental and theoretical studies of such field effects. Expectations for the magnetoresistivity will also be examined.

I. FIELD DEPENDENCE OF THE SPECIFIC HEAT

The data⁷ on LuCo₂ show that, for fixed T (< 20 K), C/T decreases when an external field is applied ($H < 9.98$ T). As recalled below, it then follows from thermodynamics that the zero-field susceptibility $\chi(T, H=0)$ should decrease for increasing temperature T . Therefore the available data⁸ on $\chi(T, 0)$ where at low T , $\chi(T, 0)$ was found to increase with T , do not reflect the approach of the ground state and ought to be pursued at lower T . They have not been so far, because of the difficulty to get rid of extra (impurity) contributions. It has been shown in another case,³ Ce(In,Sn)₃, that subtraction of the impurity part can be done with a reliable accuracy. It is then hoped that the same procedure would apply to LuCo₂ at very low T in order to check whether the temperature variation of $\chi(T, H=0)$ is consistent with the field dependence of the specific heat or not,

in which case there would be a failure of thermodynamics and one should understand why.

The first paper⁹ which related the field dependence of C/T and the T dependence of χ was written in the framework of the enhanced spin-fluctuation (or "paramagnon") theory,¹⁰ and dealt with one parabolic band of strongly interacting itinerant fermions with a spherical Fermi surface; the result directly applied to liquid ³He in its normal phase. A generalization of this calculation, to account for an arbitrary band shape, was recently^{11,12} published. A glance at the theoretical results of Refs. 9, 11, and 12 clearly shows the importance of the above suggested experiments [simultaneous measurements of $\chi(T, H=0)$ and $C(T, H)/T$ for $T \rightarrow 0$]:

(i) The *sign* of the curvature of $\chi(T)$ at $T=0$ gives the sign of the variation with H of C/T and vice versa. This just follows in a straightforward manner from pure thermodynamics: From the Maxwell relation $\delta M/\delta T = \delta S/\delta H$ (where M is the magnetization and S the entropy) it follows that

$$\frac{\delta(C/T)_{T=0}}{\delta H} = H \frac{\delta^2 \chi_{H=0}}{\delta T^2} \quad (1)$$

Therefore, since on LuCo₂ it was observed that C/T decreases when H increases, thermodynamics imposes that $\chi(T, 0)$ decreases when T increases from 0 K [independently of any further increase at much higher T (Ref. 8)]. If such a decrease can be checked in future experiments it will remain to understand why it is followed by the increase at higher T observed in Ref. 8. For a Fermi-liquid ground state where $\chi(T, 0)$ varies quadratically with T , (1) reads as well

$$\left[\frac{C(T, H) - C(T, 0)}{T} \right]_{T=0} = \frac{H^2}{T^2} [\chi(T, 0) - \chi(0, 0)] \quad (2)$$

(ii) It is in the *magnitude* of the variation with H of C/T that the paramagnons (when present) are involved through the strength of their contribution to

$[\chi(T, 0) - \chi(0, 0)]$, i.e., the relation (12) of Ref. 12 which shows how much enhanced is the field variation of C/T , through the third power of the Stoner enhancement $1/(1 - \bar{I})$:

$$\begin{aligned} \left(\frac{C(T, H) - C(T, 0)}{T} \right)_{T \rightarrow 0} &= a \chi(0, 0) \frac{H^2}{T_{sf}^2} \\ &\propto \pm \frac{H^2}{(1 - \bar{I}) T_{sf}^2} \\ &\propto \pm \chi^3(0, 0) H^2 \end{aligned} \quad (3)$$

with

$$\chi(0, 0) = \frac{\chi_{\text{Pauli}}(0, 0)}{1 - \bar{I}} \propto \frac{1}{T_{sf}}, \quad (4)$$

where H and T are expressed in appropriate units, T_{sf} is the spin-fluctuation temperature or the characteristic energy of the strongly interacting system, and a is a band-structure-dependent coefficient, which may be positive or negative as emphasized in Refs. 11 and 12; $(1 - \bar{I})$ is the inverse Stoner enhancement expressed in terms of the strong dimensionless interaction $\bar{I} (\leq 1)$. Recall that in the pure Stoner theory which neglects paramagnon effects, (3) would involve $(1 - \bar{I})^{-2}$ which is a much weaker coefficient than $(1 - \bar{I})^{-3}$ of (3). In the particular case studied in Ref. 9 (parabolic band, spherical Fermi surface) (3) reduced to

$$\left(\frac{C(T, H) - C(T, 0)}{T} \right)_{T \rightarrow 0} = -\frac{3.2\pi^2}{24} \frac{\chi(0) H^2}{T_{sf}^2}$$

and thus,

$$\frac{C(H) - C(0)}{C(0)} = -0.1 \left(\frac{H}{T_{sf}} \right)^2 \frac{(1 - \bar{I})^{-1}}{\ln(1 - \bar{I})^{-1}}$$

