

Exchange field enhanced upper critical field of the superconductivity in compressed antiferromagnetic EuTe_2

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Understanding the interplay between superconductivity and magnetism has been a long-standing challenge in condensed matter physics. Here we report high pressure studies on the C-type antiferromagnetic semiconductor EuTe_2 up to 36.0 GPa. A structural transition from the $I4/mcm$ to the $C2/m$ space group is identified at -16 GPa. Superconductivity is observed above -5 GPa in both structures. In the low-pressure phase, magnetoresistance measurements reveal strong couplings between the local moments of Eu^{2+} and the conduction electrons of Te 5p orbits. The upper critical field of superconductivity is well above the Pauli limit. While EuTe_2 becomes nonmagnetic in the high-pressure phase and the upper critical field drops below the Pauli limit. Our results demonstrate that the high upper critical field of EuTe_2 in the low-pressure phase is due to the exchange field compensation effect of Eu^{2+} and the superconductivity in both structures may arise in the framework of the Bardeen-Cooper-Schrieffer theory.

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Superconductivity in conventional Bardeen-Cooper-Schrieffer (BCS) superconductors arises from electron-lattice interaction without the involvement of magnetism¹. Below the superconducting (SC) transition temperature, electrons form coherent spin singlet cooper pairs that can be suppressed by a Pauli-limited magnetic field. In contrast, one of the hallmarks of unconventional superconductivity is the interplay between magnetism and superconductivity. For example, superconductivity in copper oxide and iron-based high-temperature superconductors occurs near long-range magnetic order where the $3d$ electrons of the transition metals across the Fermi level contribute to both the magnetic correlations and superconductivity^{2–6}. In some unconventional superconductors, electron pairing forms spin-singlet and the upper critical field needed to suppress superconductivity is also Pauli limited^{2–6}. For unconventional superconductivity with an upper critical field exceeding the Pauli limit, such as recently discovered UTe_2 ^{7,8}, electron pairing is believed to be spin triplets instead of singlets. In both spin-singlet and spin-triplet superconductors, magnetic fluctuations play an important role in the formation of cooper pairs as evidenced by the neutron spin resonance from inelastic neutron scattering spectra^{6,9}.

Although the mechanism of superconductivity for conventional and unconventional superconductors may be fundamentally different, both superconductors can host magnetic local moments not directly associated with SC layers. For example, in a class of iron-based superconductors consisting of Eu^{2+} , the $4f$ electrons with spin $S=7/2$ could form an antiferromagnetic (AFM) or ferromagnetic sublattice coexisting and interacting with the magnetic sublattice of Fe^{10,11}. However, the localized magnetism of Eu^{2+} does not interplay with the superconductivity seriously. For BCS superconductors such as $\text{RNi}_2\text{B}_2\text{C}$ series ($R = \text{Y, Er, Ho, etc}$)⁹, the interplay between the magnetic order of the rare earth layers and superconductivity can dramatically affect the physical properties of the system including the upper critical field needed to suppress superconductivity.

Previously, our group reported an antiferromagnetically colossal magnetoresistance EuTe_2 with a Néel temperature of $T_N=11.4\text{K}$ and a thermal-activation gap of 16.24meV at atmospheric pressure¹². The magnetic field drives polarization of the local moments of Eu^{2+} and results in the reconstruction of the Te $5p$ orbitals^{12,13}. It is interesting to explore the pressure effect on the small gap colossal magnetoresistance compound and investigate the interplay of the Eu^{2+} local moments with itinerant electrons of Te. Based on previous studies on CrSiTe_3 , WTe_2 ,

EuIn_2As_2 , and EuSn_2As_2 , superconductivity may emerge and the valent state transition from Eu^{2+} to Eu^{3+} may occur for EuTe_2 under pressure^{14–17}. Very recently, a high-pressure study on EuTe_2 up to 12.0GPa indeed reveals superconductivity and suggests the SC pairing mechanism is exotic¹⁸.

