Title

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Journal

Physical Review B, 42(10)

ISSN

2469-9950

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Publication Date

1990-10-01

DOI

10.1103/physrevb.42.6062

Effect of high pressure on the electrical resistivity of the heavy-fermion compound UBe_{13}

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(Received 29 November 1989; revised manuscript received 22 June 1990)

The electrical resistivity ρ of the heavy-electron superconductor UBe_{13} was investigated over temperatures ranging from 1 to 300 K and at high pressures from 12 to 152 kbar. The temperature, T_{max}, at which ρ attains a maximum increases linearly with pressure at two limiting rates: 0.25 K/kbar below ~30 kbar and 0.65 K/kbar above ~30 kbar. At low temperatures, the ρ-versus-T data can be described by the expression $\rho=\rho_0+AT^2$ over a temperature interval of ~1 K at 12 kbar to more than 16 K at 152 kbar. The coefficient A can be related to the Fermi-liquid energy scale T^* by $A^{-1/2}\propto T^*$. The pressure dependence of $A^{-1/2}$ also increases linearly with pressure at two limiting rates, with a crossover at about 50 kbar. The similarity in the pressure dependences of T_{max} and $A^{-1/2}$ suggests that both parameters are proportional to a single energy scale, while the changes in pressure dependence of T_{max} and $A^{-1/2}$ may represent a crossover from heavy-fermion to intermediate-valence behavior.

INTRODUCTION

The compound UBe_{13} (Ref. 1) belongs to a small class of heavy-electron superconductors with superconducting critical temperatures $T_c \lesssim 1$ K that includes $CeCu_2Si_2$,[2] UPt_3,[3] and URu_2Si_2.[4–6] The compounds have enormous electron effective mass m^*, derived from the zero-temperature limit of the electronic specific heat $C_e(T)$ coefficient $\gamma(T)$ [$\equiv C_e(T)/T$], which attain values as high as several hundred times the free-electron mass for $CeCu_2Si_2$ (Ref. 7) and UBe_{13}. The occurrence of superconductivity in the presence of strong electron-electron interactions, inferred from the large values of m^* and the paramagnetic susceptibility, as well as other physical properties, has led to the speculation that these materials exhibit an unconventional type of superconductivity, analogous to the triplet superfluidity displayed by liquid ^{3}He below ~2 mK.[8,9] Power-law T dependences of various superconducting-state physical properties suggest that the superconductivity of these materials is anisotropic, with the superconducting energy gap vanishing at points or lines on the Fermi surface.[8,9] In UBe_{13} doped with Th (Ref. 9) or UPt_3 subjected to applied magnetic fields,[10–12] there is evidence for the existence of more than one superconducting phase.

The application of high pressure P to heavy-electron superconducting compounds has proven to be a useful technique for obtaining information about the normal and superconducting states in these remarkable materials. For example, a study of $\rho(T,P)$ in the range $0 \le P \lesssim 20$ kbar for the compound URu_2Si_2, in which coexisting superconducting and spin-density-wave states apparently form over different parts of the Fermi surface, is consistent with an increase with increasing pressure of the area of the Fermi surface that is "gapped" by the spin-density wave.[6,13] Measurements of $T_c(P)$ in the range $0 \le P \lesssim 12$ kbar on the $U_{1-x}Th_xBe_{13}$ system[14] have yielded compelling evidence for the existence of two distinct superconducting states, one in the range $0 \le x \lesssim 0.017$ and the other for $x \gtrsim 0.017$ at atmospheric pressure. An investigation of $\rho(T,P)$ in the UBe_{13} system in the range $0 \le P \lesssim 20$ kbar indicates that both the Fermi-liquid energy scale T^* derived from the coefficient of the T^2 term of the electrical resistivity and the temperature T_{max} of the maximum in $\rho(T)$ increases with pressure.[15]

In this investigation, we have extended the measurements of $\rho(T,P)$ on UBe_{13} to pressures over 150 kbar. A preliminary report of the $\rho(T,P)$ data appears in Ref. 16. The measurements reveal that T_{max} increases linearly with P at two limiting rates: 0.25 K/kbar below ~30 kbar and 0.65 K/kbar above ~30 kbar. The Fermi-liquid parameter T^* exhibits a similar linear increase with P with a crossover at about 50 kbar. The similarity in the pressure dependences of T^* and T_{max} indicates that both parameters are proportional to a single energy scale, while the changes in pressure dependence suggest the occurrence of a crossover in the range ~30–50 kbar, possibly from heavy-fermion to intermediate-valence behavior.

