Quantum oscillations with magnetic hysteresis observed in CeTe$_3$ thin films

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We have performed magnetotransport measurements in CeTe$_3$ thin films down to 0.2 K. It is known that CeTe$_3$ has two magnetic transitions at $T_{N1} \approx 3$ K and $T_{N2} \approx 1$ K. A clear Shubnikov-de-Haas (SdH) oscillation was observed at 4 K, demonstrating the strong two-dimensional nature in this material. Below $T_{N2}$, the SdH oscillation has two frequencies, indicating that the Fermi surface could be slightly modulated due to the second magnetic transition. We also observed a magnetic hysteresis in the SdH oscillation below $T_{N1}$. Especially, there is a unique spike in the magnetoresistance at $B \approx 0.6$ T only when the magnetic field is swept from a high enough field (more than 2 T) to zero field.

FIG. 1. (a) Crystal structure of CeTe$_3$. The red and blue spheres represent Ce and Te atoms, respectively. The black lined box indicates the unit cell, with unit lattice vectors $a \sim c \approx 4.4$ Å and $b \sim 26$ Å. (b) SEM image of a typical thin film device. CeTe$_3$ is shown in yellow (false color). (c) Temperature dependence of the resistivity in the CeTe$_3$ thin film device. There are two resistivity drops at 2.7 K and 1.3 K, which correspond to the two magnetic transition temperatures $T_{N1}$ and $T_{N2}$ respectively.

This material is also known to show two magnetic phase transitions at low temperature.$^{24,32,33}$ The first magnetic transition at $T_{N1} = 3.1$ K is understood to be from a paramagnetic state to an antiferromagnetic (possibly short range ordering) state. In this phase, the magnetic moment at the Ce site is antiferromagnetically coupled and aligned to an easy axis perpendicular to the layer stacking direction. The second magnetic transition is known to be another antiferromagnetic (possibly long

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peak at 0 T is due to the DC offset of the frequency. The capacity has been observed below the case of the first transition, a clear peak in the heat served at and swept from zero to 8 T. (b) FFT for the derivative of range ordering) transition at $T_{N2} = 1.3$ K. Unlike the case of the first transition, a clear peak in the heat capacity has been observed below $T_{N2}$ and the magnetic moment is still aligned to the in-plane direction (called non-parallel easy axis), but different from the easy axis in the first transition. Nevertheless, the details of these magnetic ordering states are poorly understood. Furthermore, there are no reports on thin film transport measurements.

In the present work, we have performed magnetotransport measurements with 30-40 nm thick CeTe$_3$ devices. We have observed the coexistence of quantum oscillation with unique magnetotransport phenomena. Specifically, a clear Shubnikov-de Haas (SdH) oscillation was observed in the magnetic field $B$ range from 3 to 8 T even above $T_{N1}$, demonstrating the existence of a small Fermi surface pocket. Since such a SdH oscillation has never been reported in bulk CeTe$_3$, this result reveals its strong 2D nature which is possibly enhanced by the thin film fabrication. Furthermore, the SdH oscillation has two frequencies below $T_{N2}$. This could originate from the modification of the Fermi surface due to the second magnetic transition at $T_{N2}$. We also observed a magnetic hysteresis in the SdH oscillation below $T_{N2}$. In particular, a sharp resistance peak appears at $B = 0.6$ T only when the magnetic field is swept from a high enough magnetic field (more than 2 T) to zero field.

Single crystals of CeTe$_3$ were synthesized in an evacuated quartz tube. The tube was heated up to 900°C and slowly cooled to 550°C over a period of 7 days. To fabricate thin film devices from the bulk CeTe$_3$, we used the mechanical exfoliation technique using scotch tapes. It is noted that all the following fabrication processes should be carried out inside a glove box with Ar purity of 99.9999%, since CeTe$_3$ is extremely sensitive to ambient air. After the mechanical exfoliation process, many CeTe$_3$ thin flakes on the scotch tape were transferred onto a thermally-oxidized silicon substrate. We then spin-coated polymethyl-methacrylate (PMMA) resist onto the substrate. The substrate was taken out from the glove box and electrode patterns were printed using electron beam lithography. After the lithography, the substrate was put back into the glove box again for the development of the resist. The Au electrodes were deposited using electron beam deposition in a vacuum chamber next to the glove box. Before the deposition of Au, Ar milling was performed to remove the residual resist and any possibly oxidized layers of CeTe$_3$. It should be noted that contact resistance varies greatly with electrode material. Au electrodes were found to have a minimum contact resistance compared to Ti/Au or Cu. In order to avoid further damage in ambient condition after the fabrication, the device was capped with the PMMA shortly after the electrode deposition and the lift-off process. Figure 1(b) shows a scanning electron microscopy (SEM) image of one of the thin film devices.

