Superconductivity in Magnetically Ordered CeTe$_{1.82}$

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Abstract

We report the discovery of pressure-induced superconductivity in a semimetallic magnetic material CeTe$_{1.82}$. The superconducting transition temperature $T_{SC} = 2.7$ K (well below the magnetic ordering temperatures) under pressure ($> 2$ kbar) is remarkably high, considering the relatively low carrier density due to a charge-density-wave transition associated with lattice modulation. The coexisting magnetic structure of a mixed ferromagnetism and antiferromagnetism can provide a clue for this high $T_{SC}$. We discuss a theoretical model for its possible pairing symmetry and pairing mechanism.
The $f$-electron compounds never cease to surprise us with interesting ground states. Due to the local nature of $f$-obital wave function, these compounds display various many-body collective state(s) at low temperature – often competition/coexistance among them. Typically, the $f$-electron systems show a heavily renormalized quasiparticle (called heavy fermion) below a certain crossover temperature, and then many of them at lower temperatures show magnetism and/or superconductivity (SC) such as in CeCu$_2$Si$_2$, CeIn$_3$, CeRhIn$_5$, UNi$_2$Al$_3$, UGe$_2$, etc. [1,2]. CeTe$_{1.82}$ studied here certainly belongs to this class of materials. However, due to Te and the crystal structure this compound also shares some of the feature of the layered transition-metal dichalcogenides and NbSe$_3$, which undergo a superconducting transition at low temperature $\sim 1$ K with the charge-density-wave (CDW) ordering at far higher temperature $\sim 1000$ K [3]. The precise role of CDW with respect to SC is unclear so far and their coherent properties now constitute a separate interesting branch of correlated electron systems [4]. The compound CeTe$_{1.82}$ studied in this Letter shows all these collective states: CDW, magnetism, and SC in consecutively lowering temperature.

Here we report the first observation of pressure-induced SC in a semimetallic magnetic material CeTe$_{1.82}$ with a relatively low density of states (DOS) [5]. At ambient pressure, CeTe$_{2-\delta}$(0.13 $\leq \delta \leq$ 0.18) displays various collective ground states and exhibits highly anisotropic transport and magnetic properties. CeTe$_{2-\delta}$ crystallizes in layered tetragonal Cu$_2$Sb-type structure, where a metallic Te sheet is sandwiched by semiconducting CeTe double layers and is stacked along the $c$ axis [6]. Because of this layered crystal structure and the Te vacancy, a CDW state is stabilized even at far above the room temperature. The presence of CDW gap ($T_{CDW} \sim 1000$ K) accompanying with a lattice distortion in the Te sheet is verified by electron-tunneling spectroscopy measurements [7]. At low temperatures, this compound undergoes two different magnetic orderings. The local magnetic moments of Ce ions develop a short-range ferromagnetic (SRF) ordering in the CeTe layer with a magnetoelastic origin below $T_{SRF} \sim 6$ K. As temperature is further lowered, the SRF CeTe layers develop a long-range ferromagnetic (FM) order in the layers and simultaneously a long-range antiferromagnetic (AFM) order in the spin sequence of down-up-up-down along...
the $c$ axis below $T_N \sim 4.3$ K (see Ref. [8] and Fig. 1). Because of the two-dimensional motion of the carriers confined within the Te sheet sandwiched by the ferromagnetically coupled CeTe layers, the strong anisotropy is observed in the electrical resistivity with a ratio of $\rho_{||}/\rho_{\perp} \sim 150$ at 2 K and the isothermal magnetization with a ratio of $M_{||}/M_{\perp} \sim 7$ at 2 kG [6,8].

High-purity single crystals were grown with varying Te contents, i.e., $0.13 \leq \delta \leq 0.18$ in CeTe$_{2-\delta}$. Electron-probe microanalysis reveals the deficiency in the Te content, $\delta$, without any evidence of inhomogeneity to a resolution of 0.1%. This $\delta$ value is often observed in other rare-earth dichalcogenides such as LaTe$_{1.9}$, SmTe$_{1.84}$, and DySe$_{1.85}$ [9], where the chalcogen vacancy goes into the Te sheet and stabilizes the structural modulation. The in-plane resistivity measurements were made on single-crystal platelets by the conventional a.c. four-terminal method as a function of temperature, magnetic field, and pressure. The pressure cell is of the piston-cylinder type constructed out of high-purity non-magnetic BeCu alloy suitable for the application of external magnetic fields. The pressure was determined to $\pm 0.005$ kbar from the electrical resistance of Manganin sensor. Bulk magnetization measurements as a function of temperature were performed by means of a SQUID magnetometer (Quantum Design, MPMS7) with similar pressure cells, in which 1:1 mixture of Flurinert FC70 and FC77 was used for a pressure transmitting medium.

