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Temperature and field dependence of the magnetic susceptibility of $TiBe_2$ under high pressure

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The magnetic susceptibility $\chi(T,P)$ of $TiBe_2$ is determined at a field strength of 2.1 T as a function of both temperature from 3 to 300 K and hydrostatic pressure up to 1.3 GPa (13 kbar). At low temperatures, χ is found to decrease under pressure, the degree of depression diminishing rapidly with increasing temperature from $\partial \ln\chi/\partial P = (-9.6\pm0.4)\%$/GPa at 3 K to $(0\pm0.3)\%$/GPa at 300 K. Pressure is also found to suppress strongly the low-temperature anomaly in the field dependence of $\chi(H)$ at 5.6 T. An increase in the spin-fluctuation temperature with pressure is indicated, $\partial T_{SF}/\partial P \simeq +3.5$ K/GPa, where $T_{SF}(0) \simeq 24$ K, although it is not possible to account for the present results in terms of the variation of a single characteristic energy. The Stoner factor appears to decrease rapidly under pressure. The pressure dependence of the lattice parameter for $TiBe_2$ at room temperature is determined up to 27 GPa, yielding the initial compressibility $K = (7.6\pm0.4)\times10^{-3}$ GPa^{-1}. It is shown that the ferromagnetism of $TiBe_{2-x}Cu_x$ for $x \geq 0.15$ cannot be due solely to the volume expansion associated with increasing x. Preliminary measurements indicate that substituting Mn for Ti in $TiBe_2$ causes a strong *decrease* in the total magnetization in analogy with previous studies involving Fe substitution. A simple qualitative model of the electronic structure of $TiBe_2$ near E_F is suggested. $TiBe_2$ is not superconducting at temperatures above 150 μK.

I. INTRODUCTION

It is now generally recognized that the $C15$-structure compound $TiBe_2$ is a particularly interesting example of strongly exchange-enhanced systems such as Pd, $CeSn_3$, YCo_2, and UAl_2.[1] The low-field magnetic susceptibility $\chi(T)$ of $TiBe_2$ rises rapidly with decreasing temperature, passing through a maximum at approximately 8 K.[2,3] That $TiBe_2$ is almost capable of developing spontaneous magnetism at low temperatures is indicated by its large Stoner enhancement factor $S \simeq 65$ (Ref. 1) and low spin-fluctuation temperature $T_{SF} \simeq 22$ K.[4] Indeed, even the addition of only a few percent of (nonmagnetic) Cu to $TiBe_2$ suffices to make it ferromagnetic[3,5]; on the other hand, the addition of Fe is found to suppress the enhanced paramagnetism[3,6] instead of provoking the buildup of giant moments as in Pd. In addition, a curious maximum in the dependence of $\chi(H)$ on magnetic field at ~ 5 T has been noted at low temperatures,[7–9] leading to speculations of metamagnetism,[10] Fermi-liquid behavior,[7] and/or the onset of the quenching of spin fluctuations.[4] Unfortunately, the experimental results for $TiBe_2$ depend to some extent on the details of the sample preparation,[1–9] particularly at low temperatures; investigations carried out on a single sample are thus of particular importance.

In this paper the influence of high hydrostatic pressure on the magnetism of two polycrystalline $TiBe_2$ samples is determined. A relatively large pressure dependence of χ is found for $T < 20$ K, which would appear to be correlated with the low-temperature $\chi(T)$ and $\chi(H)$ anomalies. A preliminary discussion of some of the present results was given earlier.[11]

Wu *et al.*[12] found the Curie temperature of $TiBe_{2-x}Cu_x$ compounds to decrease linearly with pressure. Although this decrease in T_c is consistent with the observed decrease in the Stoner factor S of $TiBe_2$ with pressure, we show that the appearance of ferromagnetism in $TiBe_{2-x}Cu_x$ for $x \geq 0.15$ cannot be solely accounted for by the observed volume expansion as Cu is substituted for Be. Our results are compared to those derived from the recent magnetostriction experiments of Creuzot and Campbell,[13] some of which were carried out on the same sample. We also determine the pressure-volume relation $V(P)$ at room temperature and discuss preliminary experiments on a $Ti_{1-x}Mn_xBe_2$ compound.

II. EXPERIMENT

The polycrystalline $TiBe_2$ samples used in the present work were taken from the same ingot as sample 2 of Ref. 14 or sample B of Ref. 15 and spark cut into cylinders of 4 mm diameter and 4 mm in length for insertion into the

high-pressure clamp. The residual resistance ratio was approximately 110. See Ref. 14 for details of the sample preparation techniques.

The pressure dependence of the magnetic susceptibility of $TiBe_2$ was studied using a Faraday magnetometer located in Bochum employing a 90-g hydrostatic pressure clamp machined from pure binary Cu-Be alloy. The $\chi(T)$ data were taken using a 2.1 T main field and 0.024 T/cm gradient field, whereas the $\chi(H)$ run at 3 K was carried out by varying the main field from 0 to 6 T in a fixed gradient field of 0.040 T/cm. See earlier publications[16,17] for further details of the experimental technique.

