Superconductivity in magnetically ordered $CeTe_{1.82}$

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We report the discovery of pressure-induced superconductivity in a semimetallic magnetic material $CeTe_{1.82}$. The superconducting transition temperature $T_c = 2.7$ 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 mixed magnetic structure of antiferromagnetism coexisting with ferromagnetism can provide a clue for this high T_c. We discuss a possible theoretical model for the superconducting pairing mechanism.

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Recently, quite different collective states have been observed, i.e., superconducting state coexisting/competing with magnetic ordering or charge density waves. Certain *f*-electron systems containing the lanthanide and actinide components, e.g., $CeCu_2Si_2$, $CeIn_3$, $CeRhIn_5$, UNi_2Al_3, UGe_2, etc., appear to be a manifestation of the competition/interplay between superconductivity (SC) and magnetism.[1,2] In these *f*-electron systems, the most possible pairing potential is believed to be of a magnetic origin and the pairing symmetry is an unconventional one. Another collective state, the charge density wave (CDW) in addition to SC, is observed in such materials as layered transition-metal dichalcogenides and $NbSe_3$, which undergo a superconducting transition with the CDW ordering at far higher temperatures $\sim$1000 K.[3] The precise role of CDW with respect to SC has been unclear so far. Their coherent properties now constitute a separate interesting branch of correlated electron systems.[4] The compound $CeTe_{1.82}$ studied in this paper shows all these collective states: CDW, magnetism, and SC at consecutively lowering temperature.

Here we report an observation of pressure-induced SC in a semimetallic magnetic material $CeTe_{1.82}$ with a relatively low density of states (DOS).[5,6] At ambient pressure, $CeTe_{2-\delta}(0.13 \leqslant \delta \leqslant 0.18)$ displays various collective ground states and exhibits highly anisotropic transport and magnetic properties.[6] It crystallizes in layered tetragonal Cu_2Sb-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) is verified by electron-tunneling spectroscopy measurements.[7] At low temperatures, this compound undergoes two different magnetic orderings.[8] 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 $\rho_{\parallel c}/\rho_{\perp c} \sim 150$ at 2 K and the isothermal magnetization with a ratio of $M_{\parallel c}/M_{\perp c} \sim 7$ at 2 kG.[6,8]

High-purity single crystals were grown with varying Te contents, i.e., $0.13 \leqslant \delta \leqslant 0.18$ in $CeTe_{2-\delta}$. Electron-probe microanalysis reveals the deficiency in the Te content δ without any evidence of inhomogeneity to a resolution of 0.1%. This δ 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 ac 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 nonmagnetic BeCu alloy

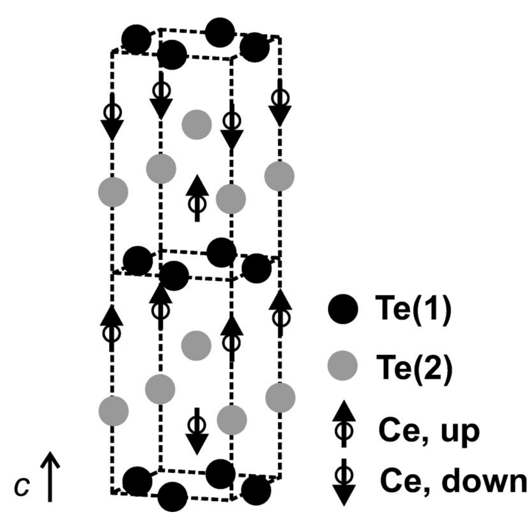

FIG. 1. The crystal and magnetic structure of $CeTe_2$ below T_N.

