Pressure dependence of the static magnetic susceptibility of the heavy-fermion superconductor UBe_{13}

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The pressure dependence of the static magnetic susceptibility $\chi(T)$ of the heavy-electron superconductor UBe_{13} was investigated over temperatures ranging from 1.5 to 300 K and at pressures from 1 bar to 8 kbar. It is linear in pressure over the full temperature range, having a small constant pressure dependence at higher temperatures that increases rapidly below about $T=150$ K. The maximal value of the relative susceptibility change $[1/\chi(d\chi/dP)]$ is about 1% per kbar. The Curie-Weiss law is obeyed in the temperature range 100 to 300 K with the effective magnetic moment $p_{\text{eff}}=3.46\mu_B$ independent of pressure, while the paramagnetic Curie temperature $\Theta \sim -100$ K becomes more negative with pressure. The lattice parameter of cubic UBe_{13} was measured as a function of pressure, permitting a determination of a compressibility value of -9.2×10^{-4} kbar^{-1} near ambient pressure. The pressure dependence of $\chi(T)$ is significantly weaker than that of the specific-heat coefficient $C(T)/T$; we discuss whether this can be resolved by (i) consideration of intersite antiferromagnetic interactions, or (ii) the quadrupolar Kondo-effect model, and conclude that the latter provides a stronger explanation of the results.

I. INTRODUCTION

UBe_{13} has received considerable attention as a heavy-fermion superconductor, particularly because it has the highest crystalline symmetry (cubic), which makes it ideal to test whether the superconducting states are intrinsically anisotropic.[1] Of course, a prerequisite to a complete understanding of the unusual superconducting properties requires an understanding of the normal state. The normal state of UBe_{13} is quite unusual, and studies of properties under applied pressure have underscored its anomalous character.

In this paper, we present measurements of the pressure-dependent magnetic susceptibility of UBe_{13} which may shed further light on the anomalous character of this material. For example, measurement of the low-temperature electronic-specific-heat coefficient C_{el}/T has given a Kondo scale T_K (or degeneracy temperature) of the order of 6 K.[2] In contrast, the high-temperature susceptibility $\chi(T)$ as measured by Troc', Trzebiatowski, and Piprek possesses a Curie-Weiss (CW) law with Curie constant $p_{\text{eff}}=3.4\ \mu_B$ and $\Theta=-70$ K.[3] Since typically one has the Curie-Weiss Θ for a Kondo system of the order of $\sqrt{2}T_K$, this is clearly an order of magnitude difference in scales. Indeed, the general behavior observed for UBe_{13}, and some of the other heavy-electron systems like $CeAl_3$ (Ref. 4) and $CeCu_2Si_2$ (Ref. 5) is somewhat different than most of the "valence fluctuation" (or possibly high T_K) systems such as $CeSn_3$ or $YbCuAl$ (Refs. 6 and 7) where

high-temperature CW behavior gives way to temperature-independent Pauli-like χ behavior be low some characteristic temperature. The $\chi(T)$ data of UBe_{13} are seen to have CW-type behavior at high temperatures which becomes more complicated below about 100 K. Indeed, the temperature dependence of $\chi(T)$ remains pronounced down to at least 1.5 K, and, in fact, $d\chi/dT$ increases monotonically with decreasing temperature.

Pressure studies have also proven to be quite useful probes of the heavy-fermion state. For many enhanced mass materials with larger degeneracy temperatures than the 6 K of UBe_{13}, one-parameter scaling in the resistivity, susceptibility, and specific heat has been observed under applied pressure. These observations are consistent with the results of Kondo-Anderson impurity and lattice theories which suggest that above any cooperative phase transitions, the Kondo temperature T_K should be the only relevant temperature scale in the problem.

UBe_{13} is rather different in this regard. The electronic specific-heat coefficient C_{el}/T undergoes a large depression with applied pressure,[8] concomitant with a shifting of the resistivity shoulder to higher temperatures, the opening of a T^2 term in the resistivity over a temperature range that broadens from essentially zero at zero pressure to ~ 11 K at 150 kbar.[9] These data in themselves resemble the work on other heavy-fermion materials. However, the one-parameter scaling does not work. Unlike the other systems studied, the *residual resistivity* of UBe_{13} diminishes with increasing pressure.[9-11] As we shall briefly discuss in the interpretation section, this is incom-

patible with any simple Kondo lattice picture. In addition, the results of this study show that the susceptibility has a very different pressure dependence than the specific heat. Taken together with the very different energy scale that appears in the high-temperature susceptibility from the specific heat, this provides strong empirical evidence against single-parameter scaling in this model. Namely, the energy scale of the susceptibility is different than that of the specific heat. There are two leading explanations for this result. (i) The susceptibility is dominated by strong antiferromagnetic correlations while the specific heat is dominated by the Kondo effect, which is compatible with a mean-field theory of the antiferromagnetic correlations. This picture could apply if the U ions had a nominal magnetic doublet ground state. (ii) The susceptibility has an entirely different physical origin (through intercrystal field excitation or van Vleck processes) than the specific heat. This picture is precisely what follows from the proposed quadrupolar Kondo theory of UBe_{13} in which the ground doublet is nonmagnetic in character.[12] We shall argue that the evidence at hand favors the latter scenario.

