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LETTER TO THE EDITOR

Huge residual resistivity in the quantum critical region of $CeAgSb_2$

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Abstract

We have studied the effect of pressure on the electrical resistivity of a high-quality single crystal $CeAgSb_2$ which has a small net ferromagnetic moment of 0.4 μ_B/Ce. The magnetic ordering temperature $T_{ord} = 9.7$ K decreases with increasing pressure p and disappears at a critical pressure $p_c \simeq 3.3$ GPa. The residual resistivity, which is close to zero up to 3 GPa, increases steeply above 3 GPa, reaching 55 $\mu\Omega$ cm at p_c. A huge residual resistivity is found to appear when the magnetic order disappears.

1. Introduction

The f electrons of cerium and uranium compounds exhibit a variety of characteristics including spin and valence fluctuations, heavy fermions, Kondo insulators and anisotropic superconductivity [1]. Recently a new aspect of these compounds with magnetic ordering has been discovered. When pressure p is applied to the cerium compounds with antiferromagnetic ordering such as $CeCu_2Ge_2$ [2], $CeIn_3$, $CePd_2Si_2$ [3] and $CeRhIn_5$ [4], the Néel temperature $T_N \rightarrow 0$ is reached at the quantum critical pressure p_c. Superconductivity and/or the non-Fermi liquid nature were observed around p_c. Very recently superconductivity was also found in a ferromagnetic state in UGe_2 [5] and URhGe [6]. The crossover from the magnetic ordered state to the non-magnetic state under pressure, crossing the quantum critical point, is the most interesting issue in strongly correlated f-electron systems.

$CeAgSb_2$ or more generally RTX_2 (R: rare earth, T: transition metal) and UTX_2, as discussed by Kaczorowski *et al* [7], crystallize in the tetragonal structure (space group

$P4/nmm$) [8, 9], which can be described as a filled UX_2 in UTX_2: planes of transition metal atoms intercalate into the unit cell of the respective binary uranium dipnictide. The crystal structure of $CeAgSb_2$ can also be understood from the stacking arrangement of CeSb–Ag–CeSb–Sb layers [8].

The neutron scattering experiment on a polycrystal of $CeAgSb_2$ done by André *et al* [10] indicated that the magnetic moment is simply oriented along the [001] direction (c-axis), with a Curie temperature $T_C = 9.6$ K and a magnetic moment $\mu_s = 0.33\ \mu_B$/Ce. However, the magnetization curve is quite anomalous [9]. The magnetization for the field along [001] indicates a typical ferromagnetic magnetization curve with $\mu_s = 0.37\ \mu_B$/Ce, while the magnetization for the field perpendicular to [001] increases almost linearly up to 30 kOe and saturates at higher fields, reaching $1.1\ \mu_B$/Ce. This corresponds to a metamagnetic transition in an antiferromagnet. These magnetization curves cannot be understood from the simple ferromagnetic structure proposed by the neutron scattering experiment.

This compound is also interesting with respect to a quasi-two dimensionality. The dHvA experiments on $CeAgSb_2$ as well as on reference compounds $YAgSb_2$ and $LaAgSb_2$ were reported by Myers *et al* [11]. A small Fermi surface was observed for $CeAgSb_2$. On the other hand, the Fermi surface in $YAgSb_2$ and $LaAgSb_2$ was found to consist of a cylindrical Fermi surface and two or three kinds of closed (ellipsoidal) ones. The reason why main Fermi surfaces were not observed in $CeAgSb_2$ was mainly due to the fact that the measurement was carried out at a high temperature of 2.1 K and the specific heat coefficient γ was reported to be 75 mJ K^{-2} mol^{-1} for the polycrystalline sample of $CeAgSb_2$ [12], which is larger than that of $LaAgSb_2$ ($\gamma = 2.62$ mJ K^{-2} mol^{-1}) [13].