Here, we present comprehensive experimental and theoretical investigations on EuTe_2 under pressure up to 36.0GPa . Neutron diffraction measurements demonstrate EuTe_2 exhibits a C-type AFM order below T_N . A pressure-induced structural transition at $\sim 16\text{GPa}$ is discovered. In the low-pressure (LP) phase, the C-type AFM transition temperature T_N increases due to the enhancement of the magnetic exchange interactions of the compressed lattice. The thermal activation gap E_a is closed progressively, and superconductivity emerges above 5.0GPa . The SC transition temperature T_{cs} spanning between $3\sim 6\text{K}$ in the pressure range of $5\sim 27\text{GPa}$ is irrespective of the structural transition and magnetism. While the upper critical field $\mu_0 H_{c2}$ for the superconductivity of the AFM LP phase is significantly larger than that of the superconductivity of the nonmagnetic high pressure (HP) phase. The $\mu_0 H_{c2}$ is affected by the microscopic spin texture of the Eu sublattice. The highest $\mu_0 H_{c2}$ is estimated to be 21.6T for the spin-flipped state at 7.0GPa . The ultra-high $\mu_0 H_{c2}$ could be understood by the compensation effect of the exchange field of Eu^{2+} , the so-called Jaccarino and Peter mechanism¹⁹. Our results, therefore, establish the pressure-temperature phase diagram of EuTe_2 and demonstrate the interesting interplay mechanism between Eu magnetic order, superconductivity, and pressure-induced structural lattice distortion.

Results

High-pressure structure. Figure 1 displays the in situ high-pressure synchrotron powder x-ray diffraction (XRD) patterns of EuTe_2 up to 36.0GPa at room temperature and the refined crystal structures below 15.9GPa and above 17.9GPa , defined as the LP phase and HP phase, respectively. The LP phase can be indexed by the tetragonal $I4/mcm$ space group (No. 140), identical to the ambient pressure crystal structure. The divalent europium is coordinated by eight nearest-neighbor tellurium ions¹². The edge-sharing octagonal units form the layers of the tetragonal crystal structure as shown in Fig. 1c.

In terms of the diffraction peaks changed under pressure, an obvious structural phase transition between 15.9 and 17.9GPa could be identified. We conducted an extensive search on the HP structure of EuTe_2 in the pressure range of $0\sim 25\text{GPa}$ via the

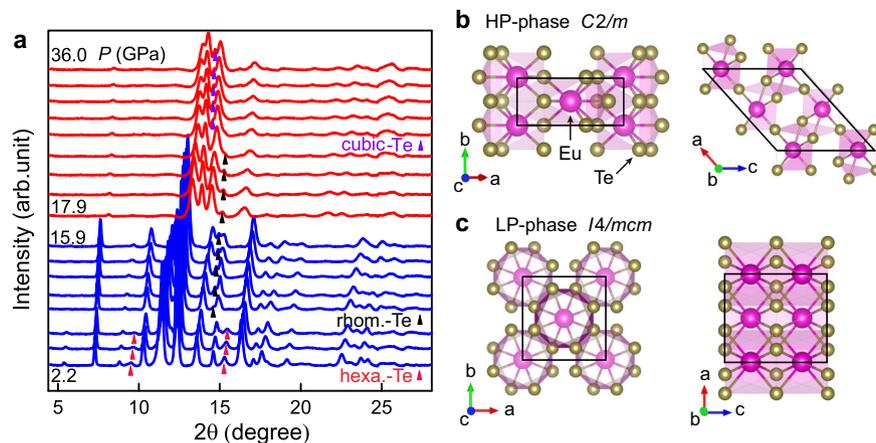


Fig. 1 X-ray diffraction (XRD) patterns and refined structures of EuTe_2 under pressure. **a** High-pressure XRD patterns of EuTe_2 from 2.2 to 36.0GPa with an x-ray wavelength of 0.6199\AA . The XRD patterns of the LP phase are in blue and that of the HP phase are in red. Tellurium undergoes two structural phase transitions within the measured pressure. The peaks from the Te impurity are marked by the triangles. **b** Crystal structures of the high pressure (HP) phase and **(c)** the low pressure (LP) phase of EuTe_2 .

CALYPSO method^{20–22}. The monoclinic $C2/m$ (No. 12) structure turns out to be a possible candidate for the HP phase at 17.9 GPa. Thus, we refined the experimental XRD pattern at 17.9 GPa by the Rietveld method through the TOPAS-Academic software²³. The $C2/m$ structure matches the XRD pattern of the HP phase well, as shown in Supplementary Fig. 1. Figure 1b shows the structure of the HP phase. The europium ions retain the eight-coordination but there is a significant deformation of the octagonal unit. This coordination unit exists in the compounds of Eu_3S_4 at atmospheric pressure, confirming that it is a stable coordination structure for europium chalcogenide²⁴. Sulfur has a smaller ionic radius than tellurium, close to pressurized tellurium.

