$TiBe_2$, a test material for spin-fluctuation theories

F. Acker and R. Huguenin
Institut de Physique Expérimentale, Université de Lausanne, 1015 Lausanne, Switzerland

M. Pelizzone
Départment de Physique de la Matière Condensée, Université de Genève, 1211 Genève, Switzerland

J. L. Smith
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 2 July 1981)

Precise magnetic susceptibility measurements for $TiBe_2$ indicate that $\chi(H,T=0)$ and $\chi(T,H=0)$ vary initially like H^2 and T^2, in formal agreement with the "paramagnon" model. Logarithmic terms $H^2 \ln H$ and $T \ln T$ are found to be inadequate. There is apparently a discontinuity in the low-temperature differential susceptibility at $H \simeq 55$ kOe, which may be indicative of a spin-density wave.

Little was known before 1978 about $TiBe_2$, a cubic Laves phase ($C15$) compound.[1,2] The prediction by Enz and Matthias[3] that this material might be a weak ferromagnet like $ZrZn_2$ started a rush at the problem. Specifically, Enz and Matthias suggested that the ferromagnetism of $ZrZn_2$ was due to a positive electron-phonon contribution to the Stoner factor. This controversial point[4] apparently set the tone for subsequent developments. The first low-temperature susceptibility measurements for $TiBe_2$ ruled out ferromagnetism and were interpreted as evidence for itinerant antiferromagnetism.[5] The achievement of $TiBe_{2-x}Cu_x$ ferromagnetic compounds[6] soon encouraged the advocates of exchange enhanced paramagnetism in $TiBe_2$.[7–9] Meanwhile, a peak in the specific heat[10] at 1.9 K was analyzed in terms of spin-density-wave antiferromagnetism (phasons).[11] Metamagnetism was also proposed[12] on the basis of the variation of the susceptibility with field.[7,8] A bibliography may be found in Ref. 13.

Clearly, no consensus has been reached yet, although the paramagnetic interpretation seems to gain support.[14] At this level the motivation of the present work was the controversy about the temperature and field dependence of the susceptibility, χ, for a Fermi liquid, which, instead of T^2 and H^2,[15] was claimed to be $T^2 \ln T$ and $H^2 \ln H$.[16] Earlier $\chi(T)$ and $\chi(H)$ measurements for TiBe have been fitted with the above logarithmic formulas[7,17] but these data are not precise enough for reliable conclusions to be drawn. A more definite answer is given by the present measurements, provided that the description of $TiBe_2$ in terms of enhanced paramagnetism is adequate. However, some features of the magnetization $M(H,T)$ are still not well understood.

The sample used was spherical, 5 mm in diameter.[7] Its magnetization M was measured to 0.1% with a moving sample magnetometer[18] down to $T=1.45$ K and up to $H=69$ kOe. Measurements to about 0.01%, of $\chi(T)$ in three constant fields (0.12, 0.5, and 5 kOe), between 1.68 and 20 K were performed in Geneva, using a newly built superconducting quantum interference device (SQUID) susceptometer.[19]

Figure 1 shows that below 46 kOe the magnetization of $TiBe_2$ at 1.45 K varies with the applied field according to the relation

$$\frac{H}{M} = \chi^{-1}(0) + BM^2 \ , \tag{1}$$

with $B<0$. There is possibly a small upturn of the susceptibility below 10 kOe. A fit of the data with Eq. (1) defines $\chi(0) = 9.70 \times 10^{-3}$ emu/mole and $B = -4.96 \times 10^{-5}$ emu/mole. Above 46 kOe the Arrott plot deviates from a straight line and H/M goes through a minimum, at $H_m(1.45\text{ K}) = 55$ kOe, as found previously.[7] The present measurements are precise enough to define a differential susceptibility $\Delta M/\Delta H$ as a function of field (Fig. 2). The calculated quantity $dM/dH = [\chi^{-1}(0) + 3BM^2]^{-1}$ [derived from Eq. (1), curve a] diverges at $H_c = 57.2$ kOe (vertical line) with the present values of the parameters. The lower the temperature, the higher is the field above which the data deviate from curve a and the sharper is the peak in $\Delta M/\Delta H$. Even by introducing higher-order terms CM^4 and DM^6 in Eq. (1) it is by no means possible to describe the measured $\Delta M/\Delta H$ below and above $H_m \simeq H_c$ with one set of parameters (see curves a and a*). Curve b was calculated with the formula $dM/dH = \chi(0) - cH^2(1 + 3\ln H/H^*)$ derived from Ref. 16. Obviously, the fit is very poor.