We have taken an interest in sample quality and the quasi-two-dimensional electronic state of $CeAgSb_2$, and have studied the effect of pressure on the electrical resistivity in the temperature range from room temperature to 90 mK. In this letter we report a huge residual resistivity in the quantum critical region.

Single crystals were grown by the self-flux method, as described in [9]. The starting materials were $3N$(99.9% pure)-Ce, $5N$-Ag and $5N$-Sb. The typical size was $8 \times 5 \times 3$ mm^3, being flat in the (001) plane.

The electrical resistivity was measured by the four-probe dc method in an indenter pressure cell, with Daphne oil (7373) as the pressure-transmitting medium, which was cooled down to 90 mK in a dilution refrigerator. The pressure value was determined from the superconducting transition temperature of lead.

First we show the general feature of $CeAgSb_2$. Figure 1(a) shows the temperature dependence of the electrical resistivity ρ in the current J along [100] and [001]. The anisotropy of the resistivity is large: the resistivity ratio between $J \parallel [001]$ and [100] at room temperature and 10 K is $\rho_{[001]}/\rho_{[100]} = 8.3$ and 18, respectively, reflecting the quasi-two-dimensional electronic state. The residual resistivity ρ_0 and residual resistivity ratio (RRR $= \rho_{RT}/\rho_0$) are 0.19 $\mu\Omega$ cm and 430 for $J \parallel [100]$, and 0.38 $\mu\Omega$ cm and 1700 for $J \parallel [001]$, indicating a high-quality sample.

Figure 1(b) shows the T^2 dependence of the specific heat C in the form of C/T, together with that of $LaAgSb_2$. The electronic specific heat coefficient γ is 46 mJ K^{-2} mol^{-1} in $CeAgSb_2$ and 2.2 mJ K^{-2} mol^{-1} in $LaAgSb_2$, values which are approximately in agreement with previous results for polycrystalline samples [12, 13]. We note that the magnetic specific heat C_m becomes dominant below $T_{ord} = 9.7$ K, and follows a T^3 dependence which is expected for antiferromagnetic excitation or antiferromagnetic ordering.

As noted above, the magnetization for $H \parallel [001]$ has a hysteresis, not shown here, as in the usual ferromagnet with a saturated moment of $0.40\ \mu_B$/Ce, as shown in figure 1(c). On the other hand, the magnetization for $H \parallel [100]$ increases almost linearly with increasing

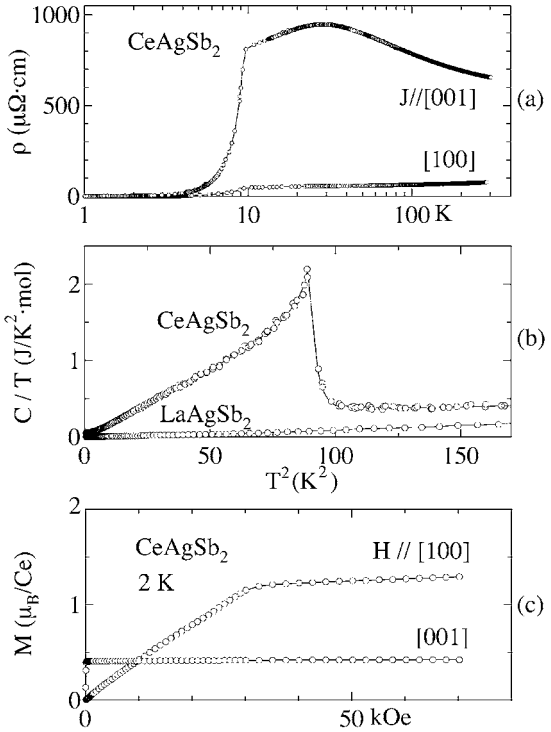

Figure 1. Temperature dependence of (a) the electrical resistivity in $CeAgSb_2$, (b) the specific heat in the form of C/T in $CeAgSb_2$ and $LaAgSb_2$ and (c) the magnetization curve for $H \parallel [001]$ in $CeAgSb_2$.