Electrical transport measurements were performed by the conventional four-probe method using a lock-in amplifier. The device was cooled with a 3He/4He dilution refrigerator down to 0.2 K and the external magnetic field was applied using a superconducting magnet. The thicknesses of all measured CeTe$_3$ thin films were confirmed by using a commercially available atomic force microscope after finishing all the electrical measurements.

The resistivity $\rho(T)$ measured with the CeTe$_3$ thin film device in Fig. 1(b) is plotted as a function of temperature in Fig. 1(c). Although the CeTe structure is half-metallic on its own, the high conductivity between the two Te sheets gives rise to a highly metallic temperature dependence in CeTe$_3$. There are two resistivity changes in the low temperature region: the first resistivity drop at $T = 2.7$ K, and the second resistivity kink structure at $T = 1.3$ K. These behaviors are consistent with the anomalies observed in bulk CeTe$_3$ resistivity measurements, which correspond to the two magnetic phase transitions at $T_{N1} = 3.1$ K and $T_{N2} = 1.3$ K. The resistivity at room temperature of this device is 81.3 $\mu\Omega$-cm, resulting in the residual resistivity ratio (RRR) of 59.2 with respect to $\rho(T = 1.5)$ K. This is 1.3 times higher than the previously reported value of 44.9. At the lowest temperature ($T = 0.2$ K), RRR reaches a value of

![FIG. 2. (a) Magnetoresistance measured with a 40 nm thick CeTe$_3$ thin film device at several different temperatures. The external magnetic field was applied perpendicular to the plane and swept from zero to 8 T. (b) FFT for the derivative of $\rho_{xx}(B)$ versus $1/B$. The main oscillation peak has been observed at $f_0 = 32.4$ T below 4 K. The secondary oscillation peak has been observed at $f_2 = 11.0$ T only below 0.8 K. The peak at 0 T is due to the DC offset of the frequency.](image-url)
140, indicating that the device is a high quality single crystal CeTe$_3$ thin film.

We next performed magnetoresistance measurements up to $B = 8$ T for the temperature range from 0.4 to 4 K, as shown in Fig. 2(a). The external magnetic field $B$ was applied perpendicular to the plane, i.e., along the $b$-axis of CeTe$_3$ and swept from zero to 8 T. A large positive magnetoresistance $\rho_{xx}(B)$ was observed in the low field regime ($B < 1$ T) along with a clear SdH oscillation which develops from magnetic fields as low as $B = 2$ T in the case of $T = 2$ K. As far as we know, such a quantum oscillation (including the de Haas-van Alphen effect) has never been reported so far even for bulk CeTe$_3$, although Lei et al. have recently reported a SdH oscillation in GdTe$_3$ thin films. This fact is possibly related to much stronger 2D nature in thin films, compared to bulk crystals. Furthermore, the magnetoresistance behavior drastically changes below $T = 0.8$ K, which is below the second magnetic transition temperature $T_{N2}$.

In order to extract the oscillatory part of the magnetoresistance, the derivative of $\rho_{xx}(B)$ with respect to $1/B$ was obtained numerically. Fast Fourier transform (FFT) was then performed in order to obtain the frequency $f$ of the quantum oscillation. The results are presented in Fig. 2(b). The main oscillation observed at all temperatures corresponds to a frequency of $f_0 = 32.4$ T, which is about two times smaller than that of GdTe$_3$. Below $T = 0.8$ K, there is an additional oscillation with $f_2 = 11.0$ T, which can be seen as the secondary peak in the FFT spectrum in Fig. 2(b). The frequencies of these oscillations are proportional to the extremal cross sectional areas of the Fermi surface perpendicular to the applied magnetic field: $S = 2\pi \epsilon f / h$, where $S$ is the Fermi surface area, $\epsilon$ is the elementary charge, $f$ is the frequency in the unit of T, and $h$ is the reduced Planck constant. Using this equation, we obtained the Fermi surface areas of $S_0 = 3.09 \times 10^{15} \text{cm}^{-2}$ for $f_0 = 32.4$ T, and $S_2 = 1.05 \times 10^{13} \text{cm}^{-2}$ for $f_2 = 11.0$ T. The main oscillation is present even above the two magnetic transition temperatures, but well below its CDW transition temperature ($T_{CDW} > 500$ K). The origin of this Fermi surface pocket can be attributed to the reconstruction of the Fermi surface due to the incommensurate CDW, which has been observed through photoemission spectroscopy experiments, as well as through quantum oscillations of other $R$Te$_3$ materials.

