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Angular Magnetoresistance of a Possible Kondo Insulator $CeOs_4Sb_{12}$ Measured at Ultra-Low Temperatures

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Angular magnetoresistance of the filled skutterudite compound $CeOs_4Sb_{12}$ has been investigated along the [110] direction. Distinct differences between the angular magnetoresistance data at $T = 0.55$ K and 4.2 K coincide with a developing of spin-density waves at $T \approx 1.7$ K in $B = 5$ T. Magnetoresistance experiments have been performed on a self-made assembly utilizing a commercial piezoelectric rotator.

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1. Introduction

Quantum mechanics not only governs the subatomic world but also dictates the organization of the microscopic particles in bulk matter at low temperatures. A prominent example is the concept of the Fermi surface (FS) that provides a precise explanation of the basic physical properties of metals. At absolute zero, electrons occupy the states with the lowest energies, up to an energy referred to as the Fermi energy. In the momentum space defined by the wave vector, this defines a Fermi surface, enclosing a Fermi volume in which all the states are occupied. Various interesting phenomena are determined by a topology of the Fermi surface and many of them are associated or even give rise to a FS reconstruction. Heavy-fermion compounds, being a prominent example of the strongly correlated systems, are exceptionally predisposed for a study of a FS reconstruction. This is because a Kondo effect strongly renormalizes an energy scale of heavy-fermion materials [1].

Direct measurements of the Fermi surfaces are typically done using the angle-resolved photoemission spectroscopy. In spite of impressive recent developments, this technique still does not have the resolution to study heavy-fermion metals in the required ultra-low-temperature range. The other well-established means to probe Fermi surfaces is the de Haas–van Alphen technique which, however, requires a large magnetic field of several dozens T. However, many important physical problems can be solved without a need to determine details of a Fermi surface. This is particularly true for an electron system displaying a low-lying phase transition or a quantum critical behavior. Simple but powerful possibility emerges when a moderate external magnetic field of a few T can be used as a tuning parameter between different ground states with distinctly different FS topologies. To investigate a FS reconstruction at ultra-low temperatures, we propose measurements of an angular dependence of the electrical resistivity taken in a transverse magnetic field. We have developed a station based on a commercially available piezoelectric rotary stepper positioner [2].

Test measurements were performed on a single-crystalline sample of $CeOs_4Sb_{12}$. This filled skutterudite compound $CeOs_4Sb_{12}$ is a rare example of a Kondo insulator [3, 4]. The Kondo insulators are prototypical strongly-correlated materials featuring $4f$-electron physics with a gap which is narrowed by antiferromagnetic interaction of conduction electrons with dense lattice of localized magnetic moments. As a result, the zero-field resistivity of $CeOs_4Sb_{12}$ increases by nearly two orders of magnitude upon cooling below $T = 60$ K [3]. Surprisingly, the Sommerfeld coefficient of the electronic specific heat is enhanced $\gamma \approx 180$ mJ K^{-2} mol^{-1}, as compared to other Ce-filled skuterrudite semiconductors [4–7]. Such an unusual duality of $4f$-electron states in $CeOs_4Sb_{12}$ is further suggested by photoemission spectroscopy experiments which demonstrated a symmetry-dependent hybridization effect [8]. Furthermore, transport measurements revealed a B-induced insulator–metal transition below 20 K [3]. Very recently, $CeOs_4Sb_{12}$ has been proposed as a candidate for a topological insulator [9].

Additionally, $CeOs_4Sb_{12}$ undergoes an antiferromagnetic (AFM) phase transition near 1 K as observed in transport and thermodynamic properties. It origin remains controversial and is a subject of intense experimental studies. We add, however, that there are several indications towards a spin-density-wave (SDW) type of the AFM order [10–13]. An important aspect of the ordered state is the field effect on the transition temperature, which markedly increases with field [10, 12]. The field–temperature phase diagram is very rich, particularly in the low-field range $B < 1.5$ T as concluded from the recent magnetostriction and magnetization measurements [13].