field and saturates above 30 kOe, indicating 1.20 μ_B/Ce at 32 kOe. This corresponds to a metamagnetic transition at 30 kOe as in an antiferromagnet. We note that the magnetization curve for $H \parallel [100]$ is reversible with increase and decrease of the field. These results for the magnetization curves cannot be understood using the simple ferromagnetic structure proposed by the neutron scattering experiment. The antiferromagnetic exchange interaction is essential in magnetization curves. Additional experiments such as neutron scattering on single crystals are required to clarify the magnetism in $CeAgSb_2$. We focus our interest on the pressure effect around the critical pressure p_c where the magnetic ordering temperature becomes zero.

Figure 2 shows the logarithmic scale of temperature dependence of the electrical resistivity in the current parallel to the (001) plane or the c-plane under several pressures. The resistivity at room temperature is 170 $\mu\Omega$ cm, which is larger than that for $J \parallel [100]$, about 80 $\mu\Omega$ cm. This is mainly due to both the small sample size and the contact of the current lead wires to the sample, related to the quasi-two-dimensional nature of the sample. The resistivity thus contains the contribution of the resistivity for $J \parallel [001]$

To clarify the magnetic ordering behaviour, we show in figures 3(a) and (b) the low-temperature resistivity. The magnetic ordering temperature T_{ord} is 9.7 K at ambient pressure, as shown by an arrow. With increasing pressure T_{ord} shifts to lower temperatures. Above about 3 GPa we could not identify any magnetic ordering in the resistivity. The resistivity peak at 5.8 K for $p = 3.6$ GPa shifts to 10 K for a higher pressure of 4.2 GPa. This characteristic temperature showing the resistivity peak approximately corresponds to the Kondo temperature, although it is influenced by the crystalline electric field effect. The temperature dependence of

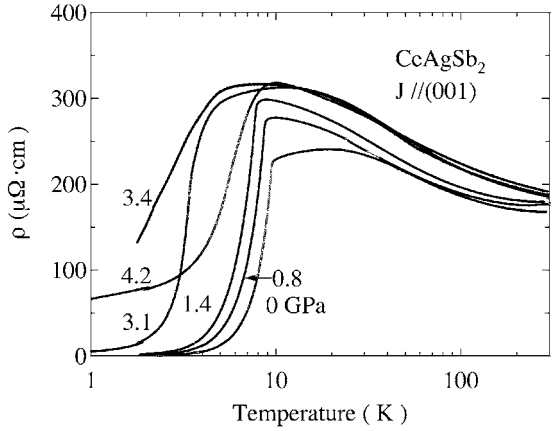

Figure 2. Temperature dependence of the electrical resistivity at several pressures in $CeAgSb_2$.

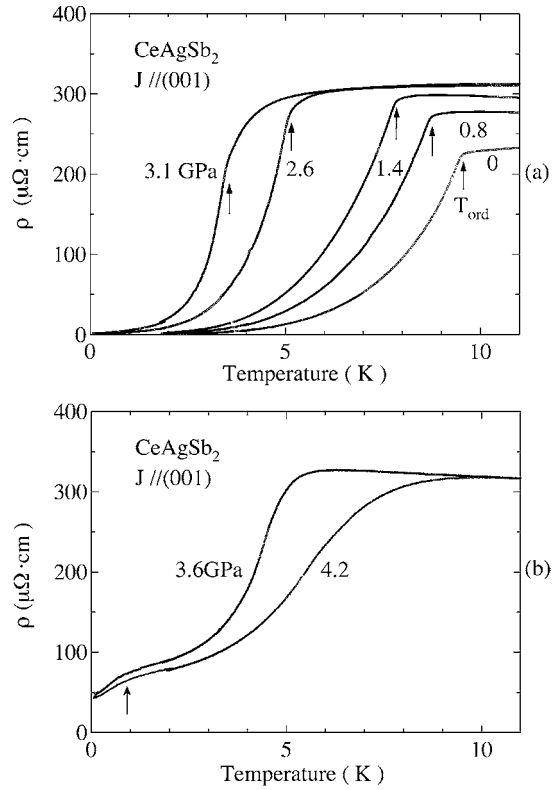

Figure 3. Low-temperature electrical resistivity at several pressures (a) below and (b) above 3.3 GPa in $CeAgSb_2$.

