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Authors

Balicas, L
Nakatsuji, S
Lee, H
et al.

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Magnetic field-tuned quantum critical point in $CeAuSb_2$

L. Balicas,[1] S. Nakatsuji,[2] H. Lee,[3] P. Schlottmann,[1] T. P. Murphy,[1] and Z. Fisk[3]
[1]*National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32306, USA*
[2]*Department of Physics, University of Kyoto, Kyoto 606-8502, Japan*
[3]*Department of Physics, University of California Davis, Davis, California 95616, USA*

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Transport, magnetic, and thermal properties at high magnetic fields (H) and low temperatures (T) of the heavy-fermion compound $CeAuSb_2$ are reported. At $H=0$ this layered system exhibits antiferromagnetic order below $T_N=6$ K. Applying B along the interplane direction, leads to a continuous suppression of T_N and a quantum critical point at $H_c \simeq 5.4$ T. Although it exhibits Fermi-liquid behavior within the Néel phase, in the paramagnetic state the fluctuations associated with H_c give rise to unconventional behavior in the resistivity (sublinear in T) and to a $T \ln T$ dependence in the magnetic contribution to the specific heat. For $H > H_c$ and low T the electrical resistivity exhibits an unusual T^3 dependence.

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I. INTRODUCTION

Quantum criticality[1] is common to a large variety of very different phenomena ranging from low-dimensional quantum systems to high-temperature superconductivity, disorder-induced criticality (e.g., Griffiths phase), and heavy-fermion compounds at the verge of antiferromagnetic (AF) order. For strongly correlated electrons a quantum critical point (QCP) is obtained when either (i) the long-range order is suppressed to $T=0$ (second-order phase transition) or (ii) the critical end-point terminating a line of first-order transitions is depressed to $T=0$.[2] A QCP can be tuned by an external variable, such as pressure, chemical composition or the magnetic field H.[3] H is an ideal control parameter, since it can be reversibly and continuously tuned towards the QCP.[4] In alloys the disorder driven effects cannot be separated from the quantum criticality of the translational invariant system.[4] Hence, it is essential to consider stoichiometric systems. Two compounds with field-tuned QCP, $YbRh_2Si_2$ and $Sr_3Ru_2O_7$, reached prominence due to the non-Fermi-liquid (NFL) behavior triggered by the quantum fluctuations associated with the QCP. In this manuscript we present a Ce compound $CeAuSb_2$ exhibiting a field-tuned QCP with unusual transport and thermodynamic properties. $YbRh_2Si_2, Sr_3Ru_2O_7$ and $CeAuSb_2$ have a field-tuned QCP as a common thread, yet their properties are considerably different. This points towards lack of universality among different systems as a fundamental component of quantum criticality.

In zero field $YbRh_2Si_2$ exhibits a second-order phase transition into an AF state at $T_N=70$ mK.[5] A field applied along the inter-plane direction drives T_N to zero at $H_c \simeq 0.66$ T, leading to NFL behavior, i.e., a logarithmic increase of $C_e(T)/T$ and a quasilinear T dependence of the electrical resistivity ρ below 10 K.[6] Above H_c Fermi-liquid (FL) behavior is recovered ($\rho \propto AT^2$ and constant Sommerfeld coefficient γ), with $A(H)$ and $\gamma(H)^2$ displaying a $1/(H-H_c)$ divergence as $H \rightarrow H_c$.[7] A similar trend was recently found in YbAgGe.[8]

Field-tuned anomalous metallic behavior in the vicinity of metamagnetism (MM) was studied in detail in $Sr_3Ru_2O_7$.[9] At a MM transition the magnetization M increases rapidly over a narrow range of fields. The transition is of first order, since there is no broken symmetry involved, and terminates in a critical "end" point $(H^\star, T^\star)$.[2] In the MM transition of $Sr_3Ru_2O_7$, $T^\star$ decreases continuously as H is rotated towards the interplane c axis,[10] thus yielding a QCP in a first order transition.[2,9] This scenario is supported by the T-linear dependence in ρ,[9] a divergence of the coefficient A of ρ,[9] the enhancement of the effective mass of the quasiparticles, and the $\ln T$ dependence of the specific heat γ.[11] Remarkably, at very low T and very close to the critical field H_c, ρ displayed unusual behavior, first reported as a T^3 dependence,[9] and then for much purer samples as an upturn.[10] The QCP of a MM transition is also believed to cause the rich phase diagram of URu_2Si_2 at high fields.[12]

Among stoichiometric Ce compounds, so far there is evidence for a field-tuned QCP only in $CeCoIn_5$ (Ref. 13) and $CeIrIn_5$.[14] Here the QCP is believed to give rise to a (possibly unconventional) superconducting (SC) phase, in addition to NFL behavior. The nature of the magnetic correlations at low T is still unclear for both compounds.