Figure 2 displays a typical feature of $\rho(T)$ showing the superconducting transition in CeTe$_{1.82}$ at $P = 5$ kbar, where $\rho(T)$ starts to drop drastically at $T_{SC} = 2.7$ K. The application of magnetic field suppresses the resistivity drop, as expected for a superconducting transition. From the onset of the superconducting transition, we have determined the superconducting phase diagram shown in the inset of Fig. 2. It is rather unusual that the upper critical field $H_{c2}$ ($\sim 5$ kG at $T \rightarrow 0$) is about an order smaller than $H_{c2}(0)$ ($\sim 100$ kG at 20 kbar) of CeRhIn$_5$ [10] having a similar $T_{SC}$ ($\sim 2.1$ K). We consider the low DOS of CeTe$_{1.82}$ as the primary reason for such a small $H_{c2}$. Then, the relatively high $T_{SC}$ with a low DOS indicates a different pairing mechanism compared to CeRhIn$_5$ and other heavy fermion superconductors.
A more conclusive evidence for SC is a diamagnetic signal below $T_{SC}$, and thus we measured the magnetization $M(T)$ of CeTe$_{1.82}$ for different pressures. Because the superconducting transition occurs just below the magnetic transition and the SC coexists with the magnetism below $T_{SC}$, a diamagnetic signal associated with SC is quite small to be easily detected. Hence, we have first plotted the difference $\Delta M (= M_{ZFC} - M_{FC})$ between the zero-field-cooled ($M_{ZFC}$) and field-cooled data ($M_{FC}$) in the inset of Fig. 3, showing a clear deviation from the linear-temperature-dependent background ($M_{BG}$). This background is defined as an extrapolation of the magnetic hysteresis of $M(T)$ below $T_N$. The main panel of Fig. 3 shows the magnetic susceptibility $4\pi \Delta M_{SC}/H$ measured at 6 kbar, where $\Delta M_{SC} = \Delta M - M_{BG}$, assuming the density of CeTe$_{1.82}$ is about 10 g/cc. The diamagnetic component is observed just below 2.8 K, which coincides with $T_{SC}$ determined by $\rho(T)$ measurements. These results support the presence of bulk SC in CeTe$_{1.82}$.

In Fig. 4, we draw the phase diagram in temperature-pressure space summarizing our measurements for CeTe$_{1.82}$. $\rho(T)$ and $M(T)$ at different pressures allow us to identify the short-range ferromagnetic ordering temperature $T_{SRF}$ and the long-range ferro/antiferromagnetic ordering temperature $T_N$. The superconducting transition temperature $T_{SC}$ is determined from $\rho(T)$. The applied pressure slightly enhances both $T_{SRF}$ and $T_N$ over the whole region of measured pressure. The SC suddenly appears in the narrow region below 2 kbar. The pressure-induced SC often occurs in heavy fermion metals in the vicinity of AFM or FM quantum criticality (QC: $T_N$ or $T_C \rightarrow 0$ K) [2] and the normal state properties exhibit various deviations from Fermi-liquid metal, so called, non-Fermi liquid (NFL) behavior [11]. However, for CeTe$_{1.82}$ there is no magnetic QC in our phase diagram and the superconducting phase exists completely inside the magnetic phase. Also, the transport and magnetic properties show no NFL behavior.

We examine possible theoretical scenarios for the SC in CeTe$_{1.82}$, focusing on pairing interactions and pairing symmetries. If the SC is mediated by magnetic fluctuations, the phase diagram in Fig. 4 appears consistent with a FM-induced SC; an AFM-induced SC tends to appear near the boundary between magnetic and non-magnetic phases [12]. In fact,
considering the ferro/antiferro-magnetic ordering structure (Fig.1), in which the main conducting Te sheet is sandwiched between two FM CeTe double layers and this FM sandwich structure is alternating its polarization along the \(c\) axis, it is quite plausible that the carriers confined in the Te sheet interact by exchange of FM fluctuations and form superconducting pairs \(^{12}\). While the traditional idea for the FM-induced SC is a triplet and odd orbital pairing, recent theoretical studies suggest that singlet s-wave pairing is also possible inside a FM phase \(^{12}\). Although it remains an important issue for further experiments to determine the symmetry of the superconducting order parameter, the possibility of FM triplet pairing in CeTe\(_{1.82}\) has a couple of problems because of its sensitivity to disorder. The sample which exhibits SC has Te vacancy of \(\sim 10\%\), and most of this vacancy is believed to go into the Te sheets that are going to develop SC. Any triplet odd orbital pairing hardly survives in this much disorder. In addition, the pressure-induced SC is observed only for a single crystal with \(\delta = 0.18\) in CeTe\(_{2-\delta}\). We have measured in-pressure resistivity of other single crystals with \(\delta = 0.15\) and 0.13, and found almost identical magnetic properties but no superconducting transition. This indifference of SC to the magnetic properties and the extreme sensitivity to the Te vacancy suggests that the magnetism is unlikely to be a primary source of SC pairing mechanism.