The $V(P)$ studies at room temperature were carried out in Paderborn, using a gasketed diamond-anvil cell[18] in the energy dispersive mode and averaging over the (220), (311), (400), and (331) diffraction lines. A 4:1 mixture of methanol-ethanol was chosen as the hydrostatic pressure medium, and the ruby R_1 fluorescence method was used for the pressure determination.[19]

III. RESULTS

In Fig. 1 we compare directly the temperature dependence of the molar magnetic susceptibilities $\chi(T)$ of Pd,[17] $CeSn_3$,[16] and $TiBe_2$. The low-temperature susceptibility of $TiBe_2$ is seen to soar well above those of the other two systems, indicating a lower value of the spin-fluctuation temperature and a possible larger exchange-enhancement factor $S=\chi(0)/\chi_{\text{Pauli}}$. We obtain the values $S=9.5$, 5.6, and 69 for Pd, $CeSn_3$, and $TiBe_2$, respectively, using the values of the Pauli susceptibility 0.078×10^{-3} emu/mol for Pd,[17] 0.34×10^{-3} emu/mol for the reference compound $LaSn_3$,[16] and 0.14×10^{-3} emu/mol for $TiBe_2$ (Ref. 1) together with the following values of the impurity-corrected susceptibility $\chi(0)$ at $T=0$ K: 0.74×10^{-3} emu/mol (Ref. 17), 1.9×10^{-3} emu/mol (Ref. 16), and 9.7×10^{-3} emu/mol (Fig. 1) for Pd, $CeSn_3$, and $TiBe_2$, respectively. The temperature dependence of the magnetic susceptibility of $TiBe_2$ in Fig. 1 agrees well with previous studies,[2,3,14] although the value at low temperature is characteristically sample dependent; our value $\chi(0)=(9.7\pm0.02)\times10^{-3}$ emu/mol at 2.1 T for two neighboring samples taken from the top-center section of the arc-melted bead compares with 9.5×10^{-3} and 8×10^{-3} emu/mol from Ref. 14 at 1.5 T and

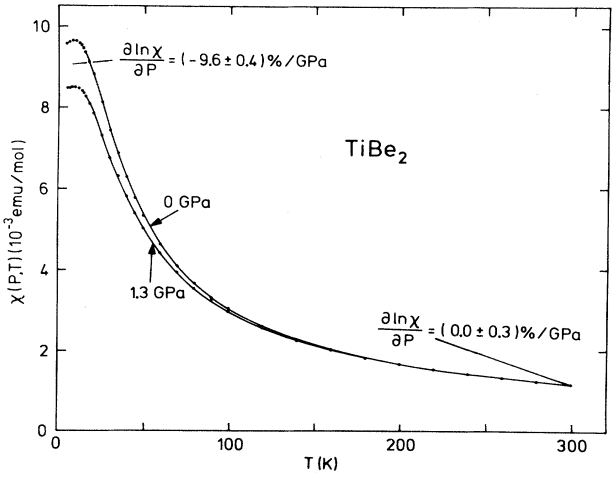

FIG. 2. Temperature dependence of the magnetic susceptibility of $TiBe_2$ at 2.1 T for 0 and 1.3 GPa pressure. $\partial\chi/\partial P$ is strongly temperature dependent.

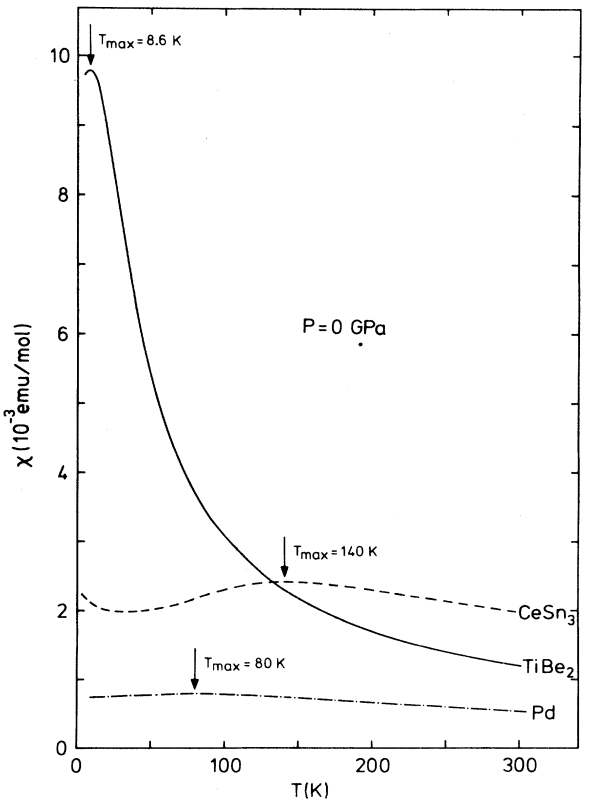

FIG. 1. Temperature dependence of the magnetic susceptibility at ambient pressure of $TiBe_2$, $CeSn_3$ (Ref. 16), and Pd (Ref. 17) at fields of $H=2.1$, 5.6, and 5.6 T, respectively.