An outline of the paper is as follows: in the Sec. II we will give the experimental details for our work, and in Sec. III present the data. In Sec. IV, we will discuss the possible scenarios for understanding our data and make connection to previous experimental and theoretical work. In Sec. V we will conclude and suggest new directions for research.

II. EXPERIMENT

The polycrystalline UBe_{13} sample was cut from the same ingot from which samples used in our previous investigations were taken.[13] The UBe_{13} ingot was prepared by arc melting high-purity elements (99.97% U, 99.9% Be) together on a water-cooled copper hearth in a Zr gettered Ar atmosphere in a manner previously described.[13] The sample quality is reflected in its high superconducting transition temperature $T_c \sim 0.905$ K and very sharp inductive transition width $\Delta T_c = 0.030$ K.

Magnetic-susceptibility measurements under pressure were made in a miniature self-clamping BeCu pressure cell.[14] The sample consisted of several disks with a total mass of 100 mg and the pressure clamp with a mass of about 45 g. The BeCu used in the pressure clamp was a custom-made alloy of 2% Be in Cu which was heat treated according to a procedure previously reported.[14] This binary alloy has a yield strength of about 12 kbar, considerably lower than the 25 kbar normal for the Co containing Berylco-25 alloy popularly used in pressure cell construction. Although the pressure range is more limited for the binary alloy, the complete temperature range of the magnetometer is available as a result of the reduction in paramagnetic and ferromagnetic background contributions. In order to further minimize background corrections, the diamagnetic susceptibility of the clamp was offset with foil of the temperature-independent paramagnet Pt. Pt was chosen to replace the Ta foil previously used because the superconducting transition of Ta obscures the observation of the manometer T_c.

Pressures were inferred from ac-susceptibility measurements of the T_c of a 1 mg Sn foil manometer contained in the Teflon sample capsule. The pressure media was 1:1 isoamyl:n-pentane solution. Magnetic susceptibility measurements were made in a Faraday balance magnetometer consisting of a balance with 0.001 mg resolution and a cryostat fitted with ± 5 T superconducting solenoid and ± 200 G/cm superconducting gradient coils. Temperature stability of $\pm 0.05K$ was maintained during measurement.

It should be pointed out that for most quasihydrostatic pressure cells there are significant changes in the applied pressure as the temperature is changed from 300 to 1.5 K.[15] These changes result primarily from differences in thermal contraction of the materials used in the construction of the pressure cell.[14] The pressures reported here are the pressures inferred from the known pressure dependence of the Sn manometer at temperatures below 4 K.[14,15] The pressures retained at 4 K were about $\frac{1}{3}$ of that calculated from the applied load at room temperature.

In order to determine the unit cell volume and check for structural transitions, pressure-dependent x-ray-diffraction measurements were made at room temperature using a single crystal of UBe_{13} for pressures ranging from 1 to 50 kbar. The sample was mounted in a Merrill-Bassett–type diamond anvil pressure cell fitted with Be supports for the diamonds. The pressure media was a 1:1 methanol:ethanol solution and the pressure was determined from the fluorescence of ruby powder contained in the sample space. The pressure was checked both before and after x-ray measurements. Single-crystal x-ray-diffraction measurements were made on a four-circle diffractometer.

III. RESULTS

Shown in Fig. 1 is a plot of the unit cell volume as a function of pressure. From the initial slope dV/dP a compressibility value

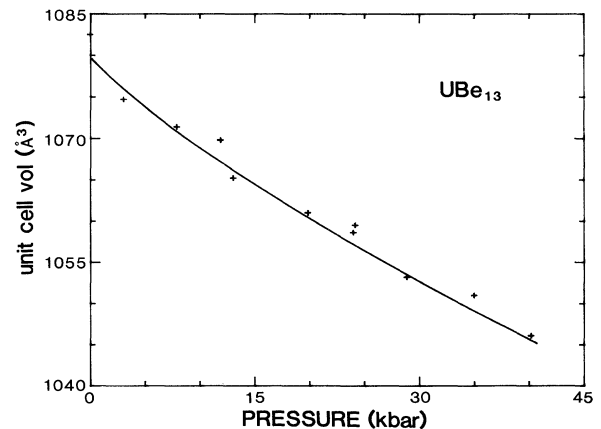$$-\frac{\partial V}{V_0 \partial P} = -9.2 \times 10^{-4} \text{ kbar}^{-1} ,$$

FIG. 1. Volume of the UBe_{13} unit cell as a function of pressure.

where V_0 is the initial volume, can be determined. Measurements with larger pressure spacings at pressures to 450 kbar were made by Benedict *et al.*[16]