In this paper, we report on anomalous properties of the tetragonal metallic compound $CeAuSb_2$, which at $H=0$ orders AF (Ref. 15) with $T_N=6.0$ K.[16] For $T<T_N$, $\rho(T)$ has the typical AT^2 dependence of a FL and extrapolating C_e/T to $T=0$ yields $\gamma \sim 0.1$ J/mol K^2. Hence, $CeAuSb_2$ can be considered a system of relatively light heavy fermions. Above T_N, on the other hand, $\rho(T)$ displays a T^α dependence with $\alpha \lesssim 1$ and, C_e/T has a $-\ln T$ dependence, both characteristic of NFL behavior due to a nearby QCP. A magnetic field along the interplane direction leads to two subsequent MM transitions and the concomitant *continuous* suppression of T_N to $T=0$ at $H_c=5.3\pm0.2$ T. As the AF phase boundary is approached from the paramagnetic (PM) phase, γ is enhanced and the A coefficient of the resistivity diverges as $(H-H_c)^{-1}$. When T is lowered for $H \sim H_c$, the T dependence of ρ is sublinear and the one of C_e/T is approximately $-\ln T$. At higher fields, $H \gg H_c$, an unconventional T^3 dependence emerges in ρ and becomes more prominent as H increases.

These observations suggest the existence of a field-induced QCP at H_c. For small fields the AFM to paramagnetic transition at 6 K has the characteristics of a second-order phase transition. This is evident, e.g., in the T dependence of the resistivity and the discontinuity of the specific heat. At lower T, the magnetization as a function of field displays two MM transitions, which become more pronounced, i.e., more steplike, as T is decreased. This is an indication that the transition acquires a first-order character when H_c is approached. This is expected, since in a magnetic field there is no symmetry breaking. However, a first-order transition is not fully developed at 1.5 K, because (i) the magnetization still varies continuously with field, (ii) the peak of the transition in the specific heat disappears continuously, and (iii) the A and A_3 coefficients of the resistivity vary as $C/(H-H_c)$ when the transition is approached for $H>H_c$. In addition no hysteresis is observed in the resistivity down to 0.7 K. The above seems to indicate that the system is close to a QCP. However, the resistivity measured at 22 mK on a different sample (with $T_N \approx 6.5$ K) showed small hysteresis between the two MM transitions. Hence, the most likely scenario is then that at very low T the transition is weakly first order and the system is very close to a QCP.

II. EXPERIMENTAL

Single crystals of CeAuSb$_2$ were grown by the self-flux method, as described in Ref. 17, using high-purity starting materials (3N-Ce, 5N-Au, and 5N-Sb) with excess Sb as a flux. Microprobe analysis indicated a single stoichiometric phase. Electrical ac transport was measured with the Lock-In technique in both, Bitter and superconducting magnets, coupled to ^{3}He and dilution refrigerators, respectively. The magnetization as a function of T and H was obtained in a Quantum Design dc SQUID, as well as with a high field vibrating sample magnetometer. The heat capacity was measured down to ^{3}He temperatures in a Quantum Design PPMS using the relaxation time technique. The $C(T)$ measurements were extended to higher T showing a Schottky-like peak centered around $T \simeq 50$ K. This suggests an excitation energy of $\Delta = 110$ K between the ground and first excited doublets.

III. RESULTS AND DISCUSSION

Figure 1(a) displays the in-plane electrical resistivity ρ as function of T down to $T=0.5$ K for $H=0$ T. The vertical arrow indicates the onset of AF order, below which a T^2 dependence of ρ is obtained. Above T_N a NFL-like T^α dependence with $\alpha \lesssim 1$ is observed. As H increases the AF order is gradually suppressed and in the vicinity of the AF to PM phase boundary a linear dependence on T emerges [see Fig. 1(b)]. At higher fields FL behavior, i.e., $\rho = \rho_0 + AT^2$, is found over a limited range of T as indicated in Fig. 1(c) by the vertical arrows. The red lines are least square fits to the T^2 dependence, which show that A decreases as H increases. At very low T and very high H, ρ displays an anomalous T^3 dependence [see Fig. 1(d)], suggesting an unusual scattering mechanism and that the T^2 dependence may just be a crossover regime between T^α with $\alpha < 1$ and the T^3 regions.

