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Suppression of spin fluctuations in $TiBe_2$ by high magnetic fields

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Measurement of the low-temperature specific heat of a well-characterized 15.6-mg sample of $TiBe_2$ was performed in magnetic fields of 0, 6.5, 11.4, 14.2, and 17.0 T. The results indicate a striking depression of the spin-fluctuation-caused upturn with increasing field in the lower-temperature specific heat and very little change at higher temperatures where the spin fluctuations are less predominant. A field for full suppression of the spin fluctuations is extrapolated to be above about 25 T. The field at which the onset of spin-fluctuation depression occurs is 5.2 ± 0.3 T, suggesting that the previously observed anomalies in the susceptibility and differential susceptibility of $TiBe_2$ at 5.5 T are connected to the onset of the depression of spin fluctuations. Furthermore, this onset of spin-fluctuation depression at 5.2 ± 0.3 T coupled with the extrapolation to full suppression above 25 T serves to unify the interpretations of previous data on $TiBe_2$ by Wohlfarth, by Acker *et al.*, and by van Deursen *et al.* which were previously thought to be in contradiction.

I. INTRODUCTION

The magnetic properties of $TiBe_2$ have been a source of controversy since the claim by Matthias *et al.*[1] in 1978 that $TiBe_2$ is an itinerant antiferromagnet. Wohlfarth predicted[2] that $TiBe_2$ would undergo a first-order transition to a ferromagnetic state in a field later stated[3] to be only 5.8 T based on magnetization data of Monod *et al.*[4] This metamagnetism proposal of Wohlfarth has recently received at least partial support from theoretical band-structure considerations coupled with the de Haas—van Alphen data of van Deursen *et al.*[5] Acker and co-workers, however, have interpreted their[6,7] magnetization data in fields to 21 T and that of Monod *et al.* as evidence for exchange-enhanced paramagnetism or spin fluctuations in $TiBe_2$. This was seen[3] to be in direct disagreement with the work of Wohlfarth[2] and also with the claim of van Deursen *et al.*[5] of ferromagnetism in $TiBe_2$ in fields above 15 T.

Stewart *et al.*[8] have recently measured the specific heat of $TiBe_2$ at low temperatures in 0 and 7 T and found convincing evidence of spin fluctuations—the $T^3 \ln T/T_{SF}$ temperature dependence in the specific heat that is characteristic[9,10] of spin fluctuations. They also saw no evidence of a first-order transition at 5.8 T. However, the relatively low (7 T) applied field used was insufficient to observe a change in the low-temperature specific heat (LTSH) of more than 4% in $TiBe_2$. In order to trace the variation of the LTSH with field, predicted[11] to go as H^2, and to investigate the claim of ferromagnetism at 15 T by van Deursen *et al.*,[5] we have measured the LTSH of a sample of $TiBe_2$ in 0-, 6.5-, 11.4-, 14.2-, and 17.0-T fields.

II. EXPERIMENTAL

For a complete description of how the LTSH was measured in high fields, the reader is referred to Ref. 12, where Stewart, Cort, and Webb describe how they succeeded for the first time in measuring LTSH in a normal-state Bitter-type magnet in fields to 18 T. The apparatus used in the present work is identical, except that temperature in the present work was measured with an encapsulated carbon-glass thermometer. Its magnetoresistance was corrected for using published data.[13] This procedure was checked at the end of the measurements by admitting He exchange gas into the vacuum can, ramping the field up to 6.5, 11.4, 14.2, and 17.0 T, and measuring the magnetoresistance of the particular encapsulated carbon-glass thermometer used in this experiment. The maximum error in temperature in the present experiment caused by the observed difference between the median published value[13] for the magnetoresistance of carbon-glass thermometers and the actual value of magnetoresistance observed for the carbon-glass thermometer used was less than 0.1 K at the highest field.

In addition, another check on accuracy was performed by measuring the LTSH of a 212-mg sample of high-purity Ge in these same fields at several temperatures and comparing the results with published[14] results in zero field. This mass of Ge was chosen to give approximately the same LTSH at 10 K as the sample of $TiBe_2$ (15.6 mg). The results of measuring the high-purity Ge standard in 17.0 T at $T=8.34$, 11.07, and 13.62 K were Ge^{meas}/Ge (Ref. 14) $=0.989$, 1.003, and 0.999, respectively. Thus, the data presented here for $TiBe_2$ in applied magnetic fields may be viewed as accurate to better than $\pm 3\%$.