Measurements of $\chi(T)$ between 1.5 and 300 K were made at five different pressures ranging from ambient pressure to 8 kbar. The curves of $\chi(T)$, for temperatures between 1.5 and 300 K, are plotted in Fig. 2(a) where it can be seen that the general shape of curves are essentially unchanged with pressure. The small relative effect is evident from Fig. 2(a) where it can be seen that the magnitude of the pressure dependent depression of χ decreases monotonically with increasing temperature, with χ appearing nearly pressure independent at high temperatures $T \gg |\Theta|$ where Θ is the Curie-Weiss temperature. In Fig. 2(b) are plots of $1/\chi$ versus T at 1 bar and 8 kbar where it can be seen that there is a significant range over which CW-type behavior appears to exist, although in previous $\chi(T)$ measurements to much higher temperatures there were some deviations observed up to about 300 K.[1] In Fig. 3 are values of CW Θ determined from linear fits to χ^{-1} from 100 to 300 K. These fits yield a constant value for the effective moment p_{eff} of 3.46 μ_B over the range of pressures studied. This result is not unexpected since at higher temperatures the moments have atomic characteristics and as long as the crystal-field levels do not cross, the magnitude of the moment should remain nearly constant. The plot of the CW Θ versus P, shown in Fig. 3 has a slope of 0.6 K/kbar.
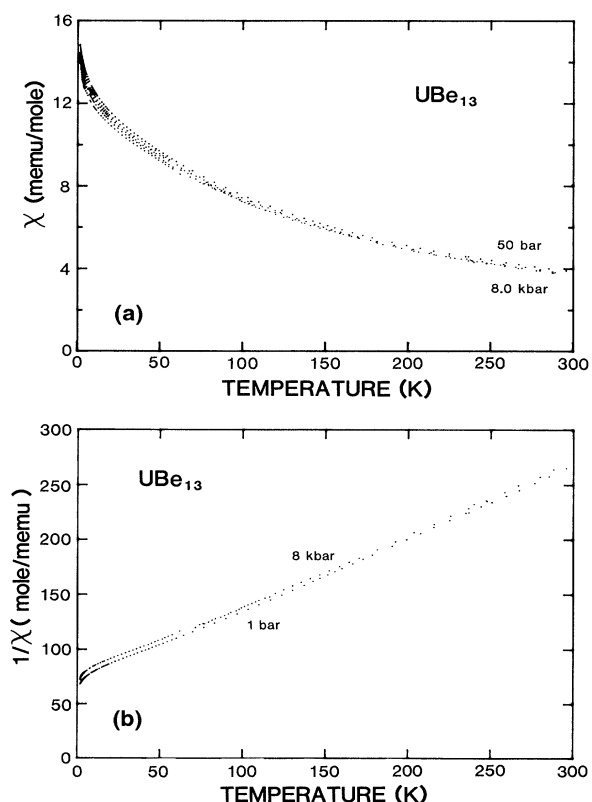
FIG. 3. The Curie-Weiss Θ determined from high-temperature exrtrapolations as a function of pressure. The line represents a linear fit to the four points clustered around the line; we do not understand the origin of the clearly deviated point.

At any given temperature, χ is observed to be linearly dependent on P. The slopes of $\chi(P)$ curves, at all the temperatures measured, are plotted in Fig. 4. The small value of $d\chi/dP$ at higher temperatures is consistent with the existence of local moments for $T \gg T_K$. Note also that in the Curie-Weiss regime one can determine $d\Theta/dP$ from the data plotted. Namely, assuming that only Θ changes with P one has by differentiating χ the relation

$$d\Theta/dP = C(-1/\chi^2)d\chi/dP \ .$$

The relative effect of pressure is more clearly shown in Fig. 5 in which the relative χ change $(\Delta\chi/\chi)$ is plotted as a function of temperature, where $\Delta\chi = \chi(P) - \chi(0)$. The large errors, evident from around 50 to 100 K, are instrumental errors associated with χ of the combined clamp and sample having values near $\chi = 0$. From these plots it is possible to see that the relative effect of pressure extrapolates to about 1% per kbar at 0 K. This effect is considerably smaller than that observed for measurements of

FIG. 2. (a) The magnetic susceptibility χ of UBe_{13} as a function of temperature $\chi(T)$ at pressures of 0.001, 1.2, 2.7, 4.3, and 8.0 kbar for T from 1.5 to 300 K, and (b) inverse susceptibility $1/\chi(T)$ at pressures of 0.001 and 8.0 kbar.
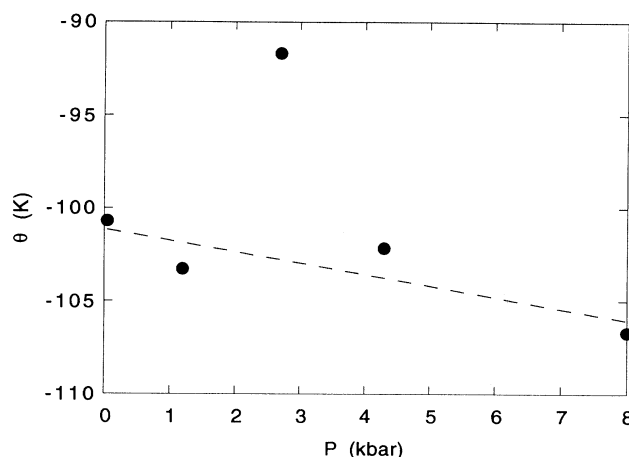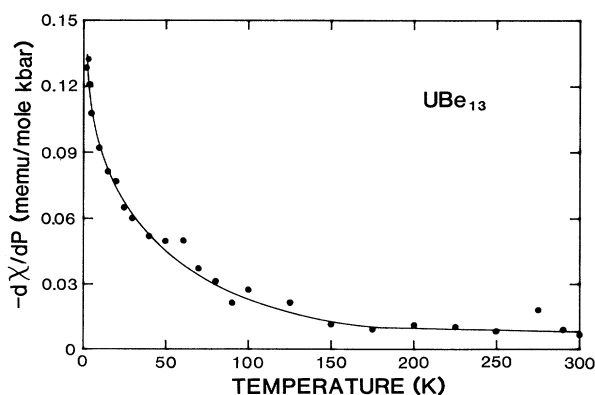
FIG. 4. Slopes $-d\chi/dP$ of plots of magnetic susceptibility χ versus of pressure P as a function of temperature. A magnetic field $H = 1$ T was used for all of the χ data utilized in this plot.