The H dependence of ρ sheds more light on the role of the magnetic fluctuations (see upper panel of Fig. 2). For

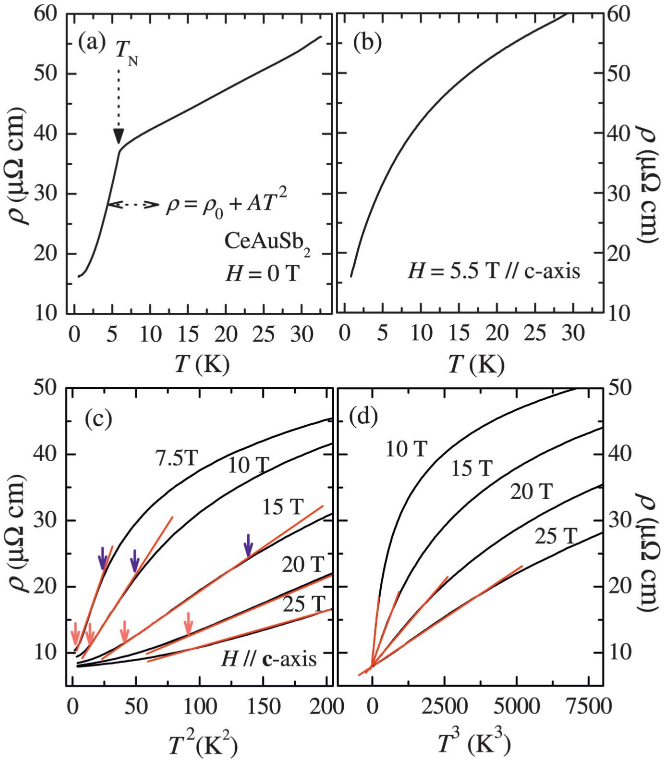

FIG. 1. (Color) (a) T dependence of the in-plane resistivity ρ of a CeAuSb$_2$ single crystal down to $T=0.5$ K. At T_N a pronounced change in slope is observed, below which FL behavior is recovered. (b) This behavior is suppressed and a sublinear T dependence is observed when $T_N \rightarrow 0$. (c) For $H>5$ T a T^2 dependence is obtained but only over a limited range of T as indicated by the vertical arrows (red lines are linear fits to a T^2 dependence). (d) For $H>6$ T a T^3 dependence is observed at lower T (red lines are linear fits to a T^3 dependence).

$T>6$ K, a crossover from positive to negative magnetoresistance is seen in $\rho(H)$ at $H \sim 5$ T. As T is lowered below 6 K, two MM transitions emerge, with negative magnetoresistance developing at the AF to PM transition. The magnetic field suppresses the AF spin correlations, giving rise to a gradual alignment of the spins and reducing this way the spin-flip scattering. The anomalous T^3 dependence in $\rho(H)$ is believed to be related to the alignment of the Ce spins. As seen below, the saturation of the magnetization m suggests that the system is nearly half metallic for very large H.

The middle panel of Fig. 2 shows the phase diagram resulting from transport and magnetization measurements. The blue triangles indicate the boundary between the PM and the AF phases for $H \| c$ axis, while the green ones define the phase boundary for H in the a-b plane. Notice the remarkable anisotropy. Surprisingly no hysteresis was detected down to 0.7 K, neither in T nor in H scans, despite the abrupt changes in the slope of $\rho(H)$. This suggests that the transition between the PM and AF phase is a weak first order one or unexpectedly of second order. The first MM transition at $H_{\mathrm{MM}} \simeq 2.8$ T, which could correspond to a spin-flop transition, is indicated with open circles.