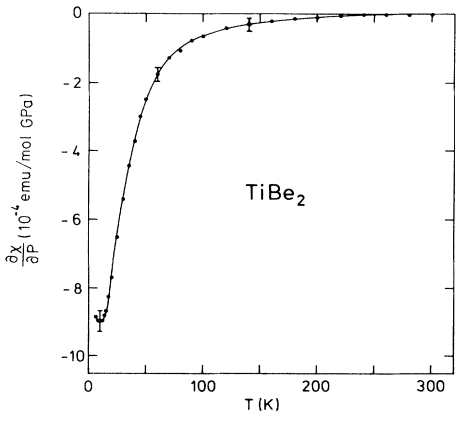

FIG. 3. Pressure derivative of the magnetic susceptibility of $TiBe_2$ vs temperature. Experimental accuracy is not sufficient to determine if the apparent minimum in $\partial\chi/\partial P$ at ~10 K actually exists.

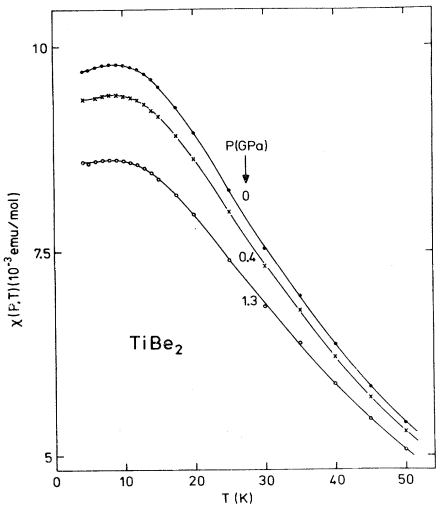

FIG. 4. Temperature dependence of the susceptibility at several pressures on an expanded scale.

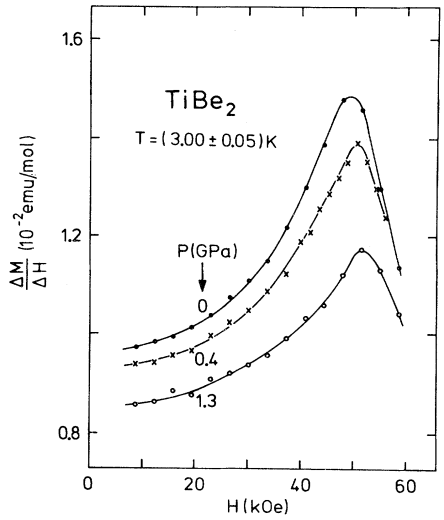

FIG. 6. Differential magnetic susceptibility $\chi(H)=\Delta M/\Delta H$ at 3 K vs magnetic field for three pressures.

9.75×10^{-3} emu/mol from Ref. 9 at 0.05 T. The former (higher) value from Ref. 14 was obtained for a sample taken from the equator of the same bead as our samples. Whereas we find a well-defined maximum in $\chi(T)$ at 8.6 K, Stewart *et al.*[14] do not, even though the susceptibility maximum should be slightly more pronounced in their case due to the lower (1.5 T) field value they used.[9] The above results evidently reflect the variation of the magnetic properties of this system within a large arc-melted bead.

In Fig. 2 we show the susceptibility of $TiBe_2$ over the temperature range 3—300 K at pressures of 0 and 1.3 GPa. At low temperatures the application of pressure is seen to decrease the susceptibility, the magnitude of this depression decreasing rapidly with increasing temperature, as seen in Fig. 3. At the lower temperatures the relative pressure dependence reaches the sizable value $\partial\ln\chi/\partial P=(-9.6\pm0.4)\%/\text{GPa}$, which compares with the values -15%/GPa for $CeSn_3$ (Ref. 16) and -1.6%/GPa for Pd.[17] In a separate experiment the pressure was varied at a fixed temperature of 300 K; within the experimental accuracy of $\pm0.05\times10^{-4}$ emu/mol GPa, which corresponds roughly to the width of the data points in Fig. 3, no change whatsoever in $\chi(300\text{ K})$ could be detected to 1.5 GPa. The data at lower temperatures are less accurate (see error bars). A less dramatic decrease in the magnitude of the pressure derivative $|\partial\chi/\partial P|$ as temperature increases is also observed for Pd (Ref. 17) and $CeSn_3$.[16]

In Fig. 4 we display the low-temperature data on an expanded scale. Within experimental accuracy, no pressure shift of the susceptibility maximum at $T_m\simeq8.6$ K can be resolved, although the maximum appears to broaden with pressure. A similar result was obtained for Pd,[17] whereas for $CeSn_3$ the susceptibility maximum was observed to shift to higher temperatures at the rate $\partial T_m/\partial P\simeq+34$ K/GPa.[16]

As seen in Fig. 5, the field dependence of the magnetization $M(H)$ at 3.0 K shows the $TiBe_2$ characteristic upturn.[7–9] The data are of sufficient accuracy to determine, even under high-pressure conditions, the differential susceptibility $\chi(H)=\Delta M/\Delta H$ displayed in Fig. 6. The ambient pressure curve is in remarkably good agreement with previous studies at 1.45 and 4.17 K by Acker *et al.*[9] In Fig. 6 the application of pressure is seen to suppress the susceptibility $\chi(H)$ at all fields to 5.6 T and shift the