EXPERIMENTAL DETAILS

The polycrystalline UBe_{13} sample was prepared by arc-melting high-purity elements together on a water-cooled copper hearth in a Zr-gettered argon atmosphere as has been previously described.[1] The quality of the material is reflected in its inductively measured high $T_c = 0.905$ K and very narrow transition width $\Delta T_c = 0.030$ K. Electrical resistivity measurements under pressure were made in a self-clamping Bridgman anvil device in a ^{4}He cryostat for temperatures between 1 and 300 K. The anvils were made of tungsten carbide and were degaussed prior to use. The sample was a small single bulk piece of UBe_{13}, which was mounted electrically in series with a piece of Pb to serve as a manometer.[17] Platinum leads were attached to the sample and Pb in four-lead configurations using silver-loaded epoxy. The sample was mounted between a pair of steatite disks with the voids around leads and sample filled with talc. The gasket was made of pyrophyllite.

The pressure was inferred from the resistivity measured T_c of the superconducting Pb manometer.[17] Electrical resistance measurements were made using a standard four-lead and lock-in detection ac technique at 220 Hz.[18] The temperature was determined from a calibrated carbon glass resistance thermometer for temperatures above ~7 K and from a calibrated Ge resistance thermometer for temperatures below ~7 K. On the basis of a room-temperature electrical resistivity value of 107 $\mu\Omega$ cm for UBe_{13}, a geometrical factor was calculated and used to convert the measured resistances to resistivities.

RESULTS

Measurements of $\rho(T)$ between 1 and 300 K were made at nine different pressures ranging from 12 to 152 kbar. The $\rho(T)$ curves for eight of these pressure are plotted in Fig. 1. The curve at 12 kbar is similar to previously reported curves at pressure;[15] in particular, the shoulder at 20 K seen in the atmospheric pressure data[1] is no longer present. The general behavior is essentially unchanged with pressure, with ρ increasing as T decreases from 300 K, attaining a maximum, and then decreasing rapidly prior to the abrupt drop to zero at the superconducting transition.

It is evident from the shapes of the $\rho(T)$ curves in Fig. 1 that pressure has a fairly strong effect, shifting the maximum in the curves to higher temperatures. A plot of T_{max}, the temperature at which $\rho(T)$ attains a maximum, versus pressure is shown in Fig. 2. The plot reveals that

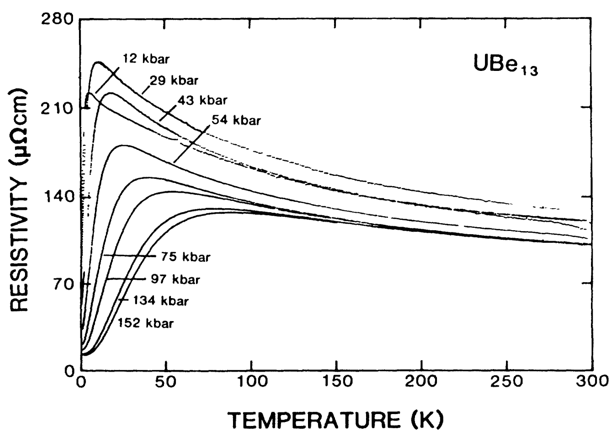

FIG. 1. Electrical resistivity ρ vs temperature T of UBe_{13} at pressures of 12, 29, 43, 54, 75, 97, 134, and 152 kbar.