The sample used in the present work was the 15.6-mg sample labeled "sample 2" in previous work,[8] with a residual resistivity ratio, $R(300\ K)/R(4\ K)$, of 110. The previous work raised some question about minor impurities affecting the field behavior of the LTSH of sample 2. However, since sample 2 has a significantly larger LTSH response to a 7-T applied field, it was the choice for measurement. Later work will measure the high-field LTSH response of sample 1 of the previous work.

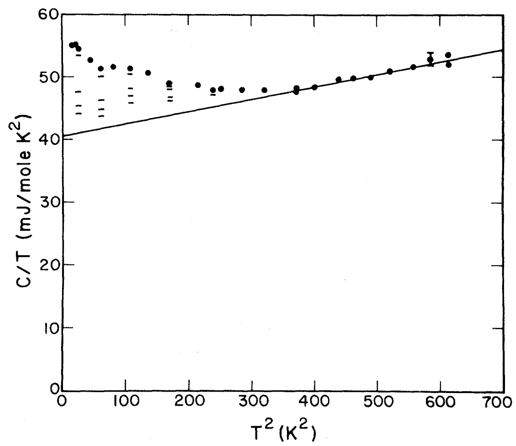

FIG. 1. Low-temperature specific heat of $TiBe_2$ in 0, 6.5, 11.4, 14.2 and 17.0 T. Zero-field data are the filled circles; field data monotonically decrease with increasing field and are represented by a series of horizontal lines. Only the 17-T-field point is shown at $T^2=238\ K^2$. See Table I for numerical values. Straight line shown is an extrapolation of the higher-temperature data where the spin fluctuations have died away. This extrapolation is used at lower temperatures to approximate where the depression of spin fluctuations would saturate. This percent decrease at saturation is used in Fig. 2.

III. RESULTS AND DISCUSSION

The LTSH data for $TiBe_2$ in 0, 6.5, 11.4, 14.2, and 17.0 T are shown in Fig. 1 and tabulated numerically at the five temperatures of field measurement in Table I. As may be seen in Fig. 1 and Table I, there is a substantial decrease of the LTSH in field at lower temperatures (19% in 17 T at 5.05 K) which becomes less pronounced as temperature is increased (2% in 17 T at 15.42 K).

The zero-field LTSH data in Fig. 1 may be fit to

$$\frac{C}{T}=\gamma_0(1+\lambda_{e\text{-}e})+\beta' T^2+\delta T^2 \ln T \ ,$$

where β' contains the lattice contribution as well as a spin-fluctuation contribution, $-\ln T_{SF}$, as is discussed in Ref. 8. The $T^3 \ln T/T_{SF}$ temperature dependence of the LTSH data in Fig. 1 and $\lambda_{e\text{-}e}>0$

TABLE I. C/T (mJ/mole K^2) data for $TiBe_2$ vs magnetic field. These data have a precision of approximately $\pm 1\%$. All these data were taken on the high-field sample platform described in Ref. 12. The agreement of the zero-field data taken on this platform in the present work with that taken on another platform in Ref. 8 is better than 2% except at $T=10.262$ K, where the data on the high-field platform is about 4.5% higher compared to that of Ref. 8. The computer fit to the zero-field data in Ref. 8 is in fact about 2% higher than the data at this temperature. Thus, the correct value of C/T at 10.262 K is probably midway between the number quoted here and the number obtained in Ref. 8.

	Field (T)				
T (K)	0	6.5	11.4	14.2	17.0
5.051	54.47	52.81	47.51	45.35	43.89
7.722	51.35	50.03	46.16	44.74	43.67
10.262	51.43	50.54	48.22	46.97	45.71
12.971	48.96	48.61	48.05	46.71	46.16
15.420	47.99				47.07

are associated[9,10] with the existence of spin fluctuations. It is this spin-fluctuation contribution which causes the upturn in the C/T-vs-T^2 plot of Fig. 1 at lower temperatures. The trend in the field data is clearly the depression of this spin-fluctuation contribution with increasing field. At higher temperatures, where this contribution is less predominant, i.e., where $T \rightarrow T_{SF}$, the amount of decrease in field is much reduced.