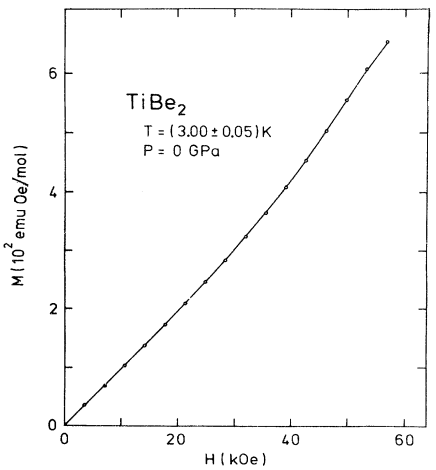

FIG. 5. Magnetization at 3 K vs magnetic field to 5.6 T at ambient pressure.

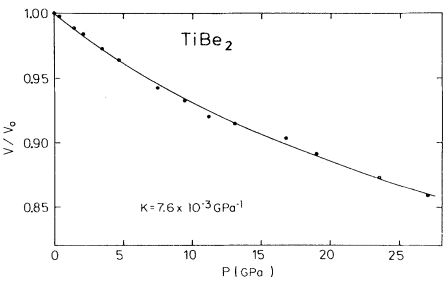

FIG. 7. Pressure dependence of relative sample volume of $TiBe_2$ at room temperature.

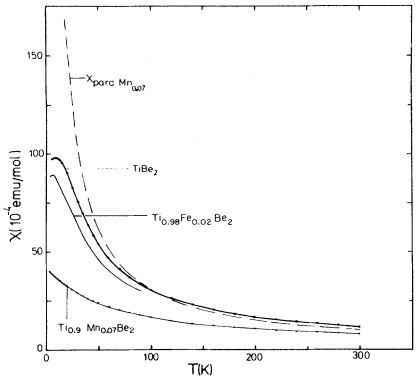

FIG. 8. Temperature dependence of magnetic susceptibility for $TiBe_2$, $Ti_{0.9}Mn_{0.07}Be_2$ (off stoichiometry), and $Ti_{0.98}Fe_{0.02}Be_2$ (Ref. 3). Calculated Curie-law susceptibility for 7 at.% Mn with $S=\frac{5}{2}$ is also shown for comparison.

peaked anomaly at 5 T to somewhat higher field values. It should be noted that the derivative $|\partial\chi(H)/\partial P|$ reaches its maximum value for $H=H_m\simeq 5$ T, decreasing particularly rapidly for higher fields $H>H_m$. This is in agreement with the magnetostriction results of Creuzet and Campbell,[13] which indicate that $\partial\chi/\partial P$ passes through zero near 6 T and changes sign; their estimate of the value of $\partial\chi/\partial P$ is also in reasonable agreement with our low-temperature value.

The pressure dependence of the relative sample volume $V(P)/V(0)$ of $TiBe_2$ to 27 GPa is shown in Fig. 7. From the initial pressure dependence, the compressibility value $K=-V_0^{-1}\partial V/\partial P=(7.6\pm 0.4)\times 10^{-3}$ GPa^{-1} can be extracted.

The results of a preliminary study of the influence of the substitution of Ti with Mn in $TiBe_2$ are shown in Fig. 8. The substitution of a few percent of Mn is seen to *decrease* the total susceptibility at all temperatures in analogy with previous studies on Fe-substituted compounds.[3,6] For comparison, also shown in Fig. 8 is the calculated paramagnetic susceptibility (Curie law) for the same volume concentration of free Mn spins as in the compound. The substituted Mn ions are not only unable to develop any local-moment free-spin paramagnetism of their own, but they, in fact, actually suppress the enhanced susceptibility of their $TiBe_2$ matrix.

IV. DISCUSSION

Perhaps one of the more interesting results of the present studies is the strong temperature dependence of the pressure derivative $\partial\ln\chi/\partial P$ seen in Figs. 2 and 3. At high temperatures $TiBe_2$ appears magnetically "hard," such as Fe or Co, with a magnetization which is initially relatively insensitive to volume change; on the other hand, at lower temperatures the stability of the magnetism appears more precarious, like in the weak itinerant ferromagnets[20] or valence-fluctuating compounds, where modest pressures can cause large changes in the magnetization.[11] This separation of the high-pressure behavior into two temperature regimes suggests the existence of a characteristic temperature, a point we will return to below. In any case, a reduction in the magnitude of the value of $\partial\ln\chi/\partial P$ with increasing temperature would appear to be a property of a large number of compounds whose magnetism either increases (e.g., Ce compounds) or decreases (e.g., Yb compounds) under pressure.[11]

A. Low-temperature susceptibility

The appreciable size of the enhancement factor $S=69$ for $TiBe_2$ would imply that paramagnon effects should dominate at sufficiently low temperatures, giving the expressions[1] for the initial functional dependence of the susceptibility on temperature

$$\chi_{\rm para}(T)=S\chi_{\rm Pauli}\left[1+\frac{\pi^2}{6}\left[2\frac{N''(E_F)}{N(E_F)}-1.2\frac{[N'(E_F)]^2}{[N(E_F)]^2}\right]S^2T^2\right] \quad (1)$$

and field

$$\chi_{\rm para}(H)=S\chi_{\rm Pauli}[1+f(N(E_F),N'(E_F),N''(E_F))S^3H^2]\ . \quad (2)$$