The variation of χ with temperature in a fixed field (0.5 kOe) is shown in Fig. 3. Such detailed measure-

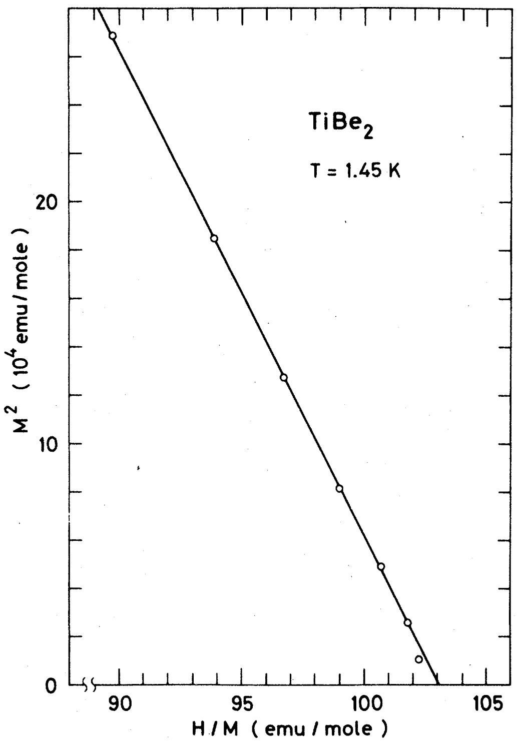

FIG. 1. Arrott plot (M^2 vs H/M) of the magnetization, in fields between 10 and 46 kOe, for $TiBe_2$ at 1.45 K.

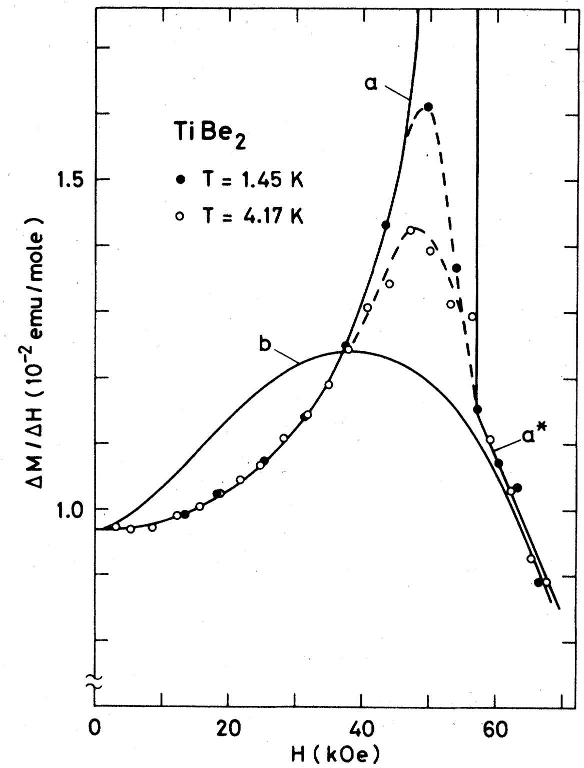

FIG. 2. Differential magnetic susceptibility $\Delta M/\Delta H$ as a function of field for $TiBe_2$ at 1.45 and 4.17 K. Solid curves are calculated: Curve a, from Eq. (1); curve a*, from Eq. (1) with a CM^4 additional term; curve b, from Misawa's logarithmic formulas (see text).

ments were repeated for $H = 0.12$ and 5.0 kOe. The data below 3.5 K were fitted with the expression $\chi(T) = \chi(0)(1 + \alpha T^2)$, yielding a strong variation of α with H (insert). For $H \rightarrow 0$ we find $\alpha \simeq 6.0 \times 10^{-4}$ K^{-2}. It is likely that for $H \simeq 25$ kOe χ will be practically constant, up to about 10 K. Further measurements are planned around H_m where χ increases rapidly with decreasing temperature.