pressure-dependent specific heat,[8] where a 30% decrease in the electronic coefficient γ at 9.3 kbar was observed (i.e., 3%/kbar).

IV. INTERPRETATION AND CONNECTION WITH PREVIOUS RESULTS

A. Previous relevant data and phenomenology

Measurements of the magnetization M as a function of magnetic field H to 5.5 T at 5 K have been shown to be linear to within 2%.[3] The M versus H measurements under pressure presented here were linear over the full 5 T field range. The observed decrease in χ with pressure is characteristic of decreasing slopes of M versus H with pressure which is consistent with our results indicating that T_K is increasing with pressure. This result is important, since the 6 K scale evident in C/T would suggest for a magnetic ground state that $M(H)$ should show curvature on the 5–6 T scale. That it does not underscores the different energy scales present in the susceptibility and specific-heat data.

Pressure dependence of the resistivity has yielded remarkable results. Not only does a T^2 region over a finite-temperature range open up with pressure (this is not seen at ambient pressure), but the T^2 coefficient de-

FIG. 5. Pressure-induced changes in the magnetic susceptibility Δ_c, normalized by χ at $P = 0.001$ kbar, as a function of temperature (a) at pressures of 1.2 and 8.0 kbar over the full temperature range, and (b) at pressures of 0.001, 1.2, 2.7, 4.3, and 8.0 kbar at temperatures below 50 K.

creases with increasing P.[9–11] The secondary peak in the resistivity near 8 K is suppressed with pressure. Most remarkable is that the apparent residual resistivity of order 80–100 $\mu\Omega$ cm at T_c is rapidly suppressed with pressure. As we shall argue, it is very difficult to reconcile this last result with any Fermi-liquid picture of the material.

It has been argued that UBe_{13} is simply a very strong-coupling superconductor in which the onset of superconductivity at 0.9 K $\approx T_F^*/10 \approx T_K/10$ obscures the development of a full Fermi-liquid regime. In any conventional Kondo lattice theory, Fermi-liquid behavior in the form of a T^2 coefficient sets in at below $T_K/10$. This argument is of course consistent with the lack of T^2 behavior in the resistivity at ambient pressure. However, in order to support this contention, one would expect to see the specific-heat coefficient C/T roll over and flatten out below T_c, when, in fact, explicit field-dependent measurements that suppress T_c and entropy balance arguments show that C/T continues to *rise* as the superconductivity is suppressed. Hence it is *unlikely* that one can cleanly account for UBe_{13} as a Fermi liquid.

It has also been argued that the magnetoresistance of UBe_{13} places it in the class of a standard magnetic Kondo lattice.[17] The data have been fit to a Kondo impurity form with a temperature-dependent Kondo scale which extrapolates to zero linearly as $T \rightarrow 0$, in at least some of the reported data. The magnetoresistance remains negative to the lowest temperature measured, and is almost entirely born by the residual resistivity. In point of fact, this is *not* the expected behavior for a conventional Kondo lattice. In a conventional Kondo lattice, the magnetoresistance would be negative and fit by the impurity result only above $T \approx 0.15 T_K$.[18] Below this temperature, the resistivity will initially rise with increasing field as the singlets are depolarized, and then fall well above $\mu_B H \approx k_B T_K$ as the scattering is frozen out of the fully polarized ions. Moreover, there is *no residual resistivity* in the pure and conventional Kondo lattice, so that all the field dependence is born by the coherent part of the resistivity at low temperature. In contrast, *all of the field dependence* at low T for UBe_{13} is carried by the residual resistivity. Hence the unusual negative magnetoresistance of this material *argues against* a conventional magnetic Kondo lattice interpretation.