When Ref. 9 was written, the paramagnon theory was recent.¹⁰ The sign of $[C(H) - C(0)]$ was not indicated in Ref. 9 as it was obvious from thermodynamics but, more importantly, because the magnitude of $\Delta C(H)/C(0)$, estimated in Ref. 9 for liquid ³He, was found to be too small to be observed, due to the smallness of the nuclear moment of the fermions (hidden in H in the present units). However, since then, data became available for electron systems, for which the moment is much larger; it was observed⁵ that in the nearly ferromagnetic compound UAl_2 , the low-temperature variation of $\chi(T)$ and the field dependence of $C(H)/T$ were in good agreement with the theoretical results derived in Ref. 9. Apart from the more recent data of Ref. 7, the only other available data, up to my knowledge, on the field variation of C/T for a strong Pauli paramagnet, are a

measurement⁴ made on CeAl_3 but at only one value of the field ($H = 10$ kOe), which appeared to be too small to induce an observable change in C/T . It is interesting to note that for LuCo_2 ,⁷ the value of C/T of Ref. 7 at 2.5 T, i.e., 25 kOe, is practically identical to the one at zero field, and higher fields (5.39, 7.62, and 9.98 T) were necessary to produce an observable shift of the C/T curves compared to those at $H = 0$ or 2.5 T.

We examine later in this paper the strength of the field effect on C/T compared to the one on M/H .

To summarize, simultaneous measurements of $[C(T, H)/T]_{T \rightarrow 0}$ and of $\chi(T, 0)$ on the same sample would be quite useful since these quantities ought to vary consistently if thermodynamics apply (which does not seem to be the case for the time being for LuCo_2 , if one compares Refs. 7 and 8). Secondly, band-structure calculations on the same sample, fixing the value and sign of a , are crucial to be compared with the experimental results to check again the consistency of the above calculation but more importantly to compute the strength of the effects.

II. FIELD DEPENDENCE OF THE MAGNETIZATION

The form $\chi^3(0, 0)H^2$ in formula (3) implies that H^2 scales like $\chi^{-3}(0, 0)$. This is indeed reasonable: It has been shown¹³ that, due to quantum effect at vanishing temperature, three-dimensional paramagnon theory actually behaves like a six-dimensional system and thus assumes mean-field critical exponents; therefore, at the transition, i.e., $T = 0$, $\bar{I} \sim 1$, H scales like M^8 with the magnetization M to a power equal to the mean-field critical exponent $\delta = 3$. Replacing M by χH , this indeed means that H^2 scales like χ^{-3} ($T = 0$), or in our present units, that H^2 is homogeneous to $[(1 - \bar{I}) T_{sf}^2]$ in agreement with (3). Such a scaling follows as well if one writes down a phenomenological Ginsburg-Landau free-energy equation $M^2/2\chi(T = 0) + (b/4)M^4 - MH$ as proposed in the first of Ref. 6; when this equation is minimized with respect to M , and then expanded to lowest order in H , it yields with $M(T = 0, H)/H \equiv \chi(0, H)$:

$$\chi(0, H) = \chi(0, 0) [1 + (\text{const}) \chi^3(0, 0) H^2] \quad (5)$$

The sign of the H^2 term is determined by the constant term in front. The microscopic derivation of the constant, within the paramagnon theory, is far from being obvious. In the pure Stoner theory (no paramagnons contribution) it can be extracted from Ref. 14 (devoted to metamagnetism) that (5) reads

$$\begin{aligned} \chi_{\text{Stoner}}(T = 0, H) \\ = \chi(0, 0) \left[1 + \frac{1}{6} \frac{H^2}{(1 - \bar{I})^3} \left(\frac{N''}{N} - 3 \frac{N'^2}{N^2} \right)_{E_F} \right] \end{aligned} \quad (6)$$