Here f is a function of the density of states N and its first and second derivatives, $N'=\partial N/\partial E$ and $N''=\partial^2 N/\partial E^2$; f is believed to be similar in form and magnitude to the corresponding function in parentheses in Eq. (1).[1] It would be expected from Eqs. (1) and (2) that $\chi(T,H)$ should initially *increase* quadratically with T and H only if N'' is sufficiently large and positive; such behavior is indicated for $TiBe_2$ by Acker *et al.*[9] and by our own data in Fig. 5 for $\chi(H)$. The present experiments did not go sufficiently low in temperature to check for a $\chi(T)\sim T^2$ law, although a positive curvature in $\chi(T)$ for $T<5$ K is indicated.

From the Maxwell relation $\partial M/\partial T=\partial\mathcal{S}/\partial H$, where $\mathcal{S}$ is the entropy, it follows that

$$H\left[\frac{\partial^2\chi}{\partial T^2}\right]_H=\left[\frac{\partial\gamma(H)}{\partial H}\right]_T, \quad (3)$$

where γ is the linear coefficient of the electronic specific heat. When using Eq. (3) care must be taken to evaluate χ and γ at the *same* temperature T, pressure P, and field H. A positive curvature in $\chi(T)$ thus implies that γ should increase with H. However, Acker *et al.*[9] have shown that $\chi(T)$ for $TiBe_2$ may continuously change from positive to negative curvature at $T<4$ K as the field H is increased from zero to a value somewhat above 2 T. The recent specific-heat results of Stewart *et al.*,[4] who find that γ *decreases* with field for $H>5$ T, with $\partial\gamma/\partial H\simeq 0$ at lower fields, are thus not inconsistent with the above susceptibility results. Recent susceptibility studies by Acker *et al.*[15] on $TiBe_2$ have shown convincingly that Maxwell's relation in Eq. (3) is indeed obeyed.

There is a second Maxwell relation connecting the change of the magnetic susceptibility with pressure to the volume change with magnetic field:

$$\left[\frac{\partial\chi}{\partial P}\right]_H = \frac{-1}{V_0 H}\left[\frac{\partial V}{\partial H}\right]_P . \tag{4}$$

Recent magnetostriction studies on our $TiBe_2$ sample by Creuzet and Campbell[13] to 20 K and 7 T agree within experimental error with our direct pressure measurements, thus confirming the validity of Eq. (4). In light of the above, it seems likely that apparent violations of the Maxwell relations will disappear if these relations are properly applied to measurements on a single sample.

B. Pressure dependence of susceptibility

From Eq. (1) we have at $T=0$ K

$$\chi(0)=S\chi_{\text{Pauli}}=2\mu_B^2\frac{N(E_F)}{1-\bar{I}N(E_F)} , \tag{5}$$

where μ_B is the Bohr magneton and $\bar{I}$ the screened Coulomb-Coulomb interaction. From Eq. (5) it follows that

$$\frac{\partial\ln\chi(0)}{\partial\ln V}=-K^{-1}\frac{\partial\ln\chi(0)}{\partial P}=\gamma_e+\frac{\partial\ln S}{\partial\ln V} \tag{6}$$

and

$$\frac{\partial\ln\bar{I}}{\partial\ln V}=\frac{1}{S-1}\frac{\partial\ln S}{\partial\ln V}-\gamma_e , \tag{7}$$

where $\gamma_e=[\partial\ln N(E)/\partial\ln V]_{E=E_F}$. In the absence of a calculated or measured value of γ_e for $TiBe_2$, we assume $\gamma_e\simeq+1.1$ as estimated for two other related systems $ZrZn_2$ (Ref. 21) and Pd.[22] Such a value lies relatively close to the value $\gamma_e=+\frac{5}{3}$ expected for the simple pressure broadening of d bands.[23] Inserting our experimental values into Eq. (6) we find that $\partial\ln\chi(0)/\partial\ln V=+12.6\pm1.2$ so that $\partial\ln S/\partial\ln V\approx+12.6-1.1=+11.5$ for $TiBe_2$ compared with, for example, the small value $+0.6$ for Pd.[17] In $TiBe_2$, therefore, the exchange-enhancement factor would appear to decrease quite rapidly with pressure.

From Eq. (7) we derive the volume dependence of the exchange interaction $\partial\ln\bar{I}/\partial\ln V=(68)^{-1}(+11.5)-1.1=-0.93$, which is opposite in sign but almost equal in magnitude to the above value $\gamma_e=+1.1$. Such a near compensation of the *pressure increase* of the exchange interaction by a *pressure decrease* of the density of states was also found for Pd (Ref. 17) and other compounds.[11] In contrast to Pd, however, for $TiBe_2$ both $S\equiv[1-\bar{I}N(E_F)]^{-1}$ and $\chi(0)$ decrease rapidly under pressure due to the very large value of the enhancement factor $S=69$ in the later case.