A separate observation is that of Fisk *et al.*[19] who have noted a correlation between the linear coefficient of electronic specific heat per unit volume $\gamma_v(\equiv C_v/T)$ and the ground-state configuration of U-based heavy-fermion systems. Plots of $C(P)/T$ versus T were constructed from the pressure-dependent specific-heat results from which the Sommerfeld coefficient could be estimated at $T = 0$; with a value of $\gamma(0) \cong 630$ mJ/mole K^2 determined at $P = 9.3$ kbar. Using a value of 0.97×10^{-3} kbar^{-1} for the compressibility of UBe_{13},[16] γ_v was calculated to have a value of 7.83 mJ/cm^{3}K^2 at 9.3 kbar, which is almost one-half its value at ambient pressure and is approaching the values for $\gamma_v \cong 5.3$ mJ/cm^{3}K^2 of the heavy-electron magnets UCd_{11} and U_2Zn_{17}.[20] Another observation supporting an increase of the magnetic character with pressure are the increases of the magnetic transition tempera-
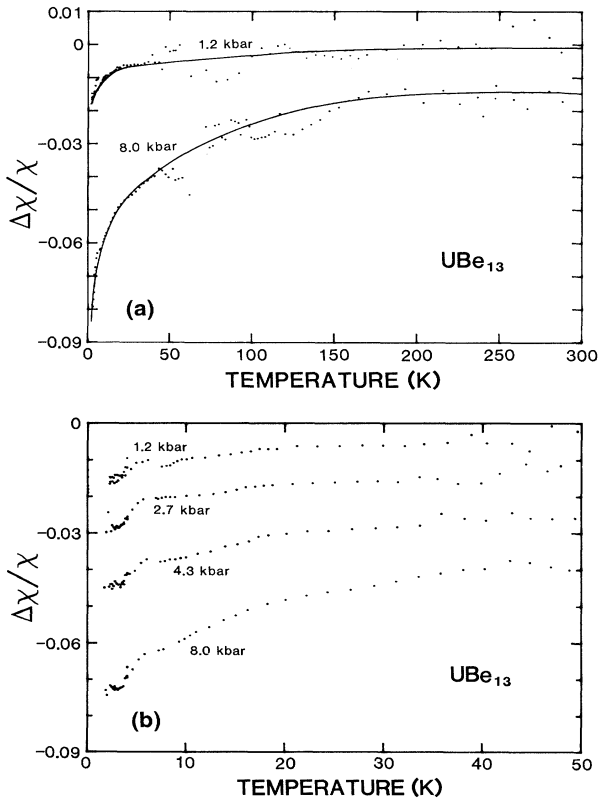
tures for the heavy-electron magnets UCd_{11} and U_2Zn_{17} and for the magnetic transition temperatures of the heavy-fermion superconductors URu_2Si_2 and UPt_3.[20] These changes are likely associated with the changes in electronic structure since the crystal symmetry does not change with uniform pressure.

It is important to note that the neutron-scattering data of Shapiro et al.,[21] the nuclear magnetic spin-lattice relaxation data of Clark,[22] and the specific-heat Schottky anomaly data of Felten et al.[23] point to the existence of highly damped crystalline electric-field excitations at about 15 meV above the lowest-lying U multiplet. In addition, the NMR spin-lattice relaxation data reveals temperature dependence on the 10 K scale. Moreover, the highly damped feature in neutron scattering almost completely exhausts the static susceptibility.[21] We observe that this crystal-field scale is comparable to the Θ value identified from our Curie-Weiss fits to $\chi(T)$, and that below 100 K ~ 10 meV the susceptibility breaks from the Curie-Weiss form. We note that more recent neutron data[24] support the presence of some low-lying frequency structure in the dynamical susceptibility, a point we shall return to below when we discuss the modification of the van Vleck susceptibility in the presence of the quadrupolar Kondo effect.

Finally, we remark on the data for the magnetic susceptibility on entering the superconducting state. There is no change of the ^9Be (Ref. 25) Knight shift or neutron form factor determined $\chi(\mathbf{Q})$ (Ref. 26) upon entering the superconducting state. At most the muon Knight shift[27] displays a small, field-*independent* "jump" upon entering the superconducting state. This information is of assistance in discriminating between the two scenarios we present in the next subsection.

B. Interpretation

What is clear from our pressure-dependent susceptibility data is that the specific heat and susceptibility have very different physics. Specifically, χ is more weakly pressure dependent than C/T, and the energy scale evident in χ from high temperatures is quite different from the low-temperature scale evident in C/T. We compare two possible explanations for this.

1. Antiferromagnetic correlations in a Anderson lattice

Let us assume the U ions are trivalent with a $J = \frac{9}{2}$ ground multiplet from Hund's rules, and a possibly stable Γ_6 ground magnetic doublet as argued by other authors.[23] Given the full multiplet effective moment of 3.63 μ_B (within LS coupling), this scenario is reasonably compatible with the observed value of 3.4 μ_B from our high-T Curie law fits. In this case, we might model the U ions with a standard *magnetic* Anderson lattice Hamiltonian, with parameters to place us in the Kondo regime so we may obtain the small T_K value of 6 K.