For the HP phase, slip occurs between the adjacent layers compared with the LP phase. As shown in Fig. 1b, c, the unit cell volume decreases sharply at the pressure-induced structural transition from 317.157(9) \AA^3 at 15.9 GPa to 262.524(3) \AA^3 at 17.9 GPa, which may be accompanied by the valent state transition from Eu^{2+} to nonmagnetic Eu^{3+} . The diffraction peaks and structural transitions of Te impurity can be observed in Fig. 1a^{25,26}. The refined XRD patterns and structural parameters for 9.7 and 17.9 GPa are shown in Supplementary Note 1.

High-pressure electrical and magnetic properties. To investigate the electrical properties of EuTe_2 under pressure, we performed electrical transport measurements below 27.7 GPa. Figure 2a shows the temperature dependence of the resistance at various pressures, revealing semiconducting to metallic and SC transitions. Resistance as a function of pressure for selected temperatures is presented in Fig. 2b. The magnitude of the resistance decreases as pressure increases. The upturn in resistance at low pressure may be attributed to the scattering of conduction electrons by local moments of the Eu^{2+} ions. An abrupt drop in resistance appears between 14.7 and 16.2 GPa, consistent with the structural transition between 15.9 and 17.9 GPa. Thus, the

structural transition pressure should occur at ~ 16.0 GPa. The resistance above ~ 50 K in Fig. 2a is fitted to the thermal activation-energy model $\rho(T) = \rho_0 \exp(E_a/k_B T)$, where ρ_0 is a prefactor, E_a is the thermal activation gap, and k_B is the Boltzmann constant. The gap of 16.24 meV for EuTe_2 at ambient pressure is gradually closed by pressure, as shown in Fig. 2c and Supplementary Note 2. The evolution of the carriers against pressure at 10 K is also investigated by the Hall resistance measurements. The Hall coefficient remains positive, revealing that the majority of carriers are holes (Supplementary Note 3). The determined density of holes shows an abrupt enhancement across the structural transition similar to the observation in EuSn_2As_2 ¹⁷.

To elucidate the magnetic state of the HP phase, we conducted systematic magnetoresistance (MR) measurements against temperature and pressure. At ambient pressure, EuTe_2 shows colossal negative MR. Under pressure, the semiconducting gap is decreased, the resistance without a magnetic field becomes much smaller and the MR is suppressed accordingly (Supplementary Note 4). The integrated MRs (defined as $\text{MR} = (\rho_H - \rho_0)/\rho_0 \times 100\%$) over the magnetic fields from -10 to 10 T as presented in Fig. 2d decrease as pressure and temperature, diminishing gradually above 16.2 GPa. The abrupt decrease in MR suggests that the HP phase is not magnetically ordered.

Figure 3 shows the resistance in a smaller temperature range as a colormap on a logarithm scale. The T_N of the AFM transition and T_c of the SC transition under pressure could be identified from the resistance (Supplementary Note 5 and Note 6). Upon increasing pressure, the derived T_N s increase from 11.4 K at ambient pressure to 16.7 K at 8.0 GPa. The superconductivity appears at 4.9 GPa with a T_c of 3.2 K, defined by the intersection of the tangent to the resistance curve during the transition process and the straight line of the normal state above the SC transition. The T_c reaches a maximum of 6.1 K at 7.0 GPa and decreases smoothly afterward across the structural transition,