If one takes for granted that the magnetic properties of $TiBe_2$ are those of a Fermi liquid, Figs. 1–3 indicate that the correct initial variations of the susceptibility with field ($T \rightarrow 0$) and temperature ($H \rightarrow 0$) for such a system are, respectively, H^2 and T^2, which differ markedly from $H^2 \ln H/H^*$ and $T^2 \ln T/T^*$. Obviously an apparent T^2 variation at low temperature may only be obtained by taking two or more $a_n T^n \ln T/T_n$ terms. This introduces at least four parameters and the fit is not unique. We wish to mention that a T^2 law possibly holds at low temperature for all the materials[15,16,20] for which χ has been tentatively described with a $T^2 \ln T/T^*$ law. The low-temperature data are rather scarce, but they deviate characteristically from the calculated curves. A small variation of χ with H has been reported for YCo_2 and $LuCo_2$.[21] Although the data for YCo_2 apparently follow a H^2 law, a fit with $H^2 \ln H/H^*$ has also been tried.[22]

Béal-Monod recently confronted the calculated low-temperature variation of χ in the paramagnon model and in the Stoner model with earlier data for $TiBe_2$.[7] The paramagnon formula,[15] which essentially differs from the Stoner result by a factor S (Stoner factor), was shown to be the most adequate.[14] It should be noticed, however, that the paramagnon prediction for the coefficient of T^2 in $\chi(T)$, the

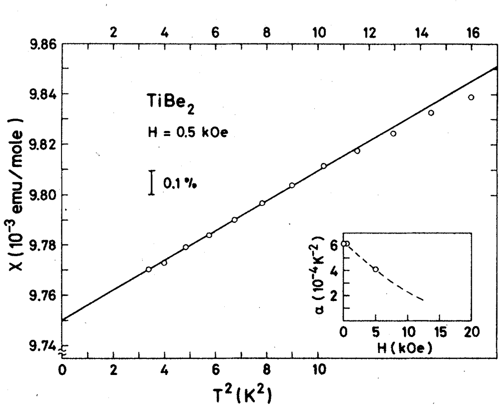

FIG. 3. Magnetic susceptibility, $\chi = M/H$, as a function of T^2 for $TiBe_2$ in a field of 0.5 kOe. Insert, variation with field of α in $\chi(T) = \chi(0)(1 + \alpha T^2)$.

present experimental value in low field and the Stoner value are approximately in the ratios 100:10:1. In Ref. 14, S was taken to be 61.4 and the values for the derivatives of the density of states at the Fermi energy were estimated from recent band-structure calculations.[23] The coefficient of H^2 in $\chi(H)$ cannot be easily obtained in the paramagnon model.[14]

Coming back to Figs. 1 and 2, we wish to point out some similarity between the low-temperature magnetization curve for $TiBe_2$ and for the cubic compound MnSi.[24] In both cases dM/dH has a singularity[25] at a critical field ($\simeq$1 kOe for a MnSi powder). A helical spin-density wave was detected in the itinerant electron magnet MnSi by low-angle neutron diffraction on a single crystal,[26] four years after the second unsuccessful investigation with neutrons (second of Ref. 24). Keeping in mind that spin-density wave antiferromagnetism was already proposed for $TiBe_2$,[11,27] further low-temperature low-angle neutron diffraction studies of this fascinating compound might prove rewarding. If the spin-density wave is longitudinal, however, its detection could be problematic.[27,28]

Finally, we want to mention that the small downturn in H/M for H decreasing below 10 kOe (Fig. 1) is in qualitative agreement with the fact that $d\chi/dH$ is very small in low fields while $d\chi/dT$ decreases rapidly with increasing field. If the Arrott plot at $T\rightarrow 0$ is to be perfectly straight (retrograde) there will be a shallow maximum in $H/M=\chi^{-1}$ versus field at finite (low) temperature. However, this effect ($\Delta\chi/\chi \simeq 0.1\%$ at 4 K) is about 10 times smaller than the observed one which may be due to the uncertainty in H or to a small impurity contribution.

In conclusion, it appears that the nature of the magnetization in $TiBe_2$ is still not fully elucidated. While the low-field, low-temperature susceptibility of this compound can be described by the paramagnon model, the presence of a spin-density wave may be inferred (in particular) from higher-field data. Based on a recent electronic structure calculation for Pd in megagauss fields,[29] the possible occurrence of itinerant metamagnetism in $TiBe_2$ also remains an open question.

ACKNOWLEDGMENTS

This research was supported by the Swiss National Science Foundation and by the U. S. Department of Energy.