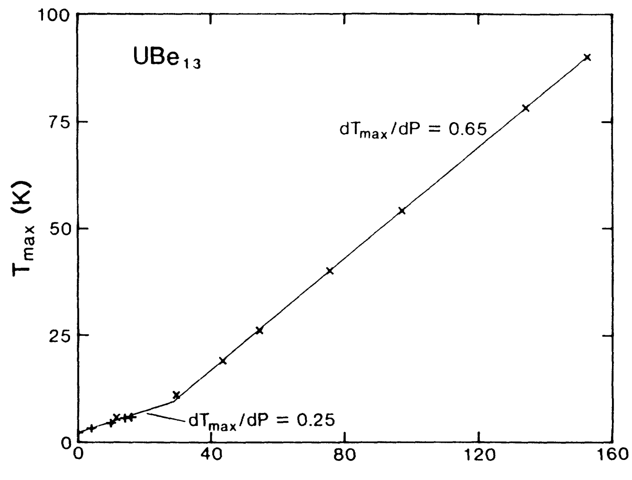

FIG. 2. Temperature of the resistance maximum, T_{max}, in UBe_{13} as a function of applied pressure P. The lines correspond to the limiting slopes at high and low pressures. The points plotted with "+" symbols are from Ref. 16.

T_{max} increases linearly with pressure at two limiting rates, 0.25 K/kbar below ~30 kbar and 0.65 K/kbar above ~30 kbar. X-ray-diffraction measurements under pressure to 450 kbar showed no evidence for a structural phase transition in this pressure range.[19]

At the higher pressures, there is evidence of a change in the behavior of $\rho(T)$ at low temperature. Plots of ρ

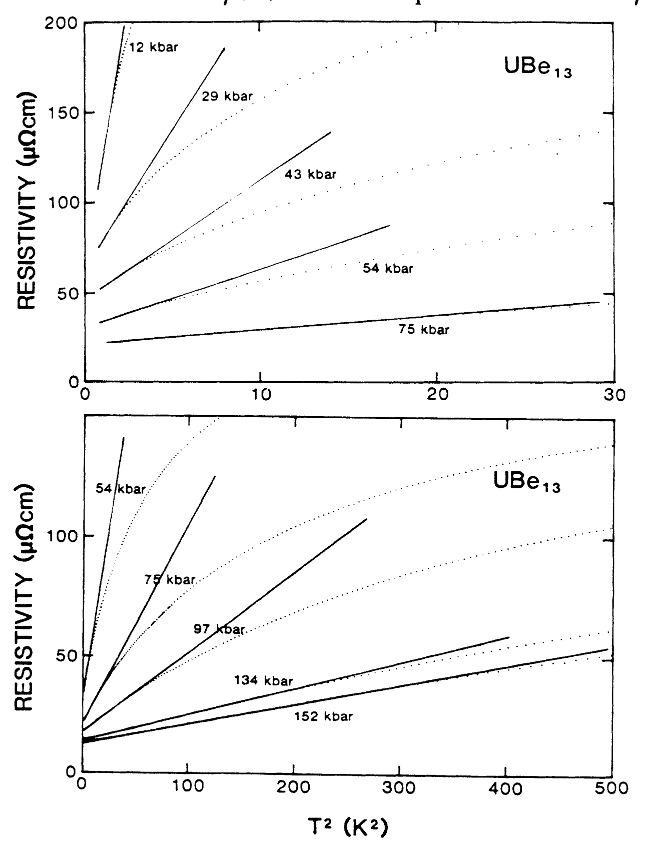

FIG. 3. Electrical resistivity ρ as a function of temperature squared T^2 for UBe_{13} at pressures of 12, 29, 43, 54, 75, 97, 134, and 152 kbar.