The high $T (> 100$ K) fits to the $\chi(T)$ would then be consistent with strong antiferromagnetic correlations between U ions. Recent calculations by Si, Lu, and Levin show how the Anderson lattice will contain effective

spin-spin interactions mediated by particle-hole pairs [Ruderman-Kittel-Kasuya-Yosida (RKKY)] and particle-particle pairs (superexchange) both of which scale as V^4, where V is the hybridization matrix element between f and conduction electrons.[28] The dominant short-distance behavior should be antiferromagnetic. Similar conclusions for the intersite exchange follow from a completely localized limit with realistic orbital anisotropies as described in Ref. 29.

This interpretation has three main strengths.

(1) In a simple minded mean-field theory,[30,31] the uniform susceptibility is given by

$$\chi(T) \approx \frac{\chi_0}{1 - qI\chi_0(T)/(k_B\mu_{eff}^2)} , \qquad (1)$$

where χ_0 is the on-site susceptibility of a model material with zero intersite interaction I and coordination number q. Assuming $I < 0$ (antiferromagnetic ordering) then we see that since $\chi_0 \sim (T + \sqrt{2}T_K)^{-1}$ that

$$\Theta \approx \max(q|I|/k_B, \sqrt{2}T_K) .$$

This interpretation thus allows the susceptibility scale of the interacting system to be different from that of the noninteracting system and to show a different temperature dependence than the specific heat. This follows since in such a mean-field analysis, until one obtains magnetic order, the specific heat would be that of the noninteracting limit. Hence, the discrepancy between Θ and T_K could be resolved in this way.

(2) Obviously, this could explain the different pressure dependence of the susceptibility as well as the specific heat and susceptibility. While the intersite interaction energy I would scale approximately as the hybridization to the fourth power, in a mean-field treatment, the specific heat would still reflect the single ion Kondo scale $\sim \exp(-A/V^2)$. Clearly, the pressure dependence of the specific-heat coefficient would be much stronger than the susceptibility, which qualitatively checks with our results.

(3) Moreover, there may be evidence for antiferromagnetic correlations in UBe_{13}.[32]

In addition to these strengths, since the conventional Kondo lattice is expected to have a Fermi-liquid excitation spectrum in the absence of cooperative instabilities, and since the Fermi temperature T_F^* is of order T_K, one would generically expect a T^2 behavior in the resistivity and $d\rho/dT^2 \sim T_K^{-2}$ to decrease with increasing pressure.

The central objection to this possible explanation is that if Θ is due to antiferromagnetic interactions and $|I| \gg T_K$ holds, we should have already observed antiferromagnetic order well above $T_K \approx 10$ K. No such order exists. If this were the correct explanation, one would also anticipate the magnitude of χ to change significantly upon dilution of the U ions by adding, e.g., Th. This is because one will significantly alter the mean coordination number of the magnetic moments. However, Kim et al.[33] find that the value of $\chi(T \to 0)$ is *insensitive* to doping (though C/T does change significantly) and have inferred that the susceptibility is local in character. Also, a conventional Kondo lattice Fermi-liquid picture would

meet considerable difficulty reconciling the temperature scale of the specific heat and NMR low-T $1/T_1$ data (10 K) with the absence of a T^2 region in $\rho(T)$, the continued rise of C/T below T_c with application of magnetic field,[34] and the absence of significant magnetic-field dependence in all but the magnetoresistance.[17] We discussed these issues in the previous subsection at length. Another issue is the fact that the Knight shift does not change on entering the superconducting state.[25,27] If we had a band of heavy magnetic quasiparticles, a Knight shift change would necessarily occur for all even-parity states and most odd-parity pairing states. Finally, the strong pressure dependence of the residual resistivity is quite incompatible with a Fermi-liquid theory. The value in ambient pressure corresponds to nearly resonant scattering; application of pressure will not change the resonance condition.

2. Quadrupolar Kondo effect

An alternative scenario is that the U ions are actually tetravalent with a $J = 4$ Hunds' rules ground multiplet. It has been argued elsewhere that in this situation a *nonmagnetic* Γ_3 ground doublet may lie lowest.[12] In this circumstance, the U ions are subject to a quadrupolar Kondo effect. In this case, the magnetic susceptibility derives from virtual magnetic excitations to the two magnetic triplet levels of the $J = 4$ multiplet, i.e., is of van Vleck form. [At a site of cubic symmetry, the $J = 4$ multiplet splits into a doublet (Γ_3), two triplets (Γ_4, Γ_4), and a singlet (Γ_1).]

A number of results follow immediately from this picture for UBe$_{13}$.

(1) The difference of energy scale and pressure dependence in $\chi(T)$ and C/T is immediately clear. The latter reflects the quadrupolar Kondo temperature T_K, the former the crystal-field splitting Δ. Theory for a single quadrupolar Kondo site shows that the van Vleck susceptibility is little changed from the ionic form,[12] and it has been noted that the 15 meV energy associated with the neutron-scattering cross section and the specific-heat Schottky anomaly is of precisely the right magnitude for the measured low-temperature susceptibility, assuming a Γ_4 triplet is at this energy. The effective moment of the susceptibility is fixed and independent of pressure in this case, in agreement with our finding of the high-temperature χ data. The crystal-field splitting should scale as V^2 (Ref. 35) while again, the Kondo scale goes as $\exp(-A/V^2)$, so the pressure dependence of C/T should be stronger.