where N , N' , and N'' are the density of states and its first derivatives at the Fermi level. While in presence of fluctuations ("paramagnons"), the combination of N , N' , and N'' might appear differently from the one in (6), the validity of mean-field critical exponents recalled above (at $T=0$) allows one to expect that the enhancement $(1-\bar{I})^{-3}$ of H^2 will remain unchanged for vanishing temperatures. Therefore, there ought to be a critical field proportional to $(1-\bar{I})^{3/2}$, i.e., of order $(1-\bar{I})^{1/2}T_{sf}$ (in adequate units) such that at very low T for $T/T_{sf} \ll H/H_0 \ll 1$, $M(T \rightarrow 0, H)/H$ deviates quadratically from the zero-field constant. For higher temperatures, the variation of $M(T, H)/H$ cannot be predicted and may be very different. It requires a full account of both T and H in the computation of $M(T, H)$. The precise value of H_0 , i.e., the coefficient λ in $H_0 = \lambda(1-\bar{I})^{1/2}T_{sf}$, will remain unknown until paramagnons contributions can be incorporated in the calculation of $\chi(T=0, H)$ or, better, in $\chi(T, H)$.

On the other hand, H_0 defines the characteristic crossover field separating the low-field region where M varies first linearly with H , from the high field one where M saturates. The quantitative knowledge of H_0 would be highly useful also to study the phase diagram of the polarized liquid ^3He system¹⁵; H_0 would be determined by a nontrivial microscopic study, in presence of a finite field, of the Wilson-type Lagrangian^{13,16} describing three-dimensional interacting paramagnons. I do not want to elaborate here more on the theoretical difficulties which would arise; this would be outside the scope of the present paper which only aims to a simple discussion of the experiments.

The theoretically expected change in the behavior for $\chi(T, H)$ when $T/T_{sf} = H/H_0$ is not inconsistent with the existing data. Indeed, experimentally,⁶ at low T (below roughly 10 K), $M/H = \chi(H)$ increases first with H , passes through a maximum and then decreases continuously, for higher H ; the maximum is less and less pronounced when the temperature increases: the extrapolation of the data seem to indicate that at higher T (above ~ 10 K), M/H continuously decreases when H increases from zero field. We recall here too that in zero field, while $\chi(H=0)$ assumes that Stoner form $\propto (1-\bar{I})^{-1}$ at 0 K, the coefficient of the T^2 term at finite T is modified^{9,11}; in particular, as emphasized in Ref. 11, the power of the enhancement is different in the T^2 term; we rewrite here these expressions for convenience, for a further comparison with the experiments⁶ on TiBe_2 :

$$\chi_{\text{Stoner}}(T, H=0) = \chi(0, 0) \left[1 + \frac{\pi^2}{6} \frac{T^2}{1-\bar{I}} \left(\frac{N''}{N} - \frac{N'^2}{N^2} \right) \right] \quad (7)$$

and

$$\chi_{\text{paramagnons}}(T, H=0) = \chi(0, 0) \left[1 + \frac{3.2\pi^2}{4} \left(\frac{5}{12} \frac{N''}{N} - \frac{1}{4} \frac{N'^2}{N^2} \right) \frac{T^2}{(1-\bar{I})^2} \right] \quad (8)$$