In Fig. 3 the field scan at 22 mK of the resistivity of a different sample with higher T_c is shown. Contrary to the

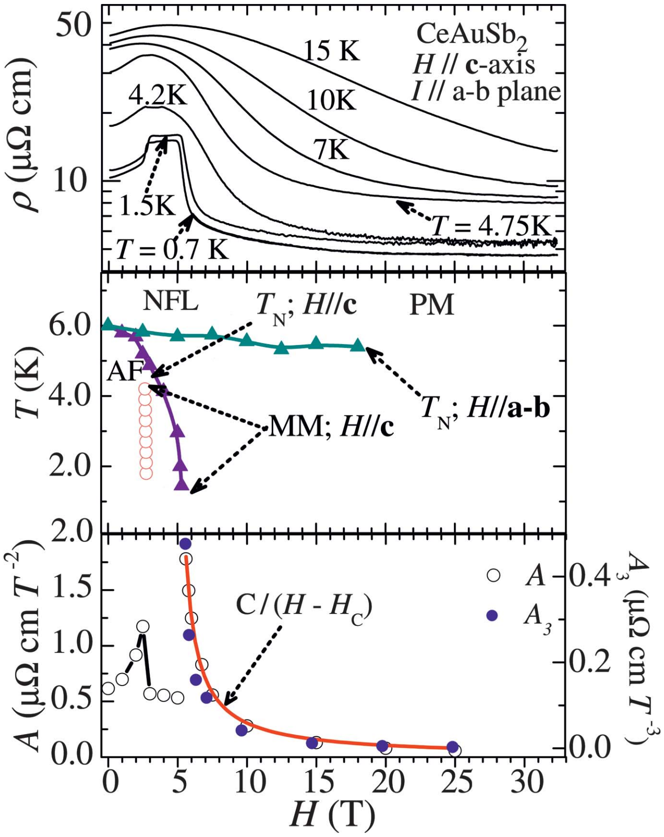

FIG. 2. (Color) Upper panel: In-plane resistivity ρ of CeAuSb$_2$ as a function of H up to H=32.5 T. Middle panel: The resulting phase diagram indicating the AF phase boundary, the PM region, and the region in field where NFL behavior is found. Arrows indicate the critical fields of the metamagnetic transitions. Note that the effect of H on T_N is very anisotropic. Lower panel: Coefficients A (open circles) and A_3 (blue circles) of the resistivity as a function of H. See text for details.

field scans presented in the upper panel of Fig. 2, which are at a slightly higher T, this scan displays a moderate hysteresis. We have to conclude from this, in addition to a possible slight sample dependence, that at very low T the system has a weak first order transition. In summary, CeAuSb$_2$ is very close to a QCP.

The field dependence of the A and A_3 coefficients (prefactors of the T^2 and T^3 terms) of the resistivity is displayed in the lower panel of Fig. 2. Both coefficients diverge as $H \rightarrow H_c$ and the red line is a fit to $C/(H-H_c)$. The same dependence was reported for YbRh$_2$Si$_2$ (Ref. 6) and interpreted as a dramatic increase of the quasiparticles linewidth throughout the entire Fermi surface. A similar increase of the A coefficient was perceived as evidence for a QCP in Sr$_3$Ru$_2$O$_7$.[9] The A coefficient also increases at the first MM transition due to an enhancement in the scattering of the quasiparticles.

The magnetic moment m of a CeAuSb$_2$ single crystal divided by the field is shown in Fig. 4(a) as a function of T down to 1.8 K. For small fields and higher T, m/H follows a Curie-Weiss law, yielding an antiferromagnetic Weiss-temperature θ_C=12.25 K and an effective moment p_{eff} $\approx 2.26\mu_B$, for the field applied along the c axis. Notice that the AF transition is observed only for $H \| c$, which is the magnetic easy axis. At higher fields, m/H saturates, suggesting the complete polarization of the Ce spins. In Fig. 4(b) both metamagnetic transitions are seen as steps in the magnetization m as a function of H. Both traces contain field-up and down sweeps measured in a SQUID magnetometer. Again, as for the resistivity, no hysteresis is observed for this sample and in this temperature range. The field of the first MM transition is only weakly T dependent. In contrast, as already seen in the resistivity data, the T_N for the second MM transition (AFM to PM) is markedly field dependent. Figure 4(c) shows m as a function of H (up to 30 T) at T=1.5 K. Above $H \sim 7$ T, m saturates at $m \simeq 1.65\mu_B$, a value that differs from the free ion moment of Ce due to crystalline electric field effects.