(2) A purely ionic ($V = 0$) van Vleck susceptibility would saturate exponentially at low T, obviously inconsistent with the data. However, when the coupling to electrons is included, the saturation turns to the form[36]

$$\chi(T) \sim [1 - A(T/T_K)^{1/2}]/\Delta \quad (2)$$

with $A \sim 1$ which does fit the data, as shown in Fig. 6, over the limited region 1–4 K. The above form is expected to be asymptotically valid for $T \le 0.3$–$0.5T_K$, given the width of the critical regime evident in the pure two-channel model calculations of Ref. 38. Thus only a small
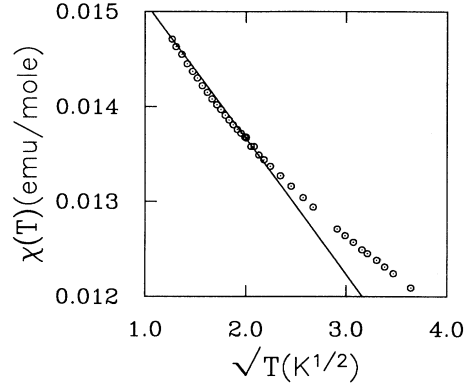

FIG. 6. Susceptibility $\chi(T)$ versus $\sqrt{T}$. The points are from experiment, the line a fit to the data in the 1–4 K range by the form $\chi(0)(1 - A\sqrt{T/T_K})$ [cf. Eq. (1)], with $A = 0.21$ (assuming $T_K = 6$ K) and $\chi(0) = 0.0165$ emu/mole. The van Vleck susceptibility should take on such a form for a quadrupolar Kondo material (see Ref. 36).

region of data is available to match to given $T_K \sim 10$ K.

The origin of the square-root term is qualitatively understood as follows. First, consider the ionic ($V = 0$) limit. In that case, one finds for a single excited crystal-field multiplet that

$$\chi(T) = \chi(0)[1 - e^{-\Delta/k_B T}] .$$

The second term arises from thermal occupancy of the excited level which is frozen out as the temperature tends to zero. Once hybridization is turned on there will always be some quantum occupancy of the higher crystal-field state which will allow virtual transitions from the occupied fraction of the higher crystal-field level to the lowest crystal-field level. Since the hybridization broadens delta-function energy distributions to continua, it is not surprising that the exponential dependence is replaced by a power law. The particular power law reflects the *singular* behavior of the ground Γ_3 doublet in the quadrupolar Kondo model. Since the low-energy physics of this model are those of a two-channel Kondo model, the spectral response of the Γ_3 is singular, diverging as

$$1/\sqrt{\max\{\omega - E_0, T\}}$$

on approach to zero temperature and the ground-state energy E_0.

(3) The pressure dependence of χ as a function of temperature seems to be semiquantitatively explained in this picture. We assume that (a) the pressure dependence of the hybridization is strong, that of the f-level splitting weak (in fact, this needs to be checked for U ions), and (b) that the crystal-field splitting $\Delta \sim g$, where $g \sim V^2$ is the dimensionless effective exchange coupling of the local quadrupole moments on the U sites to the conduction electrons, with

$$k_B T_K \simeq E_F \exp(-1/g) ,$$

and E_F the Fermi energy. Then given the above form for $\chi(T)$ and

$$C/T \sim (1/T_K)\ln(aT_K/T) ,$$

where a is a parameter determined from the background specific heat of excited crystal-field states,[37] we see with some straightforward algebra that

$$\frac{\partial \ln\chi(T)}{\partial P} \approx \frac{1}{B}\frac{\partial \ln g}{\partial \ln v}\left[1 - \frac{1}{2g}\left[\frac{T}{T_K}\right]^{1/2}\right] \qquad (3)$$

and

$$\frac{\partial \ln(C/T)}{\partial P} \approx \frac{1}{gB}\frac{\partial \ln(g)}{\partial \ln v}\frac{\ln(aT_K/eT)}{\ln(aT_K/T)}, \qquad (4)$$

where B is the bulk modulus, v is the specific volume, and e is the natural logarithm base. These formulas suggest (1) the pressure dependence of the specific-heat coefficient should be much stronger than that of the susceptibility due to the $1/g$ out front, and (2) that the slope of the $\sqrt{T}$ term in $\partial\ln\chi/\partial P$ is larger than in $\chi(T)/\chi(0)$, which is consistent with the observation of stronger pressure dependence to χ as the temperature is lowered. The square-root correction rapidly becomes less and less important as the temperature is lowered. If we take the reasonable estimate $g \approx \frac{1}{7}$ to get $T_K = 10$ K for $T_F = 10^4$ K, then at low T (1 K), we get the pressure derivative of the specific heat to be 2–4 times that of the susceptibility as we vary a from 0.41 (the universal number in the pure two-channel limit[38]) to 1.0 (reflecting a specific-heat tail rom the excited crystal-field states). Hence, this model provides a plausible *quantitative* explanation for the discrepancies in susceptibility and specific-heat pressure dependence.