I concentrate in the following to the $T < T_{sf}$, $H < H_0$, and $T/T_{sf} < H/H_0$ regime, for which it is proposed that χ varies qualitatively similarly to (6). The following discussion for $\chi(T=0, H)$ is similar to the one made in Ref. 11 for $\chi(T, H=0)$. Since $-(N''/N)^2 < 0$, when $N''/N < 0$, $\chi(T, H=0)$ will decrease when T increases, and similarly $\chi(T=0, H)$ will decrease when H increases; this was the case¹ for $\chi(T, H=0)$ in liquid ^3He [where $(N''/N) = -(4E_F^2)^{-1}$], and would be so for $\chi(T \rightarrow 0, H)$ in that system if sufficiently high fields could be achieved. In contrast, $N''/N > 0$ is a necessary (but not sufficient) condition for $\chi(T, 0)$ to increase with T and $\chi_{\text{Stoner}}(0, H)$ to increase with H . But it then remains to compare N''/N to the fraction of $-(N''/N)^2$ appropriate to either quantity. For TiBe_2 (Ref. 6) there has recently appeared¹⁷ a couple of band-structure calculations (in good agreement with each other), according to which $N'(E_F) \approx 0$ and $N''(E_F) > 0$. Under such conditions, whatever is the fraction of $-(N''/N)^2$ involved, it is practically negligible compared to N''/N and the necessary condition above becomes also sufficient and is fulfilled, in agreement with the experimental observations⁶ which show that $\chi(T, 0)$ increases with T and $(M/H)/(M/H)_{H=0}$ increases first with H and all the more that T is small. According to a rough estimate from the band-structure calculations of Ref. 17, the bandwidth is roughly equal to 2684 K, $(N''/N)^2 \sim (5107 \text{ K})^{-2}$ and $N''/N \sim (1306 \text{ K})^{-2}$; on the other hand, the enhancement $(1-\bar{I})^{-1}$ in TiBe_2 is ~ 61.4 , as obtained from the density of states at the Fermi level¹⁷ (118 states per Rydberg cell for two spin directions and with two TiBe_2 in a cell) and the measured low-temperature zero-field susceptibility^{6,18}; this gives $T_{sf} = 2684/61.4 \sim 44$ K which can be compared with the value of 20 K obtained by the extrapolation of the high-temperature part (the Curie-Weiss form) of the inverse zero-field susceptibility $(T+20)^{-1}$. Then from Ref. 11 and formula (8) above, one deduces that at, for instance, $T = 2$ K, $[\chi(T, 0) - \chi(0, 0)]/\chi(0, 0)$ is about 2.5%, to be compared to 0.5% experimentally (Acker *et al.* of Ref. 6); one has to note that the agreement is much better than in the absence of paramagnons since, in the pure Stoner theory [formula (7) above], this ratio would be only 0.02%—on the other hand, the Stoner estimate in (6) for $[\chi(0, H) - \chi(0, 0)]/\chi(0, 0)$ would yield, when $H = 20$ kOe a change of about 13% at $T = 0$ K, which, if paramagnons could be taken into account,

would be multiplied by a numerical factor, unknown here; 13% is thus just an order of magnitude; experimentally, this percentage is $\sim 8.6\%$ at $T = 1.2$ K (Acker *et al.* of Ref. 6) and between 4 and 7% (due to large error bars) at $T = 1.8$ K (Shaltiel *et al.* of Ref. 6). The fact that, experimentally, this ratio is maximum at the lowest temperature, and decreases when T increases, goes along the arguments developed above, according to which one expects a different regime when one switches from T/T_{sf} smaller to larger than $H/[(1-\bar{I})^{1/2}T_{sf}]$ (with $\lambda = 1$), i.e., from $T/44$ K $< H/42$ kOe to $T/44$ K $> H/42$ kOe. It is worth noticing that experimentally,⁶ M/H goes from being $> (M/H)_{H=0}$ to $< (M/H)_{H=0}$ at about 90 kOe, i.e., about twice the above Stoner ($\lambda = 1$) critical field ~ 40 to 50 kOe. It would be very interesting to have data on the field variation of the specific heat at low temperature on this system too.

III. COMPARED FIELD VARIATIONS OF C/T AND M/H

I wish to examine here whether the effect of a finite field is stronger on C/T or on M/H . One can define

$$\Delta \left(\frac{C}{T} \right) \equiv \left[\frac{C(T, H) - C(T, 0)}{T} \right]_{T=0} = \frac{H^2}{T^2} [\chi(T, 0) - \chi(0, 0)] , \quad (9)$$

$$\Delta \left(\frac{M}{H} \right) = \frac{M(0, H)}{H} - \left[\frac{M(0, H)}{H} \right]_{H=0} \equiv \chi(0, H) - \chi(0, 0) . \quad (10)$$

In the pure Stoner theory, (9) and (10) combined with (6) and (7) yield, for a parabolic band ($N \propto \sqrt{E}$),

$$\left[\frac{\Delta(C/T)}{\Delta(M/H)} \right] = \frac{\pi^2}{6} (1 - \bar{I})^2 . \quad (11)$$