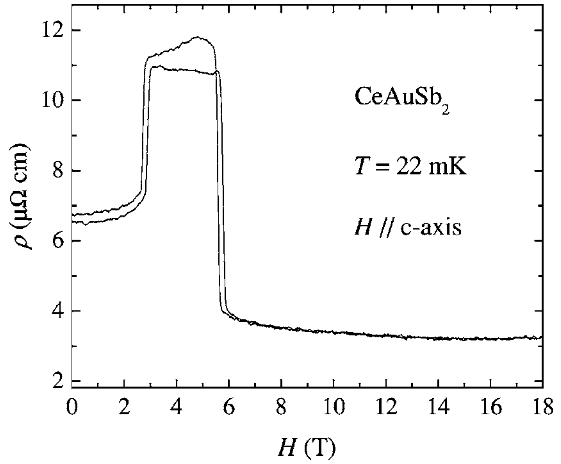

FIG. 3. Magnetic field scan of the resistivity at 22 mK on a different sample with $T_N \approx 6.5$ K. Between the two MM transitions a weak hysteresis is observed. The difference in the vertical lines for the increasing and decreasing field sweeps is likely due to the time constant of the instruments. The hysteresis is indicative of a weak first order character of the transition.

The upper panel of Fig. 5 shows $C(T)/T$ as a function of T for CeAuSb$_2$ at H=5 T, and for its isostructural nonmagnetic analog LaAuSb$_2$ at H=0 T. The large peak for CeAuSb$_2$ signals the AF transition. The subtraction of both curves yields the magnetic contribution to the heat capacity $C_e(T)/T$. For $3<T<20$ K, $C_e(T)/T$ displays a $-\ln(T)$ NFL-like dependence. The lower panel of Fig. 5 shows $C_e(T)/T$ as a function of T for several values of H. In the PM phase all curves collapse into a single trace, suggesting that the origin of the $\ln(T)$ dependence is the magnetic precritical fluctuations to the AF order. Furthermore, the effective mass of the quasiparticles, as given by $C_e(T)/T$ for $T \rightarrow 0$ increases considerably as $H \rightarrow H_c$, but remains finite. Note that because of the AFM order the effective mass cannot be defined precisely. In the inset we display $C_e(T)/T$ at T=1 K as a function of H. The $-\ln(T)$ dependence does not continue to very low T. This is similar to the behavior of the specific heat of Sr$_3$Ru$_2$O$_7$, where the $-\ln(T)$ dependence does not continue to very low T and there is a crossover to a constant C/T at the lowest T.[11]

Figure 6(a) shows the T^2 dependence of ρ down to $T \simeq 25$ mK for $H=5 \lesssim H_c$. There is evidence for a pro-

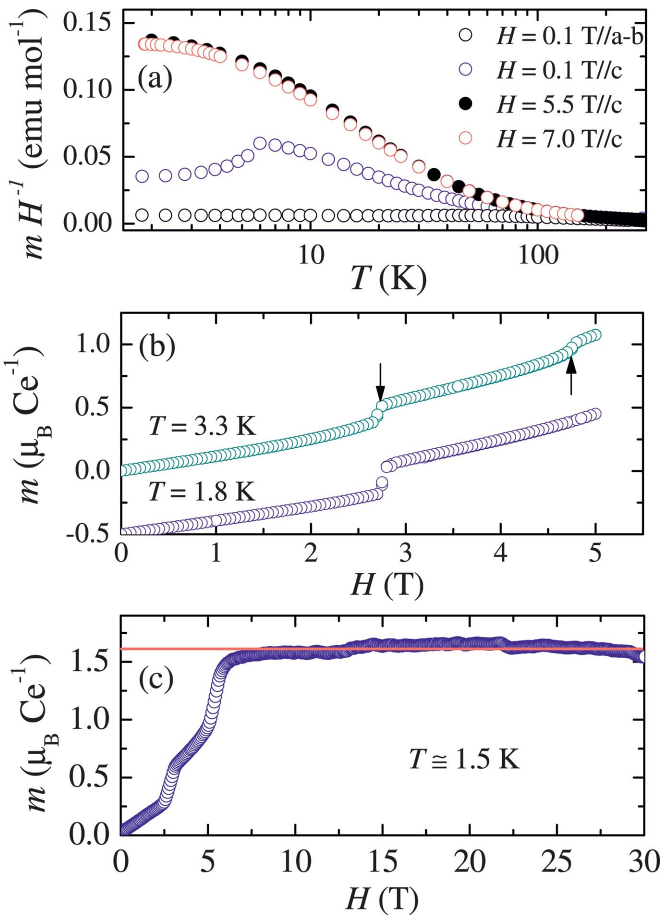

FIG. 4. (Color) (a) Magnetic moment m of $\mathrm{CeAuSb_2}$ divided by the field H, as a function of T for several values of H. (b) m of Ce, measured in a SQUID magnetometer, as a function of field H for $T=3.3$ and 1.8 K. (The vertical scale for the latter is off-set by -0.5.) Vertical arrows indicate the fields of both metamagnetic transitions. (c) m as a function of H at $T \simeq 1.5$ K measured with a vibrating sample magnetometer.