Although the angular dependence of the SdH oscillation has not been measured in the present study, it was already demonstrated in a similar tritelluride thin film device, i.e., GdTe$_3$, where the SdH oscillation follows $1/\cos \theta$ ($\theta$ is the angle between the applied magnetic field and the layer stacking direction). This indicates a highly 2D geometry of the Fermi pockets. Since the crystal structure and the high conduction of the Te layers are the same for GdTe$_3$ and CeTe$_3$, we believe that the SdH oscillation observed in CeTe$_3$ also originates from a highly 2D Fermi surface pocket. Especially, $S_0$ in the CeTe$_3$ device is two times smaller than that in a thin film GdTe$_3$ device, where the effective mass is known to be as small as $\approx 0.1 m_0$ ($m_0$ is the bare electron mass). This fact suggests that the conduction electrons between the Te sheets in the CeTe$_3$ device has a similarly small effective mass. In addition, we detected the secondary oscillation with a frequency of $f_2$, which does not exist in GdTe$_3$ and develops only after the second magnetic transition temperature $T_{N2}$. According to Ref. 32, the second magnetic transition at $T_{N2}$ is related to a spin density wave transition with formation of heavy quasi-particles. Thus, such reconstruction of the Fermi surface along with this transition could be a possible cause of this new oscillation, but further investigation is required to confirm the hypothesis.

In a typical SdH oscillation, the FFT amplitude for a given frequency decreases with increasing temperature. However, FFT amplitudes of the main oscillation remained mostly constant, with the exception at $T = 2$ K. A similar result has been reported in GdTe$_3$, where the FFT amplitude plateaus below its antiferromagnetic transition temperature, while showing a typical temperature dependence well above the transition temperature. Our scenario is consistent with this report: in other words, the unconventional temperature dependence of the FFT amplitude depends strongly on the interaction between the magnetic order at the CeTe site and the conduction electrons.

In addition to the SdH oscillations discussed above, we observed a magnetic hysteresis behavior, superimposed onto the SdH oscillations, below the first antiferromagnetic ordering temperature $T_{N1}$. This is highlighted in Fig. 3. At 4 K above $T_{N1}$, there is no hysteresis in the SdH oscillation. Below $T_{N1}$, however, a clear magnetic hysteresis is observed with a sharp peak structure at $B \approx 0.6$ T. The magnetic hysteresis in the SdH oscillation vanishes above $B \approx 4$ T, and the sharp peak appears only when $B$ is swept from a high enough field.
appears when the negative field sweeping. Note that quantum oscillations with magnetic hysteresis were reproducible for other CeTe thin film devices, even below $T_{N2} = 1.3$ K, on the other hand, SdH oscillations with two different frequencies were obtained. The FFT analysis revealed the existence of two small Fermi pockets whose sizes are $3.09 \times 10^{13}$ cm$^{-2}$ and $1.05 \times 10^{13}$ cm$^{-2}$. In addition, a magnetic hysteresis superimposed on the SdH oscillation was detected below the first antiferromagnetic temperature $T_{N1} = 2.7$ K. Especially, a sharp peak at $B \approx 0.6$ T was clearly observed when the magnetic field was swept from the high enough field to zero field. Materials where quantum oscillations and magnetic hysteresis coexist are extremely scarce. Along with the ease of thin film fabrication through mechanical exfoliation, further researches on CeTe$_3$ could pave the way for $f$-orbital spintronics and could provide an ideal stage for understanding the interplay between electronic quantum conduction and localized heavy fermion spins.

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