The question arises, when will the response of the LTSH to field saturate. The percent change in specific heat with applied field at the five temperatures is shown in Fig. 2. Clearly the change is much slower than H^2. The saturation field may be reasonably inferred to be that which decreases the LTSH to the value given by the extrapolation of the

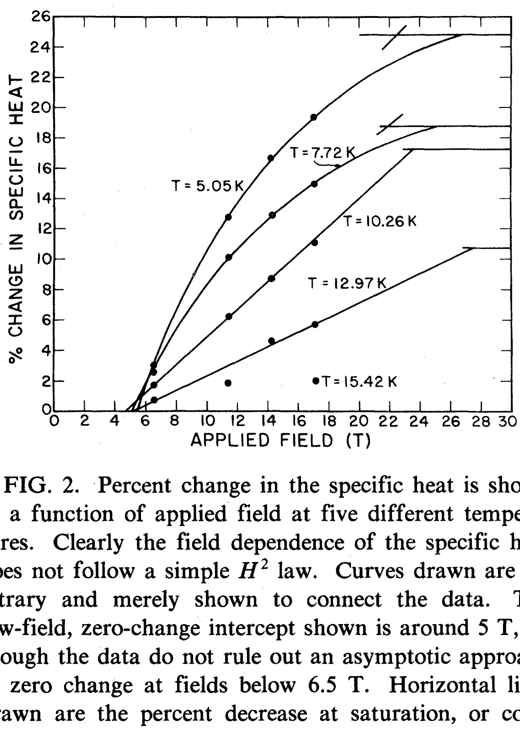

FIG. 2. Percent change in the specific heat is shown as a function of applied field at five different temperatures. Clearly the field dependence of the specific heat does not follow a simple H^2 law. Curves drawn are arbitrary and merely shown to connect the data. The low-field, zero-change intercept shown is around 5 T, although the data do not rule out an asymptotic approach to zero change at fields below 6.5 T. Horizontal lines drawn are the percent decrease at saturation, or complete spin-fluctuation suppression, at the four lower temperatures as discussed for Fig. 1. For $T=10.26$ and 12.97 K, a straight-line extrapolation fits the percent change of the data vs field. Intercepts of these straight lines give a predicted saturation field of 23.5 and 27.5 T, respectively. At the two lower temperatures, the percent change at the higher fields is clearly not linear. Thus, the correct saturation field is higher than that given by the straight-line extrapolation to the respective saturation levels shown and may be more correctly given by the arbitrary curves drawn to pass through the points. This procedure for determining the field at which the depression of spin fluctuations would saturate gives an approximate result of saturation at or above 25 T.

high-temperature data shown in Fig. 1. Although the extrapolations shown in Fig. 2 are somewhat approximate, it appears that the saturation field is above about 25 T. This is consistent with two estimates of the spin-fluctuation temperature T_{SF} for $TiBe_2$ made recently, one by Giorgi and Stewart[15] of 22 ± 4 K and one by Stewart *et al.*[8] of 50 ± 25 K. This is because, as pointed out by Brinkman and Engelsberg,[16] if a magnetic field of strength $H > k_B T_{SF}/g\mu_B$ (or $H > 0.74 T_{SF}$ for $g=2$, H in tesla and T_{SF} in kelvin) is applied to a spin-fluctuation material, the Zeeman splitting of the spin-up and spin-down band energies is large enough to quench the spin fluctuations.