C. Pressure and field dependence of susceptibility

In Fig. 9 we plot the low-temperature magnetization data $M(H)$ for $TiBe_2$ at several pressures in the form of an Arrott plot. Also shown are ambient pressure results on $Ti_{1-x}Fe_xBe_2$ and $TiBe_{2-x}Cu_x$ compounds. Whereas substituting Cu for Be initially moves $TiBe_2$ towards ferromagnetism, applying pressure moves it away, as implied by the decrease of the Stoner parameter $\bar{I}N(E_F)$ with pressure (from the above it follows that $[\partial\ln(\bar{I}N)/\partial\ln V]_{E=E_F}\approx+0.17$). Since, for $S=69$, $\bar{I}N(E_F)\simeq0.986$, it follows that a 1.4% increase in $\bar{I}N(E_F)$ should drive $TiBe_2$ ferromagnetic [i.e., $\bar{I}N(E_F)=1$]. From our present results we would thus predict that this would require a volume *increase* of $\Delta V/V_0\simeq1.4\%/0.17\simeq8\%$ or a negative pressure of approximately 11 GPa. Since the critical concentration for ferromagnetism in $TiBe_{2-x}Cu_x$ occurs at $x=0.15$ where the lattice has only expanded in volume by 1.8%, it is clear that ferromagnetism in $TiBe_{1.85}Cu_{0.15}$ cannot be a simple volume expansion effect, but must arise predominantly from the changes in electron density and impurity scattering as Cu is substituted. A similar conclusion was reached by Creuzet and Campbell[13] and Giorgi and Stewart.[24]

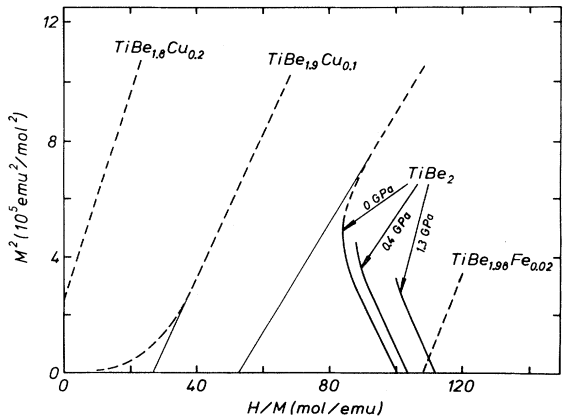

FIG. 9. Arrott plot from the field dependence of the magnetization of $TiBe_2$ at 3 K for 0, 0.4, and 1.3 GPa. Rest of data is from Ref. 6 where Fe was assumed to substitute for Be. More recent results indicate that Fe substitutes for Ti (Ref. 25).

The explanation for the loss of magnetism as Fe or Mn are substituted is not clear. However, since it is believed that these ions reside on Ti sites,[25] it has been argued[26] that the disturbance of the strong nearest-neighbor Ti—Ti bond in $TiBe_2$ by Fe or Mn substitution leads to a smearing out of the highly peaked density of states in $TiBe_2$ near E_F (Refs. 21 and 27) and to a consequent lowering of $N(E_F)$. Certainly, studies involving the substitution of Ti with other $3d$ impurities would be of considerable interest.

D. Landau expansion

We now discuss the present results in terms of the Landau expansion of the free energy F in powers of the magnetization M, field H, and pressure P:

$$F=aM^2+bM^4-MH+cP+gP^2+ePM^2+fPM^4+\cdots , \tag{8}$$

where a, b, c, g, e, and f are coefficients assumed to be independent of field H and pressure P.[28] Minimizing F with respect to M yields

$$H/M=2(a+eP)+4M^2(b+fP) , \tag{9}$$

which is appropriate for the description of straight

lines on an Arrott plot. From the low-field ($H<4$ T) data in Fig. 9 for $TiBe_2$ we obtain the values $a=(50.0\pm0.4)$ mol/emu, $b=(-1.08\pm0.01)\times10^{-5}$ mol^3/emu^3, $e=(4.8\pm1.0)$ mol/emu GPa, and $f=(1.0\pm0.2)\times10^{-6}$ mol^3/emu^3 GPa. At higher fields the lines in the Arrott plot bend over, indicating the need to include further terms in the Landau expansion. However, Acker *et al.*[9] point out that even including the next two higher powers of M in Eq. (8) or (9) does not allow a fit of the data with a single set of coefficients.

We now examine the temperature dependence of the susceptibility at different pressures using Eq. (9). Defining $\chi\equiv M/H$, we obtain the pressure derivative

$$\frac{1}{\chi^2}\frac{\partial\chi}{\partial P}=\frac{-2e-4fH^2\chi^2}{1+8bH^2\chi^3}\simeq-2e-4fH^2\chi^2\,, \tag{10}$$

the simplification following since here $8bH^2\chi^3\ll1$ for $H<4$ T. Inserting into Eq. (10) the above values of the coefficients e and f, we find that $2e$ dominates over $4fH^2\chi^2$, from which it follows that