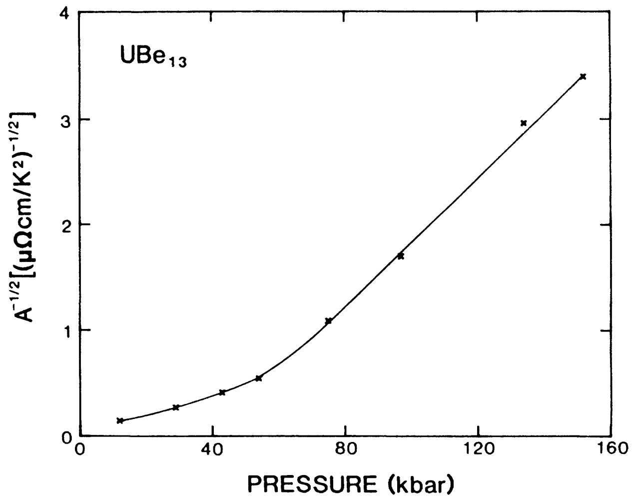

FIG. 4. $A^{-1/2}$ vs pressure for UBe_{13}, where A is the coefficient of the T^2 term of the low-temperature electrical resistivity. The line is a guide to the eye.

versus T^2 in Fig. 3 show that the temperature range over which the low-temperature $\rho(T)$ data can be described by a quadratic temperature dependence increases dramatically with pressure from a little over 1 K at 12 kbar to more than 16.5 K at 152 kbar. Values of the slope $d\rho/d(T^2)=A$ were determined from fits to the low-temperature $\rho(T)$ and data used to construct the $A^{-1/2}$-versus-P plot is displayed in Fig. 4. Similar to the T_{max}-versus-P data in Fig. 2, there are two regimes in which $A^{-1/2}$ depends linearly on P, one below ~50 kbar and another above ~50 kbar, with $A^{-1/2}(P)$ having a less distinct crossover.

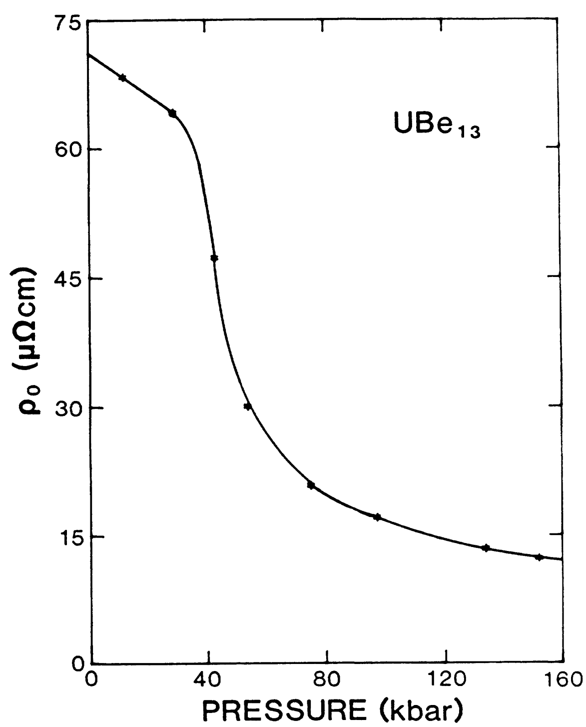

FIG. 5. The extrapolated $T=0$ K intercept, ρ_0, of the UBe_{13} $\rho(T)$ curves in Fig. 1.

Additional effects of pressure on $\rho(T)$ are evident in Fig. 1. The magnitude of ρ near and above T_{max} increases with pressure below ~30 kbar, as previously observed,[15] but decreases with pressure above ~30 kbar. At the highest pressures, $\rho(T)$ appears to approach a limiting behavior at high temperature; at pressures above 75 kbar, the $\rho(T)$ maximum continues to broaden while the magnitude of ρ at 300 K remains essentially constant. Plotted in Fig. 5 are the extrapolated 0-K intercepts, ρ_0, of the $\rho(T)$ curves which decrease monotonically with pressure.