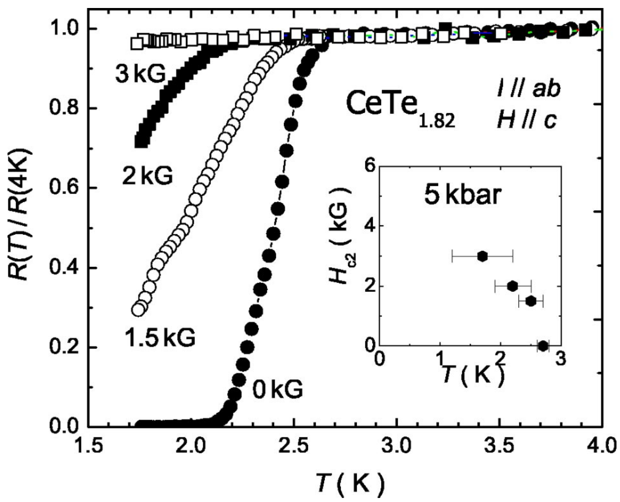

FIG. 2. Temperature dependence of the in-plane resistance $R(T)/R(4\ \mathrm{K})$ normalized to the 4 K value in pressure 5 kbar at c-axis fields 0, 1.5, 2, and 3 kG. The inset shows upper critical fields $H_{c2}(T)$ from the onset of superconducting transition, at which the resistance first deviates from the normal-state value.

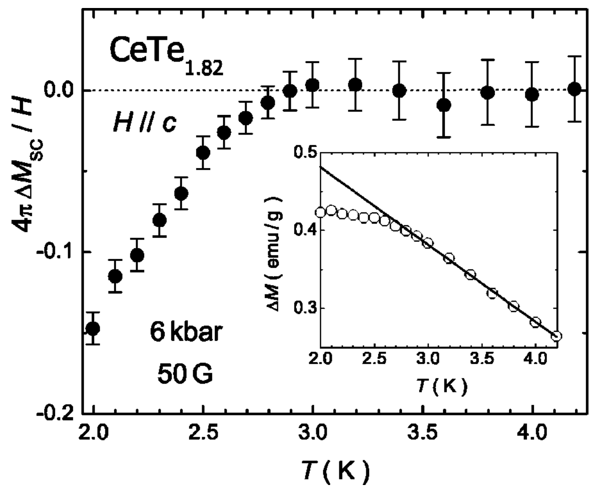

FIG. 3. Temperature dependence of $4\pi\Delta M_{\mathrm{SC}}/H$ (main panel), ΔM (open cicles in the inset), and M_{BG} (straight line in the inset) at 6 kbar, defined in the text: $\Delta M = |M_{\mathrm{ZFC}} - M_{\mathrm{FC}}|$ and $\Delta M_{\mathrm{SC}} = \Delta M - M_{\mathrm{BG}}$. Density of $\mathrm{CeTe}_{1.82}$ under pressure is assumed about 10 g/cc.

suitable for the application of external magnetic fields. The pressure was determined to ± 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 the resistivity $\rho(T)$ showing the superconducting transition in $\mathrm{CeTe}_{1.82}$ at $P = 5$ kbar, where $\rho(T)$ starts to drop drastically at $T_c = 2.7$ 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} (~ 5 kG at $T \rightarrow 0$) is about an order smaller than $H_{c2}(0)$ (~ 100 kG at 20 kbar) of CeRhIn_5 (Ref. 10) that has a similar T_c (~ 2.1 K). We consider the low DOS of $\mathrm{CeTe}_{1.82}$ (Refs. 5,6) as the primary reason for such a small H_{c2}. Then, the relatively high T_c 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_c, and thus we measured the magnetization $M(T)$ of $\mathrm{CeTe}_{1.82}$ for different pressures. Because the superconducting transition occurs just below the magnetic transition and the SC coexists with the magnetism below T_c, a diamagnetic signal associated with SC is quite small to be easily detected. Hence, we have first plotted the difference ΔM ($= |M_{\mathrm{ZFC}} - M_{\mathrm{FC}}|$) 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 normalized magnetic susceptibility $4\pi\Delta M_{\mathrm{SC}}/H$ measured at 6 kbar, where $\Delta M_{\mathrm{SC}} = \Delta M - M_{BG}$, assuming that the density of $\mathrm{CeTe}_{1.82}$ is about 10 g/cc. The diamagnetic component is observed just below 2.8 K, which coincides with T_c determined by $\rho(T)$ measurements. These results support the presence of bulk SC in $\mathrm{CeTe}_{1.82}$.