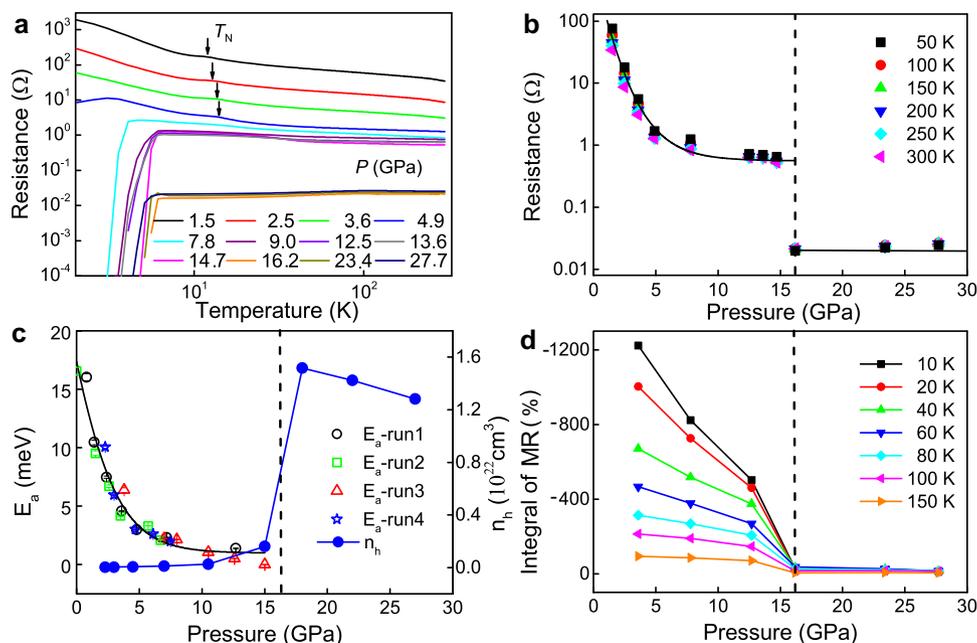


Fig. 2 Electrical transport properties under pressure. **a** Temperature dependence of the resistance upon pressures up to 27.7 GPa. The Néel transition temperatures (T_N s) are labeled on low-pressure curves. **b** Pressure dependence of the resistance at various temperatures up to 300 K shown on a logarithm scale. **c** Thermal-activation gaps (E_a s) derived from fittings of the resistance curves within the temperature range from 60 to 300 K using $\rho(T) = \rho_0 \exp(E_a/k_B T)$, where k_B is the Boltzmann constant. Different shapes of data points are measured on different samples. The black solid line is a guide to the eyes. The right axis represents the scale of carrier density. Error bars originating from the fitting process are smaller than the data points. **d** Integrals of magnetoresistance (MR) over the magnetic fields from -10 to 10 T as a function of pressure for selected temperatures. The dashed lines in (b–d) at 16.0 GPa mark the pressure of the structural transition.

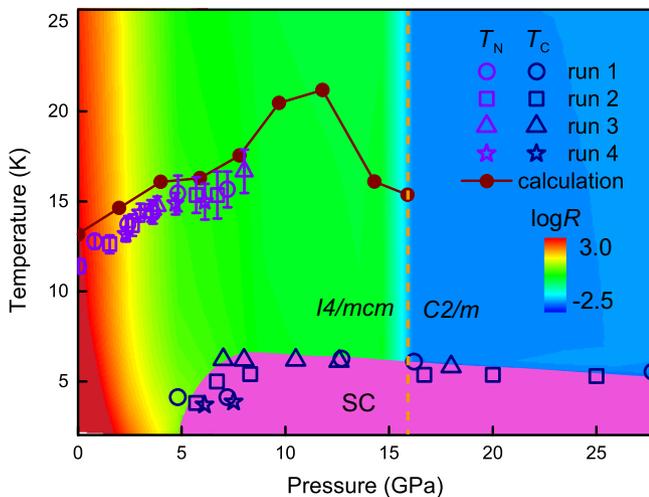


Fig. 3 A phase diagram of magnetism and superconductivity under pressure. The antiferromagnetic transition temperature T_N and superconducting (SC) transition temperature T_c against pressure. The filled circles are calculated T_N s. The color represents different resistance on a logarithm scale. Different shapes of data points are obtained from different measurements. The errors of the T_N s are estimated from resistance measurements.

indicating that the AFM order and spin fluctuations of Eu^{2+} have not contributed to the cooper pairing mechanism directly.

At ambient pressure and low temperature, the calculated energy difference between the *A*-type AFM and the *C*-type AFM is almost neglectable (about 1.5 meV per Eu)¹². Neutron diffraction measurements were employed to distinguish the two magnetic structures. Although the neutron absorption from Eu atoms is serious, the magnetic reflections associated with the *C*-type AFM are observed unambiguously (Supplementary Note 7). To understand the underlying mechanism for the enhanced T_N in compressed EuTe_2 , we investigate its exchange couplings based on the following spin model:

$$H = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + A \sum_i (\mathbf{S}_i^z)^2. \quad (1)$$

Considering the small gap of EuTe_2 , six nearest-neighbor (NN) Heisenberg exchange couplings are considered. In Eq. (1), A is the single-ion magnetic anisotropy parameter. For EuTe_2 at ambient pressure, our density functional theory (DFT) calculations show that it exhibits a *C*-type AFM ground state with a small gap of 18 meV and out-of-plane magnetic easy axis, consistent with our neutron scattering measurements. Our Monte Carlo simulations reveal the T_N is 13.17 K close to previous studies¹². Under pressure, both the DFT calculations and Monte Carlo simulations indicate that the ground state is also the *C*-type AFM order. Four NN exchange couplings are strengthened obviously in regard to ferromagnetic ($J < 0$) or antiferromagnetic ($J > 0$) terms up to 11.8 GPa (Supplementary Table 2). This is understandable because the distances between the Eu^{2+} ions decrease under pressure. Correspondingly, the calculated T_N s increase from 13.17 to 21.21 K at 11.8 GPa as shown in Fig. 3. For higher pressures near the structural transition, the Ruderman-Kittel-Kasuya-Yosida interaction may involve and lower the third and fourth NN exchange couplings, resulting in the decrease of the T_N .