(4) The Curie constant works out quite reasonably if, as per Ref. 39, we take the excited magnetic triplet levels (Γ_4 and Γ_5) to be essentially degenerate. This requires the Γ_1 to be substantially higher in energy (at about 400 K). Thus we may estimate the Curie constant from the lowest eight states alone. In LS coupling (weak spin-orbit basis), we obtain 3.34 μ_B for the effective moment, while in j-j coupling (strong spin-orbit basis) this goes up to 3.58 μ_B. The U ion is described by intermediate coupling, though it is closer to the LS limit[40]; clearly these estimates adequately bracket the observed value.

The above results provide the core of our application of the quadrupolar Kondo model to interpret our data. In addition, the model supports several other results, including the following.

(a) The lack of strong magnetic-field dependence is understood since the quadrupolar doublet couples to field only at order H^2. The linear magnetization[41] and absence of a Knight shift change at T_c (Refs. 25 and 27) are understood to arise from the van Vleck susceptibility. In the case of the Knight shift, no change is expected in the van Vleck χ since it corresponds to such a high-energy scale.

(b) The absence of Fermi-liquid properties at low T follows from the fact that a quadrupolar Kondo model is an example of a so-called overcompensated multichannel Kondo model. For a single site, the specific-heat coefficient diverges logarithmically. The resistivity will show no T^2 behavior, and in the absence of coherence among the quasiparticle spins a kind of residual resistivity corresponding to "spin disorder scattering" results.[36,42]

(c) Pressure increases the overlapping of the crystal-field levels and drives the model towards Fermi-liquid behavior.[36]

(d) A possible resolution of the magnetoresistance puzzle is also offered. The essential idea is that application of a field to a single impurity drives the system to a Fermi liquid which is described by a phase shift.[43] Hence, in the lattice at low temperatures the field will remove the incoherent "spin-disorder scattering" (a periodic phase shift simply renormalizes the band electron potential). Hence the resistivity should fall, and the crossover field for this resistance fall will tend to zero as the temperature tends to zero, though not necessarily linearly.

C. Summary and conclusions

In summary, we have performed pressure-dependent magnetic susceptibility measurements on the heavy Fermion superconductor UBe$_{13}$. We find that the susceptibility is weakly pressure dependent in the high-temperature regime where a Curie-Weiss law (with $\Theta \approx -100$ K, $\mu_{\rm eff} \approx 3.4\ \mu_B$) describes the data, and grows more strongly pressure dependent at low T. However, the low-temperature pressure dependence (at 1.4 K) is about $\frac{1}{3}$ that of the specific-heat coefficient, which clearly is compatible with an order 10 K Kondo scale.

Taking into account the other properties of the material, we have considered alternative interpretations for the data. First, we assumed that the differences between χ and C/T arise from antiferromagnetic correlations. While this provides an explanation for the different temperature scale and pressure dependence of χ relative to C/T, it fails to account for the absence of magnetic order, the insensitivity of χ to doping on the U sublattice, and the absence of a Knight shift change on entering the superconducting state. Second, we considered the quadrupolar Kondo picture, in which a nonmagnetic Kondo effect produces the specific heat while the susceptibility is van Vleck in origin. This picture appears to provide a semi-quantitative explanation of the data presented here, while providing a more comprehensive view of related data.

It is important to specify further tests of the quadrupolar Kondo picture. If, as suggested, uniform pressure increases the overlap between crystal-field levels, then a substantial broadening of the 15 meV feature observed in neutron and Raman scattering should be observed, and concomitantly the Schottky anomaly in the specific heat should broaden considerably, while the features observable in spin-lattice relaxation data at 10 and $\sim$100 K should coalesce.

It should also be interesting to study Th-doped UBe$_{13}$, especially in the dilute limit. This will allow a clearer establishment of the relevance of the quadrupolar Kondo picture, because the single site model of course becomes rigorously applicable in that limit. Even at relatively high concentrations, the suppression of the superconductivity by the Th doping allows examination of the $\chi(T)$ behavior at lower temperatures to see if the $\sqrt{T}$ suggested above holds in the true asymptotic regime ($T \ll T_K$). In addition, the $T\ln T$ specific heat expected for a two-

channel Kondo model should become apparent. This does occur for some intermediate concentrations in the data of Aliev et al.[44]; in particular, it is quite clear for $U_{0.64}Th_{0.36}Be_{13}$. Finally, Th has the effect of negative pressure on the lattice since the Th ions increase the lattice constant. Thus examination of the high-energy scale neutron-Raman excitation peak near 15 meV should show narrowing relative to UBe_{13}. Application of pressure would then drive the spectra Th-doped material back towards those of UBe_{13}. Since the pressure affects the crystal-field linewidths (which are essentially Kondo scales) more than the line positions (which scale as V^2) then the magnitude of χ at low temperatures ($\sim 1/\Delta$, where Δ is the crystal-field splitting) would be little affected. The data of Aliev et al. shows a narrowing of the overall resistivity maximum upon Th doping which would be consistent with this narrowing hypothesis.[44]

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