nounced reduction ρ at the lowest T, which could be induced by the weak first-order transition. In this case quantum criticality is not observed at the lowest T. Figure 6(b) depicts a qualitative sketch of the H-T phase diagram. It shows the dependence of the exponent $n \simeq \partial \ln[\rho(T)-\rho_0]/\partial \ln T$ on H and T. Here different values of ρ_0 were used in the PM and AF phases. The PM phase is indicated by the blue region which is influenced by the QCP leading to the anomalous NFL value $n \lesssim 1$. This is analogous to the behavior of $\mathrm{YbRh_2Si_2}$ (Ref. 7) and $\mathrm{Sr_3Ru_2O_7}$.[9] The FL state (in green) is recovered below the Néel temperature but is gradually suppressed as $H \rightarrow H_c$. As for $\mathrm{YbRh_2Si_2}$ (Refs. 6 and 7) a FL-like $n=2$ exponent is obtained above H_c but only over a limited range of T.

A spin-polarized metallic phase (in red) with an anomalous T^3 dependence is observed for $H > H_c$ at low T. A T^3 dependence at very low T and very close to the critical field H_c was also first reported in $\mathrm{Sr_3Ru_2O_7}$,[9] but then for much purer samples found to be an upturn.[10] Such dependence has also been predicted for half-metallic ferromagnets in terms of a one magnon scattering process.[18] This could be a plausible explanation for our system, which is spin polarized precisely in the region where $n=3$ is observed. Although $\mathrm{CeAuSb_2}$ is not a FM in zero field, it could have similar properties to a FM half-metal in a strong magnetic field. As seen in Fig. 1(d) at 25 T the T^3 dependence extends over one decade.

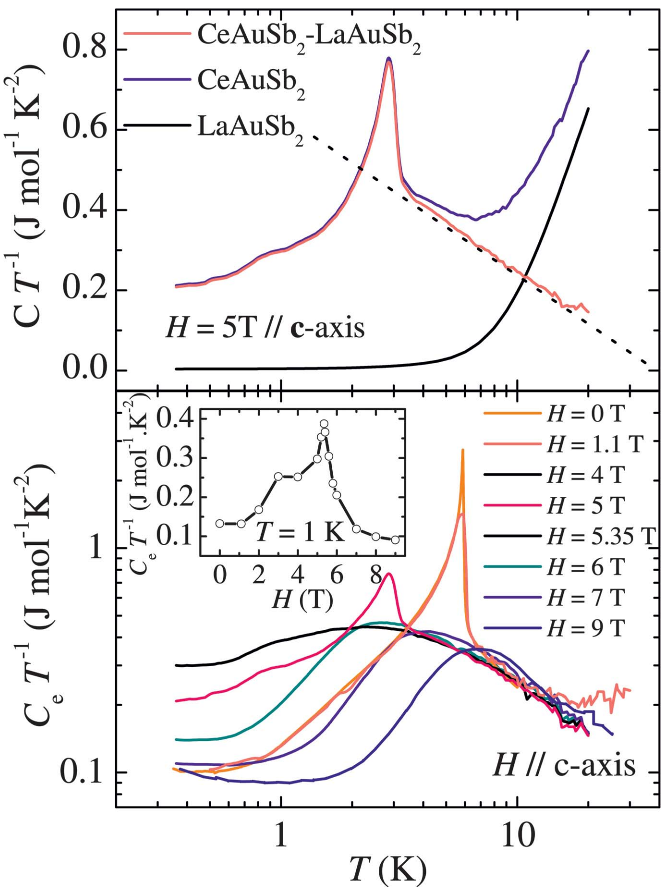

FIG. 5. (Color) Upper panel: Heat capacity divided by temperature C/T vs T down to 0.38 K for $\mathrm{CeAuSb_2}$ with $H=5$ T applied along the c axis (blue line), as well as for $\mathrm{LaAuSb_2}$ at $H=0$ T (black line). The difference is the magnetic contribution to the heat capacity C_e/T (in magenta), which shows a ln T dependence. Lower panel: C_e/T for several magnetic fields. All curves collapse into a single curve at high T and C_e/T at $T=1$ K increases as H approaches H_c (see inset).