In Fig. 4, we draw the phase diagram in temperature-pressure space summarizing our measurements for $\mathrm{CeTe}_{1.82}$. $\rho(T)$ and $M(T)$ at different pressures allow us to identify the short-range ferromagnetic ordering temperature T_{SRF} and the long-range ferromagnetic/antiferromagnetic ordering tem-

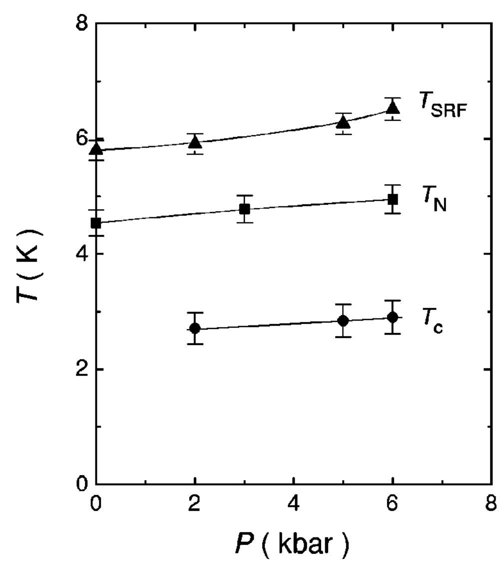

FIG. 4. Various critical temperatures as a function of applied pressure: the short-range ferromagnetic ordering temperature T_{SRF}, the long-range antiferromagnetic ordering temperature T_N, and the superconducting transition temperature T_c. T_{SRF} and T_c are determined from $\rho(T,P)$ data and T_N from $M(T,P)$ data. The solid and dotted lines are guides for eyes.

perature T_N. The superconducting transition temperature T_c 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 to be consistent with a FM-induced SC. This compared with an AFM-induced SC that tends to appear near the boundary between magnetic and nonmagnetic phases.[12] Considering the ferromagnetic/antiferromagnetic ordering structure (Fig. 1), in which the main conducting Te sheet is sandwiched by two FM CeTe 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. 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 displaying 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 suggest that the magnetism is unlikely to be a primary source of SC pairing mechanism.

From the sensitive dependence of SC to δ, 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's). 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 sixfold CDW order. The main difference between TMD 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 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.[7] Also the piezoelectric coupling is, in general, unlikely in metals.[13] Therefore, the application of the pairing theory for TMD to our case is not straightforward. However, on general grounds 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.[12] (2) The tunneling data from Ref. 7 show that the zero-bias conductance increases below T_{SRF}, indicating that DOS increases due to the magnetic ordering. As for the role of pressure, the *c*-axis lattice distance is crucial to determine the actual T_c. Because the overlap along the *c* axis is so small (as one can see the *c*-axis resistivity compared with the in-plane resistivity), small displacement along the *c* axis can produce large changes in the hybridization matrix element. Thus, we believe that the application of pressure changes the interlayer coupling by reducing the *c*-axis lattice distance, as found in many other layered superconductors such as TMD and high-T_C superconductors.[4,14] Figure 4 shows that the SC, coexisting with magnetism, abruptly appears at $T_c = 2.7$ K with pressures as low as 2 kbar. As anticipated from Fig. 4, we cannot rule out that the superconducting phase appears sharply below 2 kbar.

To conclude, we report the superconducting transition at $T_c = 2.7$ K in $CeTe_{1.82}$ under pressure ($P > 2$ kbar). This T_c is remarkable to be high among *f*-electron systems. $CeTe_{2-\delta}$ displays various collective states, CDW (~ 1000 K), SRF (~ 6 K), and AFM (~ 4.3 K) at ambient pressure, and finally it shows a SC transition for $\delta = 0.18$ under pressure. The pressure is required to become superconducting as a result of the interlayer coupling enhanced by decreasing the interlayer distance along the *c* axis. Combining available data and phase diagram, we suggest that the primary possible 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, leading to SC.

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