Spin state transitions and high upper critical field. Superconductivity emerges in both the LP and HP phases, which have distinct structures and magnetic ground states. To explore the

role of the local moments of Eu^{2+} in superconductivity, we conducted resistance measurements at 7.0 and 18.0 GPa. Figure 4a, b shows the resistance as a function of the magnetic field and temperature at 7.0 GPa. A color map of the resistance is plotted in Fig. 4c. The *C*-type AFM structure of EuTe_2 at ambient pressure undergoes a spin-flop transition at ~ 3.0 T and a spin-flip transition at ~ 8.0 T¹². The magnetic fields for the spin-flop and spin-flip transitions below the T_N of 15.6 K at 7.0 GPa are extracted from the resistance in Fig. 4a, yielding 5.5 T for the spin-flop and 12.5 T for the spin-flip transitions at 5 K (Supplementary Note 8). The increased magnetic fields compared with that at ambient pressure are proportional to the increase of T_N . The SC transition temperatures T_c s at 7.0 GPa are determined from the resistance shown in Fig. 4b and displayed in Fig. 4c (Supplementary Note 9). We find the $\mu_0 H_{c2} - T_c$ relation does not follow a simple Ginzburg-Landau (GL) formula, $\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0)[1 - (T/T_c)^2]$. The experimentally determined T_c s against magnetic field could be separated into three segments, coincident with the AFM, spin flop, and spin flipped magnetic states. The $\mu_0 H_{c2}$ s for the spin flop and spin flipped states are well above the Pauli limit of $\mu_0 H_{c2} = 1.84 \times T_c = 11.2$ T, where T_c is 6.1 K at 7.0 GPa and zero field²⁷. We compare the resistance under various magnetic fields at 18.0 GPa in Fig. 4d. The color-map of resistance suggests that the HP phase is nonmagnetic. The $\mu_0 H_{c2}$ can be described by a single GL formula with the $T_c = 5.5$ K and $\mu_0 H_{c2} = 6.15$ T within the Pauli limit of 10.12 T.

Discussion

The superconductivity in EuTe_2 arising from Te impurity can be excluded because of the high upper critical field^{28,29}. In some compounds consisting of Te, the $\mu_0 H_{c2}$ could achieve the magnitude of several teslas, such as CrSiTe_3 , WTe_2 , HfTe_5 , Bi_2Te_3 , and CsBi_4Te_6 , where the normal states are not magnetically ordered^{14,15,30–32}. The $\mu_0 H_{c2}$ s of these compounds are still below the Pauli limit. UTe_2 superconducts below 1.6 K at ambient pressure with an ultra-high $\mu_0 H_{c2} > 45$ T³³. The *5f* electrons of uranium cross the Fermi level and contribute to the magnetism and superconductivity directly, resulting in the heavy Fermi property and possible triplet pairing mechanism^{34,35}.