the electrical resistivity at 4.2 GPa in $CeAgSb_2$ is very similar to that of a typical non-magnetic heavy fermion compound $CeCu_6$ where the electrical resistivity increases with decreasing temperature, has a maximum at 15 K and decreases rapidly at lower temperatures [1].

Figure 4 shows the pressure dependence of the magnetic ordering temperature. The full curve is a guideline:

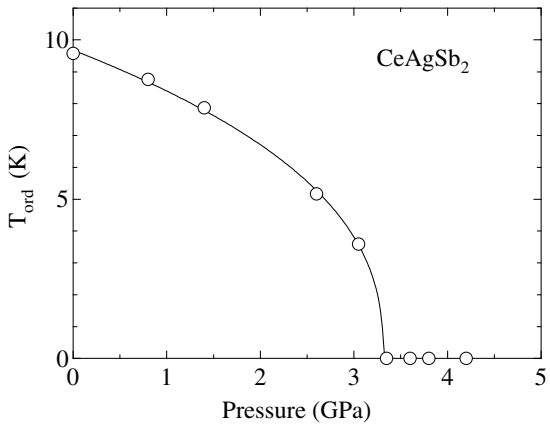

Figure 4. Pressure dependence of the magnetic ordering temperature in $CeAgSb_2$. The full curve connecting the data points is a guideline.

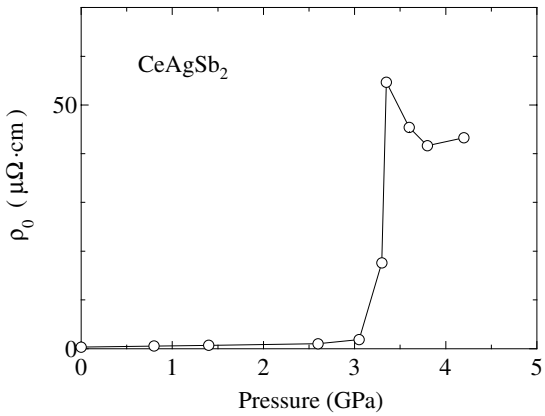

Figure 5. Pressure dependence of the residual resistivity in $CeAgSb_2$.

$$T_{\rm ord}(p) = T_{\rm ord}(p=0)\left(1-\frac{p}{p_{\rm c}}\right)^{n} \tag{1}$$

where $T_{\rm ord}(p=0) = 9.7$ K, $p_{\rm c} \simeq 3.3$ GPa and $n = 0.38$. An interesting finding is that above $p_{\rm c} = 3.3$ GPa, the residual resistivity becomes extremely large, as shown in figure 3(b). The residual resistivity is about 50 $\mu\Omega$ cm, which is much larger than the 0.5 $\mu\Omega$ cm value below $p_{\rm c}$.

Figure 5 shows the pressure dependence of the residual resistivity ρ_0, showing a maximum in the residual resistivity at $p_{\rm c}$. When the pressure was released, the residual resistivity was close to zero as in the case of the initial experiment. The present large residual resistivity is intrinsic and is a characteristic feature in the quantum critical region of $CeAgSb_2$.

Here the resistivity above 3.6 GPa decreases below 0.9 K, as shown by the arrow in figure 3(b). This resistivity decrease below 0.9 K was observed at 3.6, 3.8 and 4.2 GPa, and was also confirmed for another sample. There is a possibility that this is due to superconductivity, although the resistivity zero is not obtained. To clarify the present decrease in resistivity we measured the electrical resistivity under a magnetic field.