DISCUSSION

The general shapes of the $\rho(T)$ curves in Fig. 1 are characteristic of intermediate-valence and heavy-fermion behavior displayed by many rare-earth and actinide compounds.[20,21] At high temperatures, the large magnitude of ρ and the increase in ρ with decreasing T are consistent with a lattice of independent single-ion Kondo scattering sites.[20] At low temperature, there is a rapid decrease of ρ with decreasing T that is generally attributed to increasing coherence between the Kondo scattering sites, which leads to the formation of the low-temperature Fermi-liquid ground state.[8]

The low-T downturn in $\rho(T)$ may also be attributed, in part, to the splitting of the U $5f$-electron energy levels by the crystalline electric field (CEF). This explanation was first given by two of the authors[22] and developed further by Rao and Wallace[23] for the compound $CeAl_2$ in which the low-T downturn in $\rho(T)$ was ascribed to a decrease in magnetic scattering due to the depopulation of the Ce CEF-split $4f$-electron energy levels as the temperature was lowered. In ambient pressure $\rho(T)$ data for UBe_{13}, a broadened peak can be seen at about 20 K.[1] A similar feature was observed in $CeCu_2Si_2$ which was interpreted by Horn *et al.* as resulting from the onset of additional scattering associated with an excited Ce CEF-split $4f$-electron energy level as the temperature was increased.[24] The presence of zero-field splittings (ZFS) (e.g., CEF and spin-orbit effects) of the U $5f$-electron energy levels in UBe_{13} is supported by the high-temperature specific-heat data of Felton *et al.*[25] who concluded that the U ions have a $5f^3$ configuration with a Γ_6 ground state, a $\Gamma_8^{(1)}$ first excited state at 180 K, and a $\Gamma_8^{(2)}$ excited state at higher energy. The ZFS of the U $5f$-levels may determine the pressure where a possible crossover out of the heavy fermion regime takes place.[26]

The plots in Fig. 3 reveal the T^2 dependence of $\rho(T)$ characteristic of low-temperature Fermi-liquid behavior. Fermi-liquid theories relate the T^2 coefficient of resistivity A to the inverse square of a characteristic temperature T^* [$A \propto 1/(T^*)^2=(A_0^a/T_F)^2$, where A_0^2 is the singlet forward-scattering amplitude] that sets the energy scale for the Fermi-liquid behavior.[8] Since $A^{-1/2} \propto T^*$, a plot of $A^{-1/2}$ versus P is shown in Fig. 4. Similar to $T_{max}(P)$, there are two limiting rates of increase of $A^{-1/2}$ with P, with a crossover in the vicinity of 50 kbar. The plot of $A^{-1/2}$ versus T_{max} in Fig. 6 shows that these two quantities are linearly related to one another and are probably proportional to a single energy scale in both of the two

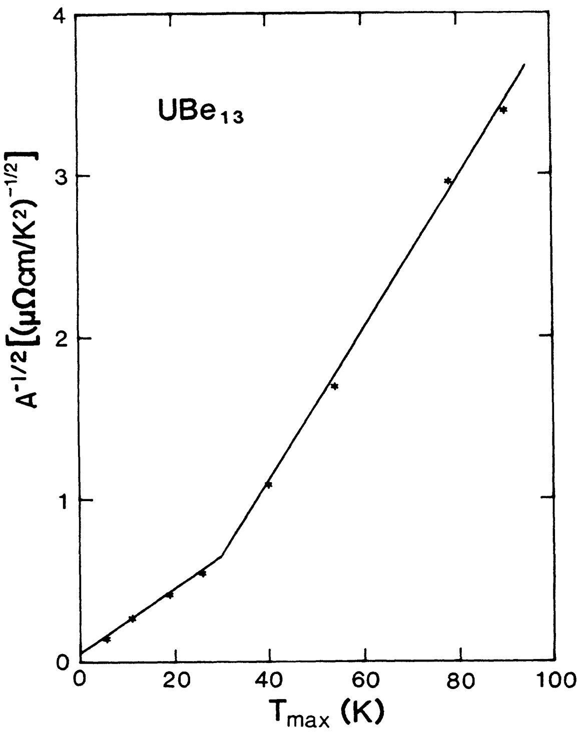

FIG. 6. $A^{-1/2}$ vs T_{max} for UBe_{13}. Lines correspond to the limiting slopes at high and low pressures.