2. Experimental details

Single crystal of $CeOs_4Sb_{12}$ was grown using a molten-metal flux method with Sb flux [4]. The electrical resistivity $\rho(T)$ was studied along the [110] direction.

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Measurements in the temperature range $2\ \mathrm{K} \leq T \leq 300\ \mathrm{K}$ were performed using a commercial ^{4}He cryostat (PPMS). The resistivity at ultra-low temperature down to 0.08 K was investigated utilizing a ^{3}He–^{4}He dilution refrigerator. A standard four-point ac technique was applied in a zero magnetic field and an applied magnetic field of 5 T. An excitation current as low as 500 nA was used in the mK temperature range. Low-ohmic ($< 0.5\ \Omega$) electrical contacts were made using a spot welding technique. The angular magnetoresistance (AMR) was measured on a ^{3}He–^{4}He dilution refrigerator utilizing attocube system ANC350 controller for driving a piezoelectric rotary nanopositioner.

A special sample holder was designed for rotating the sample(s) with respect to the magnetic field below 1 K. Figure 1a shows its 3D assembly [14] and 1b depicts the completed self-made sample holder†. The heart of the setup is a rotary stepper positioner ANRv51\RES\LT\HV from attocube systems AG [2]. Its motion drive is comprised of two piezoelectric actuators. The rotator itself provides a clockwise and counter-clockwise endless rotations. In the \RES version it is integrated with a resistive position encoder which covers a finite range between 25° and 315°. A very precise readout of the absolute position during a turn allows operating in a closed-loop mode.

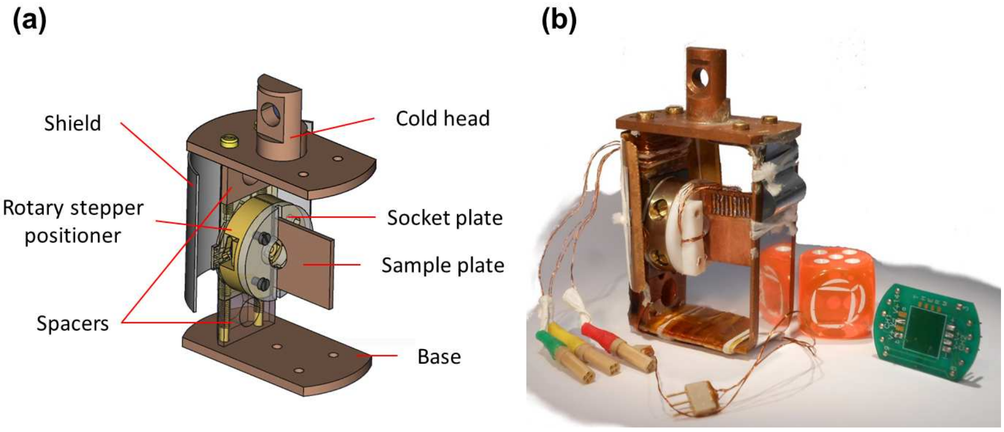

Fig. 1. Sample holder for the angular magnetoresistance measurements based on the piezoelectric rotary stepper positioner ANRv51\RES\UHV\LT from attocube systems AG as a rotation stage. (a) 3D assembly drawing of the sample holder designed to rotate the specimen(s) inside the 2-inch bore of the superconducting magnet. Some pieces are made semitransparent to emphasize the large cross-sections of the joints between the parts. (b) The completed self-made sample holder, to give a sense of its size, is compared with the standard sample plate for the commercial mechanical rotator for the PPMS (Quantum Design Inc.).

A few experimental conditions have to be fulfilled in order that the piezoelectric rotator may operate correctly: (i) The maximum load for the ANR51 series should not exceed 30 g (for a vertical arrangement the limit is about 20 g). (ii) The torque introduced by the measuring wires connected to the sample(s) should be as small as possible, because the frictional "slip-stick" driving mechanism provides only a limited torque for the rotator. (iii) To guarantee a sufficient voltage drop across the piezoceramic actuator's metallized plates, the rotator drive circuit has to be low-ohmic.