Figure 6 shows the low-temperature resistivity under $H = 0$ and 70 kOe for another sample. The configuration between $J \parallel (001)$ and $H \parallel [001]$ is transverse, $J \perp H$. A

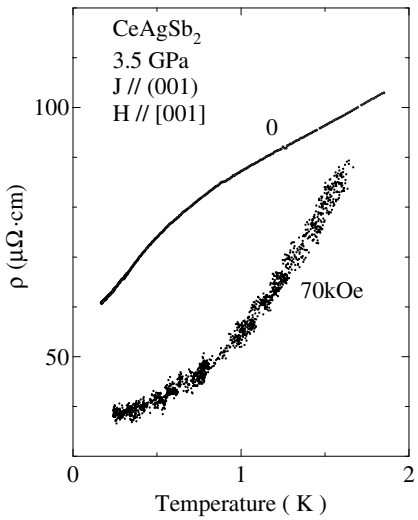

Figure 6. Temperature dependence of the electrical resistivity at 3.5 GPa under the magnetic field of 0 and 70 kOe in $CeAgSb_2$.

negative magnetoresistance is observed, as shown in figure 6. An interesting finding is that the resistivity under $H = 70$ kOe follows the usual Fermi liquid relation for the T^2 dependence. The present result is very similar to the non-Fermi liquid result in $CeCoIn_5$, which is known as a heavy fermion superconductor in the quantum critical region [14]. In other words, $CeCoIn_5$ indicates a clear T^1 dependence of the resistivity and shows negative magnetoresistance. At present it is not clear whether the resistivity decrease in $CeAgSb_2$ is due to the onset of superconductivity or not.

A new finding in the present experiment with $CeAgSb_2$ is that the residual resistivity possesses a huge value in the critical pressure region: $\rho_0 = 0.3\ \mu\Omega$ cm at ambient pressure but $\rho_0 = 55\ \mu\Omega$ cm at $p_c = 3.3$ GPa. Recently Miyake and Maebashi [15] explained theoretically the origin of the huge residual resistivity around the quantum critical region. This theory explains the sharp peak in the residual resistivity observed in $CeCu_2Ge_2$ at $p \simeq 17$ GPa where the superconducting transition temperature also exhibits a sharp peak: $\rho_0 = 2.5\ \mu\Omega$ cm at ambient pressure and 39 $\mu\Omega$ cm at 17 GPa in $CeCu_2Ge_2$ [16]. A similar pressure-induced superconductor is $CeRhIn_5$: for $CeRhIn_5$ $\rho_0 = 0.2\ \mu\Omega$ cm at ambient pressure and 13.2 $\mu\Omega$ cm at 2.5 GPa [17]. The theory is based on impurity scattering due to critical valence fluctuations of the 4f electrons in the cerium heavy fermion system. The present enhancement of the residual resistivity in $CeAgSb_2$ is large compared with the values for $CeCu_2Ge_2$ and $CeRhIn_5$.

Finally we note the relation between the crystal structure and pressure-induced superconductivity. The tetragonal crystal structure of $CeCu_2Ge_2$ and $CeRhIn_5$ has inversion symmetry which might be needed for superconductivity in a ferromagnet [18]. On the other hand, the tetragonal structure of $CeAgSb_2$ has no inversion symmetry. We need to make a further study of the decrease in resistivity in a higher-pressure region where the huge residual resistivity region is expected to exist over a wide pressure range.

In conclusion we have studied the pressure effect on high-quality single crystal $CeAgSb_2$ which has a net ferromagnetic moment of 0.4 μ_B/Ce. The critical pressure p_c was determined at $p_c \simeq 3.3$ GPa. When the pressure crosses p_c, a huge residual resistivity was observed, changing from 1.0 $\mu\Omega$ cm at $p_c = 2.6$ GPa to 55 $\mu\Omega$ cm at p_c.

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