Another feature of Fig. 2 is that there is no measurable LTSH response to a field below 5.2 ± 0.3 T. This field is where the peak in χ vs H occurs[6] in $TiBe_2$ and where a rather substantial peak in the differential magnetic susceptibility occurs in $TiBe_2$. Thus, it is tempting to attribute these previously observed anomalies at 5.5 T in the susceptibility to the onset of the suppression of spin fluctuations. Thus, above this field, the spin fluctuations are gradually depressed, and therefore their contribution[17] to the magnetic susceptibility starts to decrease, implying a monotonic decrease in the susceptibility above about 5.5 T—as is observed. Since this depression of the spin fluctuations in $TiBe_2$ is caused by the field splitting of the spin-up and spin-down bands, in energy, the state of the $TiBe_2$ above 5.5 T may in fact be called, by definition, "ferromagnetism." However, this transition into the spin-aligned or ferromagnetic state is quite gradual and is not complete until the bands split fully enough apart so that interband thermal promotion does not occur, at an applied field above approximately 25 T.

Can this explanation of the behavior of $TiBe_2$ also be applied to $TiBe_{2-x}Cu_x$? For $x > 0.16$, $TiBe_{2-x}Cu_x$ is ferromagnetic with, however, no saturation in fields to 21 T and no hysteresis or remnance in its magnetization behavior. Specific heat shows[8] the presence of spin fluctuations equally in $TiBe_{1.8}Cu_{0.2}$ as in $TiBe_2$ (the data for ferromagnetic $TiBe_{1.8}Cu_{0.2}$ overlaps within 2% of those shown for pure $TiBe_2$ below 22 K in zero field in Fig. 1). Thus, $TiBe_{2-x}Cu_x$ is not simply $TiBe_2$ displaced in behavior from $TiBe_2$ by an internal field of 10 T (the separation of the parallel χ-vs-H curves[6]) as suggested by Wohlfarth.[3] Otherwise, the spin fluctuations would be depressed in $TiBe_{1.8}Cu_{0.2}$ from pure $TiBe_2$. A consistent picture must therefore include at least two different bands in $TiBe_{2-x}Cu_x$, with one band responsible for the spin fluctuations of strength equal to that observed in $TiBe_2$ and another band fully split by the addition of Cu into spin-up and spin-down subbands, leading to the observed ferromagnetism. The suggestion that only a small fraction of the electrons at the Fermi energy take part in ferromagnetism in $TiBe_{1.8}Cu_{0.2}$ has already been proposed.[8]

Another means of plotting the field data is shown in Fig. 3, where the percent change in the LTSH is plotted at constant field versus temperature. Thus, Béal-Monod's prediction[18] that the field dependence of the LTSH of $TiBe_2$ would increase at lower temperatures (although not borne out at 7 T; see Fig. 3 and Ref. 8) is clearly correct at higher fields.

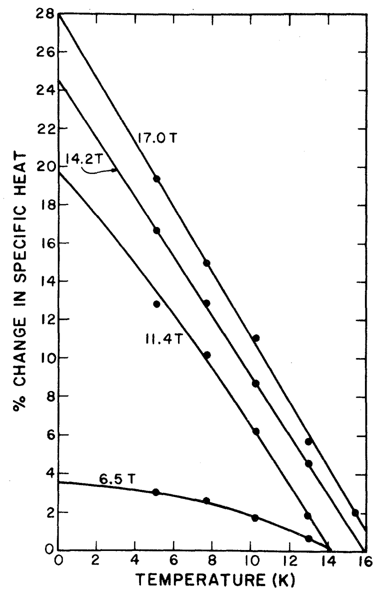

FIG. 3. Percent change in the specific heat at a given field is plotted vs temperature. These data show clearly that, at fields above 6.5 T, the specific heat responds more to field at the lower temperatures. This supports the assignment of this decrease as being due to the suppression of the spin-fluctuation term in the specific heat which becomes more important at lower temperature. Curved lines drawn for 6.5 and 11.4 T are arbitrary. It is likely that the percent change in the 14.2- and 17.0-T fields would curve over at lower temperatures below the straight extrapolations shown. If not, then the possible trend shown in Fig. 2 of the saturation field decreasing with decreasing temperature may in fact be true, since a 28% decrease at $T=0$ in the LTSH at 17 T corresponds quite closely to the extrapolated saturation C/T value at $T=0$ in Fig. 1.