To minimize the load weight, we combined different materials keeping in mind relation between their thermal expansion coefficients [14]. The socket plate was made of teflon whereas the sample plate was made of copper. During a turn the sample temperature, T_s, is increasing mainly due to heat generated by the friction between the parts of the rotator. Even though we used teflon, after the rotator stops the T_s is restored to the value of the base temperature in less than 2 min. This is precisely known because a thermometer is mounted on the other side of the sample plate in virtually the same place in the experimental space as the investigated specimen. Very good thermal contact assures measuring a valid T_s during either a rest or a rotation. The large cross-sections of the joints between the parts provide a good heat sink to the mixing chamber.

The small torque of the measuring wires was achieved by forming them in a small free planar spiral and letting them into the rotator table's feedthrough placed exactly around the axis of rotation. A similar solution was introduced in Ref. [15]. The metallic shield shown in Fig. 1 prevents the unrolling planar spiral from touching the wall of the internal vacuum can during a turn of the rotary stepper.

†Assembly drawings were made using the FreeCAD open-source parametric 3D CAD modeler (LGPL2+). The rotator-drawing-part was provided by the manufacturer — attocube systems AG.

The voltage drop needed to cause a turn must compensate for both the load's weight and the wires' torque. A simple solution is to use, e.g., Cu wires with resistance less than 5 Ω. In the case of low-T experiments this is problematic due to the large thermal conductivity of copper. To ensure that the heat leak to the mixing chamber be small, as well ensuring the low resistance of the drive circuit, NbTi superconducting wires were used to separate the high-T Cu wires from the low-T Cu wires. The first ones run between the top of the ^{3}He–^{4}He dilution refrigerator (being at room temperature) and a 1 K-pot stage. The latter ones run from the mixing chamber down to the sample holder inside the superconducting magnet. Applying some non-superconducting material minimizes the influence of the magnetic field on the drive-voltage across the piezo.

3. Results and discussion

Figure 2 shows the temperature dependence of the electrical resistivity $\rho(T)$ for a $CeOs_4Sb_{12}$ single crystal measured between 0.08 K and 300 K. The $\rho(T)$ results, obtained in zero and magnetic field of $B = 5$ T applied along the [110] direction, are presented on a double logarithmic scale. Our $B = 0$ data are quite similar to the $\rho(T)$ behavior reported previously [3]. This holds true for, e.g., a metallic behavior down to about 60 K at which temperature a broad minimum is observed. Main characteristic of the zero-field resistivity of $CeOs_4Sb_{12}$ is an increase by nearly two orders of magnitude upon further cooling. However, there are a few minor differences: (i) the resistivity increases approximately as $T^{-0.6}$, while the previous works reported the $T^{-1/2}$ dependence [3]. (ii) At $T \lesssim 0.3$ K, the resistivity increases weaker than $T^{-0.6}$ and displays a tendency to saturation in the limit $T = 0$. (iii) A zero-field anomaly due to phase transition is hardly visible in our $\rho(T)$ data. We note that previously reported AFM transition temperatures in $CeOs_4Sb_{12}$ are significantly different among various measurements, indicative of its substantial sample-to-sample dependence. The sample-dependent T_N is likely a cause of incomplete and slightly different filling of the Os_4Sb_{12} cage by the Ce atoms. This suggestion is in line with several measurements performed on various collections of single crystals that pointed at obscure anomalies at around 1 K [13].