[1] B. T. Matthias, T. H. Geballe, and V. B. Compton, Rev. Mod. Phys. 35, 1 (1963).
[2] H. Saji, T. Yamadaya, and M. Asanuma, J. Phys. Soc. Jpn. 21, 255 (1966).
[3] C. P. Enz and B. T. Matthias, Science 201, 828 (1978); see also Z. Phys. B 33, 129 (1979).
[4] D. Fay and J. Appel, Phys. Rev. B 20, 3705 (1979).
[5] B. T. Matthias, A. L. Giorgi, V. O. Struebing, and J. L. Smith, J. Phys. Lett. (Paris) 39, L-441 (1978).
[6] A. L. Giorgi, B. T. Matthias, G. R. Stewart, F. Acker, and J. L. Smith, Solid State Commun. 32, 455 (1979).
[7] F. Acker, Z. Fisk, J. L. Smith, and C. Y. Huang, J. Magn. Magn. Mater. 22, 250 (1981).
[8] P. Monod, I. Felner, G. Chouteau, and D. Shaltiel, J. Phys. Lett. (Paris) 41, L-511 (1980).
[9] D. Shaltiel, P. Monod, and I. Felner, J. Phys. Lett. (Paris) 41, L-567 (1980).
[10] G. R. Stewart, B. T. Matthias, A. L. Giorgi, E. G. Szklarz, and J. L. Smith, Solid State Commun. 30, 709 (1979).
[11] C. P. Enz and G. R. Stewart, Solid State Commun. 35, 951 (1980).
[12] E. P. Wohlfarth, J. Phys. Lett. (Paris) 41, L-563 (1980).
[13] E. P. Wohlfarth, Comments Solid State Phys. 10, 39 (1981).
[14] M. T. Béal-Monod, Phys. Rev. B (in press); Physica (Utrecht) B + C (in press).
[15] M. T. Béal-Monod, Shang-Keng Ma, and D. R. Fredkin, Phys. Rev. Lett. 20, 929 (1968); M. T. Béal-Monod and J. M. Lawrence, Phys. Rev. B 21, 5400 (1980), and references cited therein.
[16] S. Misawa, Phys. Lett. 32A, 153, 541 (1970); G. Barnea, J. Phys. C 8, L-216 (1975); G. Barnea and D. M. Edwards, J. Phys. F 7, 1323 (1977); S. Misawa, *ibid.* 8, L-263 (1978).
[17] S. Misawa (unpublished).
[18] F. Acker and R. Huguenin, J. Magn. Magn. Mater. 12, 58 (1979).
[19] M. Pelizzone and A. Treyvaud, Appl. Phys. 24, 375 (1981).
[20] S. Misawa, J. Phys. F 10, L-115 (1980); S. Misawa, Solid State Commun. 16, 1215 (1975); 15, 507 (1974); J. Beille, D. Bloch, and J. Voiron, J. Magn. Magn. Mater. 7, 271 (1978); A. Hahn and W. Treutmann, Z. Angew. Phys. 26, 129 (1969); see also R. J. Trainor, M. B. Brodsky, and H. V. Culbert, Phys. Rev. Lett. 34, 1019 (1975), for an example of decreasing χ with increasing T.
[21] D. Bloch, D. M. Edwards, M. Shimizu, and J. Voiron, J. Phys. F 5, 1217 (1975); C. J. Schinkel, *ibid.* 8, L-87 (1978).
[22] S. Misawa, J. Phys. F 8, L-263 (1978).
[23] R. A. de Groot, D. D. Koelling, and F. M. Mueller, J. Phys. F 10, L-235 (1980); T. Jarlborg and A. J. Freeman, Phys. Rev. B 22, 2332 (1980).
[24] H. J. Williams, J. H. Wernick, R. C. Sherwood, and G. K. Wertheim, J. Appl. Phys. 37, 1256 (1966); L. M. Levinson, G. H. Lander, and M. O. Steinitz, in *Magnetism and Magnetic Materials—1973,* edited by C. D. Graham, and J. J. Rhyne, AIP Conf. Proc. No. 18 (AIP, New York, 1974), p. 1138.
[25] Measurements of the ac susceptibility of $TiBe_2$ below 1 K are undertaken, in order to follow the probable development of the peak in $\Delta M/\Delta H$.
[26] Y. Ishikawa, K. Tajima, D. Bloch, and M. Roth, Solid State Commun. 19, 525 (1976).
[27] V. C. Rakhecha, G. P. Felcher, S. K. Sinha, J. L. Smith, and B. T. Matthias, Solid State Commun. 33, 495 (1980).
[28] A. Arrott, in *Magnetism,* edited by G. T. Rado and H. Suhl (Academic, New York, 1966), Vol. II B.
[29] T. Jarlborg and A. J. Freeman, Phys. Rev. B 23, 3577 (1981).