To choose an example, for a Stoner enhancement of, say, $10 = (1 - \bar{I})^{-1}$, $\Delta(C/T)$ is about 5% of $\Delta(M/H)$; i.e., the field effect on C/T is much weaker than on M/H . If one takes paramagnons into account, (9) can be made explicit using (8), but (10) is, so far, unknown as explained above. However, if the scaling argument for $\chi(0, H)/\chi(0, 0)$ varying as $H^2/(1 - \bar{I})^3$ is correct, aside from the (band-structure-dependent) numerical factor multiplying $H^2/(1 - \bar{I})^3$ in $\chi(0, H)/\chi(0, 0)$, and $T^2/(1 - \bar{I})^2$ in $\chi(T, 0)/\chi(0, 0)$, there is still a factor $(1 - \bar{I})$ difference between $\Delta(C/T)$ and $\Delta(M/H)$. This might ex-

plain why, although M/H has been observed to vary¹⁹ with field on YCO_2 , the experimentalists of Ref. 7 who have searched for a field dependence on C/T of YCO_2 did not find any.²⁰

IV. FIELD DEPENDENCE OF THE ELECTRICAL RESISTIVITY

In zero field and at very low temperature, the electrical resistivity $\rho(T)$ due to scattering of the conduction electrons of the extended band on the spin fluctuations in the narrow band of strongly interaction electrons has been computed to be

$$\rho(T \rightarrow 0) \propto \frac{T^2}{T_{sf}^n} \propto \chi^n(0, 0) T^2 = \left(\frac{M}{H} \right)_{H=0}^n T^2 , \quad (12)$$

where $n = 2$ or 0.5 depending on the approximations.^{21,22} The magnetoresistance $\rho(H)$ has been computed for locally enhanced systems,²³ in nearly magnetic dilute alloys, where $\rho(T, H)$ was shown to be smaller than $\rho(T, H = 0)$ for a given sign of a particular combination of N , N' , and N'' (different from the ones involved here), but $\rho(T, H)$ would have been larger than $\rho(T, H = 0)$ for the other sign of that same combination.²⁴ In the present uniformly enhanced system, in presence of a field, the various spin-spin correlation functions $\chi^{\alpha\beta}$ (where $\alpha, \beta = \pm 1$ corresponding to Zeeman energies $\pm H$) will enter in the calculation of the resistivity. However, to continue with the phenomenology, it seems reasonable to expect the low-field variation of the resistivity to be given by formulas analogous to (12) with M/H replacing $(M/H)_{H=0}$. If that is so, to lowest order in H one expects

$$\frac{\Delta\rho}{T^2} = \left[\frac{\rho(T, H) - \rho(T, 0)}{T^2} \right]_{T=0} \propto H^2 , \quad (13)$$

$$\Delta\rho \gtrless 0 \text{ for } M/H \gtrless (M/H)_{H=0} . \quad (14)$$

The inequalities (14) seem satisfied on preliminary resistivity data obtained for $TiBe_2$ (Ref. 25) and for YCO_2 .²⁶

To conclude, the few experiments listed above seem to support the following theoretical expectations, to lowest order in H : $C(H)$, $\chi(H)$, and $\rho(H)$ should vary quadratically with H ; the sign of the variation is band-structure dependent, however $[C(T, H)/T]_{T=0}$ increases or decreases when H increases, as $\chi(T, 0)$ increases or decreases when T increases; $\Delta\rho$ is positive or negative depending upon whether M/H increases or decreases when H increases compared to $(M/H)_{H=0}$. The existence of maxima in $\chi(T, 0)/\chi(0, 0)$ vs T and in $(M/H)/(M/H)_{H=0}$ vs H , therefore depends on the band structure. Consequently, in contrast to the results of

Ref. 27, C/T and ρ are not necessarily depressed by an applied field. It also appears that field effects on the thermodynamic properties are all the more observable when the spin-fluctuation temperature of the system is low²⁸; also the effects are all the more pronounced when the temperature of the experiment is low (and of course much lower than the spin-fluctuation one). Finally, there exists a critical field separating the low-field regime including the lowest-field corrections discussed in this note from a high-field one. It is hoped that in the future, field dependence of the specific heat, magnetization, and electrical resistivity will be measured on the same samples, at low temperatures and for varying fields. This

should, in particular, provide information concerning whether TiBe_2 is only a strongly exchanged enhanced paramagnet or if it will develop metamagnetism in a high-field regime.²⁹

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