$$\frac{\partial\chi}{\partial P}\approx-2e\chi^2\,. \tag{11}$$

We conclude from Eq. (11) that the rapid decrease of $|\partial\chi/\partial P|$ with increasing temperature observed for $TiBe_2$ in Fig. 3 can be qualitatively understood as arising from the observed rapid decrease in χ itself with increasing temperature (see Figs. 1 and 2). To facilitate a more quantitative analysis, we plot in Fig. 10 $\chi^{-2}(T)\partial\chi(T)/\partial P$ vs $\chi^2(T)$, which should give a straight line if Eq. (10) is appropriate and e and f are constants. Inserting the above low-temperature values of e and f into Eq. (10) gives the dashed line in Fig. 10 which is nearly horizontal, supporting the approximation leading to Eq. (11). The fact that the present data in Fig. 10 exhibit for $T\geq50$ K a strongly nonlinear dependence is evidence that the Landau coefficients possess non-negligible temperature dependences. The nonlinearity in Fig. 10 is another expression of the fact that χ becomes very insensitive to pressure for $T>50$ K, much more so than would be expected on the basis of Eq. (10) or (11).

E. Spin-fluctuation temperature

Because of the apparently very large exchange enhancement in $TiBe_2$, it is natural to attempt to account for the temperature and pressure dependences of the susceptibility $\chi(T,P)$ in terms of a spin-fluctuation picture with characteristic temperature $T_{SF}(P)$. Such an analysis in terms of a single characteristic energy kT_{SF} has been very successful for both intermediate-valence compounds[16,29,30] and dilute impurity alloy systems[29–31] where $\chi T=f(T/T_{SF})$ was shown to depend only on T/T_{SF}. To determine if the present data on $TiBe_2$ obey such a law, we plot in Fig. 11 $\chi T/C=[\mu(T)/\mu(\infty)]^2$, "the effective-moment squared," versus $\ln T$. Here $C\equiv N\mu^2(\infty)/(3\text{ K})$ is the Curie constant which we arbitrarily set equal to the value appropriate for trivalent Ti ions with $S=\frac{1}{2}$ and quenched orbital angular momentum, i.e., $\mu(\infty)=1.73\mu_B$. The quantity $\chi T/C$ is seen to have a rather peculiar temperature dependence with a slope change near 40 K. In addition, at different pressures the temperature dependence of $\chi T/C$ changes shape, so that the present data, in contrast to the data for the above systems,[16,29–31] *cannot* be interpreted in terms of a *single* characteristic energy kT_{SF}. We can, however, use the data for $T<100$ K to obtain the rough estimate that $T_{SF}(0)\approx25$ K and T_{SF} (1.3 GPa)≈29.5 K, where T_{SF} is defined as that temperature where $\chi T/C=\frac{1}{2}$.[29] This gives the value $\gamma_{SF}\equiv-\partial\ln T_{SF}/\partial\ln V=K^{-1}(\partial\ln T_{SF}/\partial P)\simeq+18$ for $TiBe_2$. Another way to estimate γ_{SF} is to assume the validity of the scaling relation[29,30] $T_{SF}\sim\chi(0)^{-1}$, which gives the value $\gamma_{SF}\approx+13$, in reasonable agreement with the first estimate. We compare these values of γ_{SF} with those for $CeSn_3$ ($+7.6$),[16] Pd ($+2$),[17] $(U_{0.06}Th_{0.94})S$ ($+2.7$),[32] and values between $+8$ and $+84$ for various dilute magnetic alloys.[31]

F. Simple model

One of the possibly more significant results of the high-field specific-heat studies by Stewart *et al.*[4] is that a *critical value* of the field $H_c\simeq5$ T must be exceeded before the apparent suppression of the spin-fluctuation effects in $TiBe_2$ actually begins. It would seem to be no accident that the anomaly in the low-temperature susceptibility $\chi(H)$ occurs near the same field value ($H\simeq5.6$ T) and that the low-field susceptibility maximum occurs at $T_c\simeq8$ K

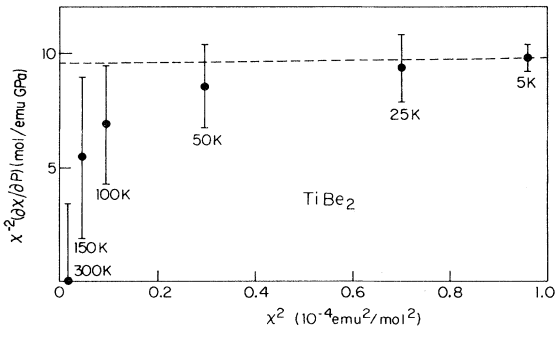

FIG. 10. Plot of $\chi^{-2}(\partial\chi/\partial P)$ vs χ^2 at different temperatures. Dashed line is fit using Eq. (10) with $e=4.8$ mol/(emu GPa) and $f=1.0\times10^{-6}$ mol^3/(emu^3 GPa) (see text).

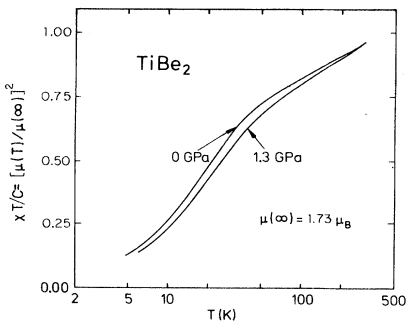

FIG. 11. Relative moment squared vs $\ln T$ for $TiBe_2$ at two pressures.