regimes, although by different constants of proportionality. It is interesting to note that the high-temperature limits of T^2 behavior under pressure are $(15\pm3)\%$ of the corresponding Kondo temperature T_K values extracted from $A^{-1/2}$ values. Here, T_K at low pressures ($0 \leq P \lesssim 10$ kbar) was estimated from the relation $\gamma(0)=0.68R/T_K$,[27] where $\gamma(0)$ is the zero-temperature value of the electronic specific-heat coefficient and R is the universal gas constant, while T_K at high pressures ($P \gtrsim 10$ kbar) was inferred from $T_K \approx CA^{-1/2}$ (see discussion below), where the scaling constant C was determined from the relation between the values of T_K and $A^{-1/2}$ at low pressures ($0 \leq P \lesssim 10$ kbar). In a recent magnetoresistance study on UBe_{13} in the range $0<P<95$ kbar, there was a change in sign of the magnetoresistance at pressures above 50 kbar, providing further evidence for the formation of a coherent ground state describable as a Fermi liquid.[28]

The decrease in ρ_0 with increasing P seen in Fig. 5 indicates the presence of magnetic scattering that has not been fully suppressed by the coherence formation at the lowest temperatures. Further evidence for this is provided by the H dependence of ρ_0 which was shown to approach a limiting value of ~ 18 $\mu\Omega$ cm with either applied H or applied H and P.[29] The reduction in ρ_0 and slope $d\rho_0/dP$ at pressures above the identified crossover suggests that the high-pressure regime is considerably less magnetic than the low-pressure regime. Pressure-dependent changes in the magnitude of $\rho(T)$ for the heavy-electron superconductor $CeCu_2Si_2$ have previously been attributed to changes in the geometrical factor.[30] Changes in geometrical factor for the bulk UBe_{13} specimen studied in this work are too small to account for the observed changes in the magnitude of $\rho(T)$.

Many of the attributes characteristic of dilute paramagnetic impurity (Kondo) systems appear, in some cases, to persist in lattices of paramagnetic ions (Kondo lattices). For dilute impurity systems, T_K is given by $T_K \cong T_F \exp[-1/N(E_F)|J|]$ where T_F is the Fermi temperature, $N(E_F)$ is the density of states at the Fermi level, and J is the exchange coefficient.[31] The exchange coefficient J can, in turn, be approximated as $J \cong -\langle V_{kf}^2 \rangle/\varepsilon_f$,[31,32] where V_{kf} is the matrix element mixing localized f-electron states and conduction electron states and ε_f is the energy separating the f-electron state f^n and E_F. The states f^n and f^{n-1} are separated by the correlation energy U with f^{n-1} an energy $U-\varepsilon_f$ above E_F. Hybridization with the condition electrons broadens the f-electron states to a width $\Delta=\pi\langle V_{kf}^2 \rangle N(E_F)$.[33] The Kondo effect is characterized by a transition from a perturbative conduction electron-localized electron interaction ($T \gg T_K$) to a strong interaction with the formation of a quasibound state ($T \leq T_K$).[34] Associated with this transition is the formation of a many-body resonance in the density of states near E_F, the Abrikosov-Suhl resonance (ARS), which has an amplitude and a width of $(\pi\Delta)^{-2}$ and T_K, respectively.[35,36] The location (energy) of the ASR is determined by the angular momentum j of the scattering channels available to the conduction electrons.[37] For a $j=\frac{1}{2}$ state, there is a single ASR centered at E_F, while for states where $j \neq \frac{1}{2}$, the associated ASR's are centered away from E_F. It is generally accepted that in small T_K Kondo lattice systems, the large amplitude of the ASR results in the characteristic energy scale being determined by the width of the ASR. It follows that the characteristic energy scale is T_K, which is proportional to $A^{-1/2}$ and, therefore, also to T_{max}. The differences in the linear relationships between $A^{-1/2}$ and T_{max} seen in Fig. 6 above and below the crossover result from the different constants relating $A^{-1/2}$ and T_{max} to T_K.