The most remarkable feature of a possible Kondo insulator $CeOs_4Sb_{12}$ is a field-induced insulator–metal transition. As reported in Ref. [3], the low-T resistivity of $CeOs_4Sb_{12}$ is reduced by nearly two orders of magnitude in a magnetic field of $B = 14$ T being applied perpendicularly to the [100] direction. Simultaneously, the metallic behavior of $\rho(T)$ above the broad minimum at ~ 60 K is unaffected by strong magnetic fields. On the other hand, the transition temperature to the ordered state is substantially affected by B in accord with its magnetic origin. The B–T phase diagram shows an enhancement of T_N in moderate fields, e.g., T_N is shifted to the maximum value of about 1.75 K for $B = 5$ T [13]. The

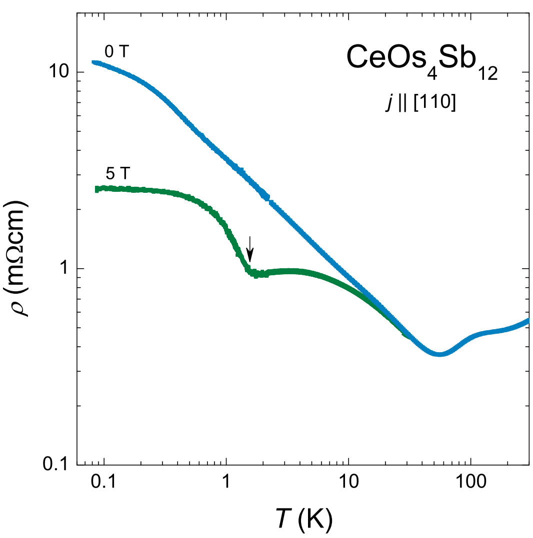

Fig. 2. The electrical resistivity of $CeOs_4Sb_{12}$ measured along the [110] direction. A magnetic field of $B = 5$ T was applied along the current flow. Arrow indicates a temperature at which a transition to a low-T ordered phase occurs.

temperature dependence of the resistivity measured in a field of 5 T (applied along the current flow) is exemplified in Fig. 2. Whereas differences between transverse [3] and longitudinal (our work) arrangements have a minor significance only, anisotropy effects in the cubic compound $CeOs_4Sb_{12}$ remain to be solved. We note that indications towards a field-induced anisotropy effect on a narrow hybridization gap have been observed in the closely related system $CeOs_4As_{12}$ [7].

Figure 3 presents results of angular magnetoresistance measurements performed for the sample $CeOs_4Sb_{12}$ whose $\rho(T)$ was discussed above. Our tentative AMR data in $B = 5$ T were collected at $T = 0.55$ K and 4.2 K, i.e., far below and much above a probable formation of SDW. Both curves were normalized to the value obtained for B perpendicular to the (001) plane. The inset shows the definition of a tilt angle θ. Owing to the cubic structure of the filled skutterudites, we have restricted our experiment to the first quadrant of θ.

At $T = 4.2$ K, the angular magnetoresistance rapidly increases with increasing the tilt angle and reaches the maximum value of about 45% for $\theta = 55°$. Upon further rotation, AMR decreases down to 35% for $\theta = 90°$. At $T = 0.55$ K, i.e., deep in the ordered state, an overall AMR is by a factor of nearly 7 larger than the magnetoresistivity measured at liquid helium temperature (*cf.* the dashed line in Fig. 3). Consequently, the magnetoresistivity as high as 325% as observed. More intriguingly, however, we have observed an additional enhancement of AMR close to the [111] direction, as marked by the shadowed area. This finding may suggest a reconstruction of