IV. CONCLUSIONS

The LTSH of $TiBe_2$ in fields to 17 T shows clearly the partial suppression of spin fluctuations. The lack of any significant decrease of the LTSH in 17 T from the zero-field values at higher temperatures calls into question the interpretation of previous LTSH work[19] on Pd in fields to 11 T, where a decrease in field of the LTSH independent of temperature (0.2 to 11 K) is assigned to spin-fluctuation effects.

The depression of spin fluctuations in $TiBe_2$ should be complete above about 25 T, consistent with earlier estimates of the spin-fluctuation temperature of 22 ± 4 K (Ref. 15) and 50 ± 25 K.[8] This observation of depression of the spin fluctuations, caused[16] by the splitting in energy of a spin-up and spin-down band by an applied field, serves to unify the different views and experiments of Wohlfarth,[2,3] Acker *et al.*,[6,7] Monod *et al.*,[4] and van Deursen *et al.*[5] on $TiBe_2$ which were previously thought to be in contradiction.[3,7] Thus, the suggestion by Wohlfarth[2,3] of a ferromagnetic state in $TiBe_2$ for $H>5.5$ T is correct, with the clarification that (1) this ferromagnetic state is not possessed of the properties of the usual ferromagnet, and (2) the transition to this spin-aligned state is not complete until well beyond 21 T where data have been obtained. The observation of band splitting (2.6 mRy in energy) in the de Haas—van Alphen data of van Deursen *et al.*[5] above 15 T, which was interpreted[5] as showing that $TiBe_2$ is in a ferromagnetic state in fields above 15 T, is also consistent with our results. Finally, the claim[6] by Acker *et al.* that paramagnons or spin fluctuations persist in $TiBe_2$ up to 21 T is again consistent with our results, and with the data and interpretations of Wohlfarth and of van Deursen *et al.*, because our work has shown conclusively that both spin fluctuations and a spin-aligned (ferromagnetic) state exist in $TiBe_2$ between 5.5 and 25 T applied field.

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[1]B. T. Matthias, A. L. Giorgi, V. O. Struebing, and J. L. Smith, J. Phys. (Paris) Lett. 39, L441 (1978).
[2]E. P. Wohlfarth, J. Magn. Magn. Mater. 20, 77 (1980).
[3]E. P. Wohlfarth, Comments on Solid State Phys. 10, 39 (1981).
[4]P. Monod, I. Felner, G. Chouteau, and D. Shaltiel, J. Phys. (Paris) Lett. 41, L511 (1980).
[5]A. P. J. van Deursen, J. M. van Ruitenbeek, W. A. Verhoef, A. R. de Vroomen, J. L. Smith, R. A. de Groot, D. D. Koelling, and F. M. Mueller, Physica (Utrecht) (in press).
[6]F. Acker, Z. Fisk, J. L. Smith, and C. Y. Huang, J. Magn. Magn. Mater. 22, 250 (1981).
[7]F. Acker, R. Huguenin, M. Pelizzone, and J. L. Smith, Phys. Rev. B 24, 5404 (1981).
[8]G. R. Stewart, J. L. Smith, A. L. Giorgi, and Z. Fisk, Phys. Rev. B 25, 5907 (1982).
[9]S. Doniach and S. Engelsberg, Phys. Rev. Lett. 17, 750 (1966).
[10]N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett. 17, 433 (1966).
[11]M. T. Béal-Monod, Phys. Rev. B 24, 261 (1981).
[12]G. R. Stewart, B. Cort, and G. W. Webb, Phys. Rev. B 24, 3841 (1981).
[13]H. H. Sample, B. L. Brandt, and L. G. Rubin, Rev. Sci. Instrum. (in press).
[14]P. Flubacher, A. J. Leadbetter, and J. A. Morrison, Philos. Mag. 4, 273 (1959).
[15]A. L. Giorgi and G. R. Stewart (unpublished).
[16]W. F. Brinkman and S. Engelsberg, Phys. Rev. 169, 417 (1968).
[17]M. T. Béal-Monod, J. Phys. (Paris) 41, 1109 (1980).
[18]M. T. Béal-Monod, private communication.
[19]T. Y. Hsiang, J. W. Reister, H. Weinstock, G. W. Crabtree, and J. J. Vuillemin, Phys. Rev. Lett. 47, 523 (1981).