From the sensitive dependence of SC to \(\delta\), one could speculate that the SC in CeTe\(_{1.82}\) is most likely to be associated with the crystal-lattice instability. The existence of CDW instability driven by the Fermi surface nesting is realized with a small periodic lattice distortion within the Te sheet, which is stabilized by a vacancy order in the Te sheets \(^7\). Related to this, an interesting pairing mechanism has been proposed by Castro Neto \(^4\) in order to explain CDW-SC in transition-metal dichalcogenides (TMD). In this theory, the SC pairing occurs with the Dirac fermions formed after a gapless CDW ordering, which couple with acoustic phonons via piezoelectric coupling due to the inversion symmetry breaking by a six-fold CDW order. The main difference between TMD compounds and CeTe\(_{2-\delta}\) is that CeTe\(_{2-\delta}\) has \(f\)-orbital moments from Ce ions and these moments develop magnetic orderings at low temperature (6 K and 4.3 K). There are experiments indicating possible interplay
between CDW and magnetic order in CeTe$_{2-\delta}$, but the details are still unclear\cite{4}. Also the piezoelectric coupling is, in general, unlikely in metals\cite{13}. Therefore, the application of the pairing theory for TMD to our case is not straightforward. However, on a general ground the CDW ordering and accompanying lattice modulation should create a new optical phonon mode, which then couples to electrons in the Te layers. Hence, we speculate that the primary pairing interaction is mediated by phonons forming a s-wave singlet SC.

As for the role of magnetism for SC, we can think of two effects: (1) In addition to the phonon pairing potential, the FM fluctuations in the FM phase can contribute to a s-wave singlet pairing\cite{12}; (2) The tunneling data from Ref.\cite{7} shows that the zero-bias conductance increases below $T_{SRF}$, indicating that DOS increases due to magnetic ordering. Finally as for the role of pressure, we think that the $c$-axis lattice distance is crucial to determine the actual $T_{SC}$ as in TMD. Figure 4 shows that the SC, coexisting with magnetism, abruptly appears at $T_{SC} = 2.7$ K with as low pressure as 2 kbar. As indicated by the dotted line, we cannot rule out that the superconducting phase appears sharply below 2 kbar. Then the pressure might decrease the $c$ lattice constant and this will increase the interlayer coupling to increase $T_{SC}$ as in other layered superconductors.

To conclude, we report the superconducting transition at $T_{SC} = 2.7$ K in CeTe$_{1.82}$ under pressure ($P > 2$ kbar). CeTe$_{2-\delta}$ displays various collective states at different temperatures; CDW ($\sim 1000$ K), SRF ($\sim 6$ K), $c$-axis AFM ($\sim 4.3$ K), and finally SC transition for $\delta = 0.18$. Combining available data and phase diagram, we conclude that the primary pairing mechanism is a phonon-mediated s-wave SC, enhanced by the FM fluctuations inside the magnetic ordering phase and the increased DOS due to the FM ordering. The unique magnetic structure of antiferromagnetically alternating FM CeTe layers cancels the internal fields on the Te sheets, which become superconducting. This fact negates the crucial negative effect of FM on SC. All these factors make CeTe$_{1.82}$ a remarkably high $T_{SC} = 2.7$ K among $f$-electron systems. Finally, the pressure is required to increase the interlayer coupling and $T_{SC}$ in the layered superconductors by decreasing the interlayer distance along the $c$ axis.

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FIG. 1. The crystal and magnetic structure of CeTe$_2$ below $T_N$. 
FIG. 2. Temperature dependence of the in-plane resistance $R(T)/R(4K)$ normalized to 4 K value in pressure 5 kbar at c-axis fields 0, 1.5, 2 and 3 kG. The inset shows upper critical fields $H_{c2}(T)$ from the onset of superconducting transition, at which the resistance first deviates from the normal-state value.
FIG. 3. Temperature dependence of $4\pi \Delta M_{SC}/H$ (main panel), $\Delta M$ (open circles in the inset), and $M_{BG}$ (straight line in the inset) at 6 kbar, defined in the text: $\Delta M = M_{ZFC} - M_{FC}$ and $\Delta M_{SC} = \Delta M - M_{BG}$. Density of CeTe$_{1.82}$ under pressure is assumed about 10 g/cc.
FIG. 4. Various critical temperatures as a function of applied pressure: the short-range ferromagnetic ordering temperature $T_{SRF}$, the long-range antiferromagnetic ordering temperature $T_N$, and the superconducting transition temperature $T_{SC}$. $T_{SRF}$ and $T_{SC}$ are determined from $\rho(T, P)$ data and $T_N$ from $M(T, P)$ data. The solid and dotted lines are guides for eyes.