IV. CONCLUSIONS

In summary, electrical transport, magnetization and thermal properties indicate that $\mathrm{CeAuSb_2}$ displays either a weak first-order or a second-order phase transition from a NFL-like PM metallic phase to a FL phase with long-range AF-order upon cooling at zero field. A magnetic field continuously reduces T_N, which vanishes for a field of $H_c=5.3\pm0.2$ T along the interlayer direction. The physical properties of the PM phase emerging at H_c are anomalous, i.e., $\rho(T) \propto T^{1/2}$, and $C(T)/T \propto -\ln(T)$, and are a strong indication that H_c corresponds to a field-tuned QCP (or at least is very close to a QCP). In addition, the A and A_3 coefficients of the resistivity diverge as $H \rightarrow H_c$. The consequences of the

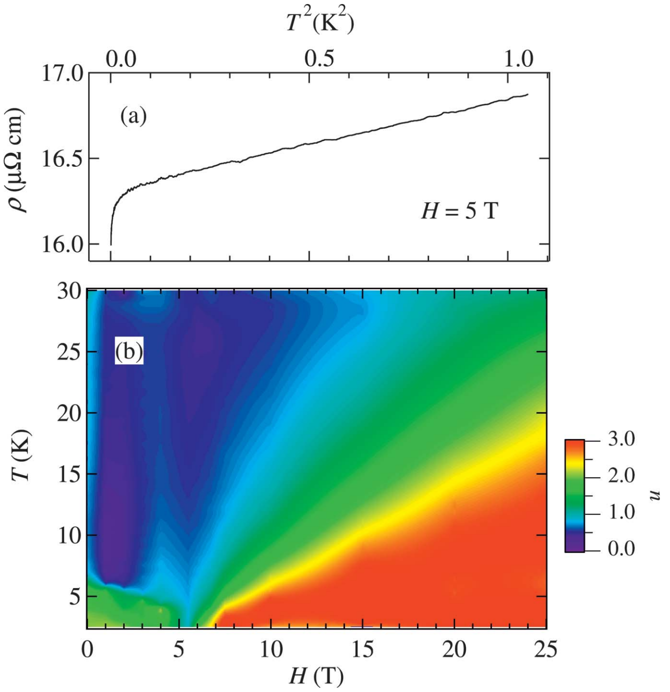

FIG. 6. (Color) (a) The T^2 dependence of ρ down to $T \simeq 25$ mK for $H = 5\text{T} \lesssim H_c$. Notice the pronounced deviation from the T^2 dependence at the lowest T, which is observed only the vicinity of H_c. b) The exponent n of $\rho(T)$ in the T-H plane.

QCP in $CeAuSb_2$ are different and perhaps more dramatic than for other field-tuned QCP systems. This is the case because the T^2 dependence of the resistivity is not recovered for H and T sufficiently far away from the QCP. The narrow region in Fig. 6 with $n=2$ should be considered a crossover region and not a FL phase.

Evidences for the existence of a field-induced QCP in $CeAuSb_2$ are the dramatic enhancement of C/T at low T's, the scaling of the resistivity A coefficient at the MM transition with field and the anomalous exponent of the T dependence of the resistivity. These were claimed to be solid evidence in favor of quantum-critical behavior in other compounds. However, a slight hysteresis is observed in one sample at 22 mK suggesting that the transition is weakly first order at very low T's. It is possible, that a modest tilt respect to the c axis may lead to a second-order transition down to the lowest temperatures, implying that the system effectively is very close to a QCP. The application of pressure in the other hand, which leads to an increase in the hybridization between itinerant and localized f electrons, is also expected to suppress the metamagnetic behavior leading to QC behavior. These hypothesis will be explored in great detail in future work. In conclusion, the field-tuned QCP systems represent a remarkable challenge from both the experimental and the theoretical perspectives, since the different compounds revealing some common aspects do *not* seem to belong to the same universality class.

ACKNOWLEDGMENTS

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[16] The present samples have lower residual resistivity, a higher T_N, more pronounced discontinuities of $M(H)$, and a sharper and higher heat capacity jump at T_N, as compared to those reported in Ref. 15. This strongly suggests that the present samples are of high quality.

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