One effect of pressure is to destabilize the larger ion, which in the case of f-electron systems will stabilize f^{n-1} in favor of f^n.[20,38] Therefore, pressure will increase the energy of the f^n state, resulting in a decrease in ε_f. In f-electron systems, pressure has been shown to produce large increases in V_{kf} and small decreases in the $N(E_F)$, with the net result being a rapid increase in T_K.[38,39] Such effects are consistent with experiments on UBe_{13} where strong reductions in the electronic specific-heat coefficient γ (30% at 8 kbar) (Ref. 27) and magnetic susceptibility χ (8% at 8 kbar) (Ref. 40) and rapid increases in T_{max} and $A^{-1/2}$ as a function of applied pressure have been observed. The application of sufficiently high pressure can also result in an intermediate-valence state in which there are temporal fluctuations between the configurations f^n and f^{n-1} and the width of the resonance at E_F can be related to the fluctuation time.[20] The two limiting rates of increase of T_K with P, along with the apparent reduction in the moment above the crossover, discussed earlier, suggest that UBe_{13} undergoes a transition from a heavy-fermion system (HFS) to an intermediate-valence system (IVS) in the range ~30–50 kbar.

An alternative interpretation considers a HFS and an IVS to be limiting regimes in which a HFS and an IVS correspond, respectively, to T_K smaller than and larger than the f-level ZFS.[26] The general behavior of both the HFS and IVS can be ascribed to a single temperature scale T_K associated with a many-body resonance at or near E_F: the HFS with a single $j=\frac{1}{2}$ ASR at E_F and the IVS with an effective resonance resulting from the combination of more than one ASR at or near E_F. It follows that a pressure-induced increase in T_K could result in a crossover from HFS ($T_K<$ZFS) to IVS ($T_K>$ZFS). In addition, the number of scattering channels will increase as the system crosses into the IVS regime and there will be a shift in E_F through the maximum of the ASR in the HFS to a position well below the ASR in the IVS, resulting in a reduction in $N(E_F)$. This may account for the pressure dependence observed for the magnitude of $\rho(T)$. The IVS would be characterized by an effective resonance that should broaden more rapidly with P than the single $j=\frac{1}{2}$ ASR of the HFS. In this characterization of the crossover, the reduction in moment inferred from the decrease in ρ_0 at high pressure follows from the Kondo ground state developing from an ion with several populated zero-field split energy levels.

In summary, pressure-induced crossovers in $d\rho_0/dP$, dT_{max}/dP, and dT^*/dP in the pressure range ~25 to ~55 kbar from one regime to another are evident in the $\rho(T,P)$ data for UBe_{13}. While the lower-pressure regime clearly corresponds to a HFS, we tentatively identify the higher-pressure regime with an IVS. One possible interpretation of the crossover is that the increase in T_K with P inferred from the $\rho(T,P)$ data in the low-pressure regime moves the system from a regime where $T_K<$ZFS (HFS) to one in which $T_K>$ZFS (IVS). The energy scale set by T_K appears to be the only one necessary to account for the behavior observed in both regimes.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy, Division of Materials Science of the Office of Basic Energy Sciences. The research at the University of California at San Diego (USCD) was supported by the U.S. Department of Energy under Grant No. DE-FG03-86ER45230. Two of us (M.B.M. and J.L.S.) would like to thank the Aspen Center for Physics (Aspen, CO), where contributions to writing this paper were made, for their hospitality. We would like to acknowledge D. M. Newns, T. Penney, K. S. Bedell, B. H. Brandow, S. von Molnar, and S. Schiferl for valuable discussions, and W. L. Hults for technical assistance.

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