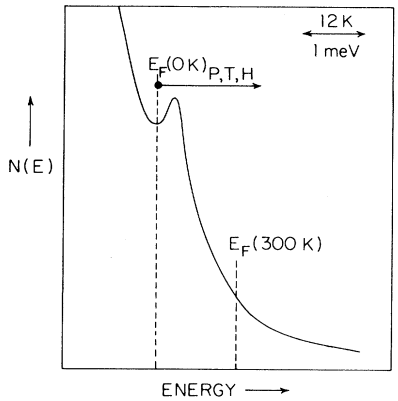

FIG. 12. Hypothetical fine structure in the energy dependence of the density of states near E_F for $TiBe_2$.

which is equivalent to the field ~ 6 T [for $g=2$, H (T)$=0.74T$ (K)]. A $\chi(P)$ anomaly at low T and H has not yet been observed, but would be anticipated. We also note that if any of the parameters exceed their critical values, the χ anomalies for the other parameters either disappear or are at least strongly diminished. Thus the $\chi(T)$ maximum vanishes for $H>5$ T (Ref. 15) as does the $\chi(H)$ maximum for $T>10$ K.[8,9] From Figs. 4 and 6 we see that a pressure of $P \geq 1$ GPa is quite effective in broadening the $\chi(T)$ anomaly and suppressing the $\chi(H)$ anomaly. It is interesting to note that from Fig. 7 the work done on $TiBe_2$, $\Delta E = \int P\,dV$, for a pressure increase of 1 GPa equals ~ 1 meV per formula unit, corresponding in energy to ~ 10 K or 7 T, which are near the critical values T_c or H_c. It thus appears that the parameters P, T, and H are all capable and willing to do away with each others' χ anomalies.

The above discussion would seem to indicate that certain critical values of the parameters must be exceeded in order to suppress the anomalous χ dependences. In the spirit of Eqs. (1) and (2), which contain both spin-fluctuation and band-structure effects, we suggest that the above anomalies can be qualitatively accounted for by a simple hypothetical model of the electronic structure of $TiBe_2$. In this model the Fermi energy at $T=0$ K would lie in a narrow (~ 10 K or ~ 1 meV) local minimum of the density of states, as represented schematically in Fig. 12. We emphasize that this discussion is purely speculative and is not based on microscopic evidence; indeed, the width of this local minimum would be well below the resolution of current band-structure calculations[21,22,27] or photoemission experiments.

Since according to Fig. 12 we would have $N''>0$ and $N'=0$ at the Fermi energy E_F, it would be expected from Eqs. (1) and (2) that $\chi(T,H)$ should initially increase with either temperature or field. This can also be easily understood directly from Fig. 12 since increasing T or H would shift E_F towards the local $N(E)$ maximum, thus increasing both $N(E_F)$ and χ. Pressure would broaden the $N(E)$ structure and shift E_F, which could account qualitatively for the pressure effects seen in Figs. 4 and 6. From Fig. 12 it would also be anticipated that the $\chi(H)$ anomaly should be broadened and shifted to lower field values as the local maximum in $N(E)$ is effectively smeared out by an increase in temperature, as observed in experiment.[9]

In this picture the strong increase in the magnetic response of $TiBe_2$ as the temperature is lowered would be associated with the Fermi energy E_F moving to lower energes and up the steep flank in $N(E)$ in Fig. 12. Ferromagnetism would be, however, prevented when E_F falls into the local $N(E)$ minimum for $T<10$ K. On the other hand, because of the initial pinning of E_F at the local minimum in $N(E)$, the suppression of spin-fluctuation effects would only begin for fields greater than 6 T which are large enough to push E_F past the local maximum and down the steep flank in $N(E)$. Stewart *et al.*[4] estimate that $H \geq 25$ T would be required to completely suppress the spin fluctuations, corresponding to the temperature $T_{SF} \simeq 34$ K, a value which is close to both our estimate and those of others.[4,14] The present description would, of course, cast doubt on the validity of using low-temperature data (here $T<10$ K) to determine the value of T_{SF} for $TiBe_2$. We note that the rather sensitive variation in the temperature dependence of the susceptibility at low temperatures from one $TiBe_2$ sample to another could be accounted for by considering the smearing effect of defect scattering on the fine structure in $N(E)$.

In the present picture, we suggest that the low-temperature $\chi(T)$ and $\chi(H)$ anomalies may arise from fine structure in the energy dependence of the density of states $N(E)$. Spin-fluctuation and band-structure effects are considered here as two distinct but interrelated entities. Because of these possible complexities, it would be easy to understand why the susceptibility data over a wide range of the parameters T, H, and P cannot be accounted for by including only the first few terms in the Landau expansion with a single set of coefficients. A quantitative interpretation of the highly anomalous behavior of $TiBe_2$ remains a challenge for the future.

In conclusion, we would like to point out that a $TiBe_2$ sample taken from an adjacent section of the same melt as ours was found to be nonsuperconducting at temperatures above 150 μK.[33]

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