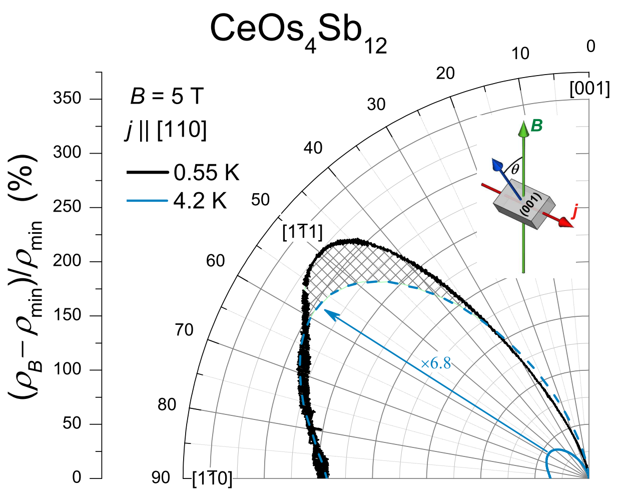

Fig. 3. Low-temperature angular magnetoresistance of $CeOs_4Sb_{12}$ measured in $B = 5$ T. Data are normalized to the minimum value for $B \parallel [001]$. The inset shows the definition of a tilt angle. Three main crystallographic directions along which a magnetic field was penetrating our sample are highlighted.

a Fermi surface when the electron system undergoes the phase transition at about 1.7 K. In a SDW scenario, a directional character of the observed anomaly may be interpreted as a result of the presence of parallel electron and hole sheets of the Fermi surface in $CeOs_4Sb_{12}$. Forthcoming AMR experiments for samples prepared along the [100] and [111] crystallographic directions should clarify if the observed anomaly in the angular magnetoresistance may be attributed to nesting properties of the Fermi surface of $CeOs_4Sb_{12}$.

Acknowledgments

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References

[1] Q. Si, F. Steglich, *Science* **329**, 1161 (2010).

[2] Technical specifications and press releases taken from attocube systems AG.

[3] H. Sugawara, S. Osaki, M. Kobayashi, T. Namiki, S.R. Saha, Y. Aoki, H. Sato, *Phys. Rev. B* **71**, 125127 (2005).

[4] E.D. Bauer, A. Slebarski, E.J. Freeman, C. Sirvent, M.B. Maple, *J. Phys. Condens. Matter* **13**, 4495 (2001).

[5] B. Andraka, C.R. Rotundu, K. Ingersent, *Phys. Rev. B* **81**, 054509 (2010).

[6] R.E. Baumbach, P.C. Ho, T.A. Sayles, M.B. Maple, R. Wawryk, T. Cichorek, A. Pietraszko, Z. Henkie, *Proc. Natl. Acad. Sci. U.S.A.* **105**, 17307 (2008).

[7] T. Cichorek, Ł. Bochenek, R. Wawryk, Z. Henkie, *Phys. Status Solidi B* **250**, 646 (2013).

[8] M. Matsunami, R. Eguchi, T. Kiss, K. Horiba, A. Chainani, M. Taguchi, K. Yamamoto, T. Togashi, S. Watanabe, X.-Y. Wang, C.-T. Chen, Y. Senba, H. Ohashi, H. Sugawara, H. Sato, H. Harima, S. Shin, *Phys. Rev. Lett.* **102**, 036403 (2009).

[9] B. Yan, L. Müchler, X.-L. Qi, S.-C. Zhang, C. Felser, *Phys. Rev. B* **85**, 165125 (2012).

[10] T. Namiki, Y. Aoki, H. Sugawara, T. Sato, *Acta Phys. Pol. B* **34**, 1161 (2003).

[11] M. Yogi, H. Kotegawa, G.Q. Zheng, Y. Kitaoka, S. Osaki, H. Sugawara, H. Sato, *J. Magn. Magn. Mater.* **45**, 272 (2004).

[12] C.R. Rotundu, B. Andraka, *Phys. Rev. B* **73**, 144429 (2006).

[13] T. Tayama, W. Ohmachi, M. Wansawa, D. Yutani, T. Sakakibara, H. Sugawara, H. Sato, *J. Phys. Soc. Jpn.* **84**, 104701 (2015).

[14] F. Pobell, *Matter and Methods at Low Temperatures*, 3rd ed., Springer, 2007.

[15] L.A. Yeoh, A. Srinivasan, T.P. Martin, O. Klochan, A.P. Micolich, A.R. Hamilton, *Rev. Sci. Instrum.* **81**, 113905 (2010).