Figure 5 shows the field dependence of T_c s in pressurized EuTe_2 . The $\mu_0 H_{c2}$ s are fitted to the experimentally determined AFM, spin flop, and spin flipped states to the GL formula, resulting in the upper critical fields of 10.1, 16.2, and 21.6 T, respectively. The Werthamer-Helfand-Hohenberg (WHH) formula of $\mu_0 H_{c2}(T) = -0.69 \times dH_{c2}/dT|_{T_c} \times T_c$ is also adopted to estimate the upper critical fields for the three distinct magnetic states, resulting in $\mu_0 H_{c2}$ s of 10.7, 15.9, and 20.8 T, respectively^{36,37}. The high $\mu_0 H_{c2}$ s for the spin flop and spin flipped states may be attributed to the compensation effect of the exchange field H_J produced by the local moments of Eu^{2+} . We note for the spin-flipped state the direction of H_J is antiparallel to the direction of magnetic moments of Eu^{2+} on the Te sites. In this case, the net magnetic field H_T acting on the conduction electrons is $H_T = H_{c2} - |H_J|$ ³⁸. As the Jaccarino-Peter mechanism, AFM spins do not contribute to the exchange field. The spin-flipped state with fully polarized moments of Eu^{2+} has the maximum H_J . If the sign of the coupling between the local spins and conduction electron spins is negative, the measured $\mu_0 H_{c2}$ should be larger than the Pauli limit¹⁹. As for $\text{Eu}_{0.75}\text{Sn}_{0.25}\text{Mo}_6\text{S}_{7.2}\text{S}_{0.8}$, an applied magnetic field can progressively tune the compound from SC to normal, to SC again, and finally back to the normal state below 1 K³⁹. To estimate the Pauli limit critical field H_p and H_J of $\text{Eu}_{0.75}\text{Sn}_{0.25}\text{Mo}_6\text{S}_{7.2}\text{S}_{0.8}$ for the second SC phase with the lower and upper $\mu_0 H_{c2}$ s at 4 and 22 T, we have the constraints of: (i) $4\text{ T} - \mu_0 H_J = -\mu_0 H_p$, and (ii)

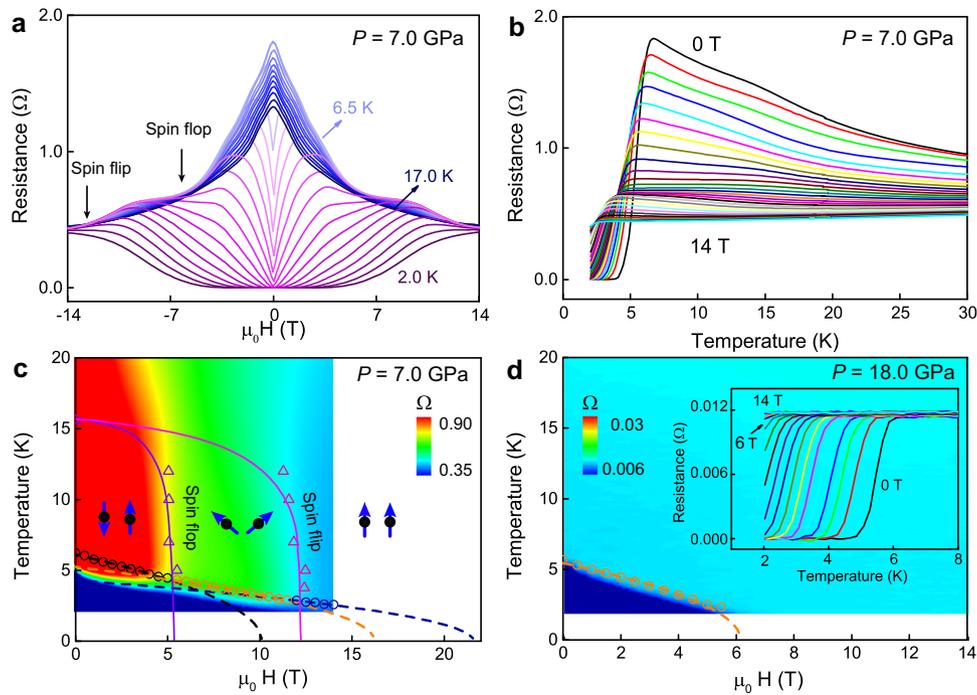


Fig. 4 Superconductivity and magnetism under pressure. **a** Magnetic field dependence of the resistance at 7.0 GPa and selected temperatures from 2.0 to 17.0 K. **b** Resistance from 2.0 to 30.0 K in different magnetic fields from 0 to 14.0 T measured every 0.5 T at 7.0 GPa. **c** A phase diagram with the antiferromagnetic order, spin flop, spin flip, and superconducting transitions. The solid lines are a guide for magnetic transitions. The dashed lines are the Ginzburg-Landau formula fittings to the segments of the superconducting transition temperatures. The color represents the magnitude of the resistance in **(b)** on a linear scale from 0.35 to 0.9 Ω . The magnetic fields of spin flop and spin-flip transitions are obtained from the magnetic field dependence of resistance in **(a)**. **d** Phase diagram of the superconductivity against the magnetic field at 18.0 GPa. The inset is the resistance curves measured on selected magnetic fields. The color represents the magnitude of the resistance in the inset of **(d)** on a linear scale from 0.006 to 0.03 Ω . The dashed line is fitting to the Ginzburg-Landau formula.

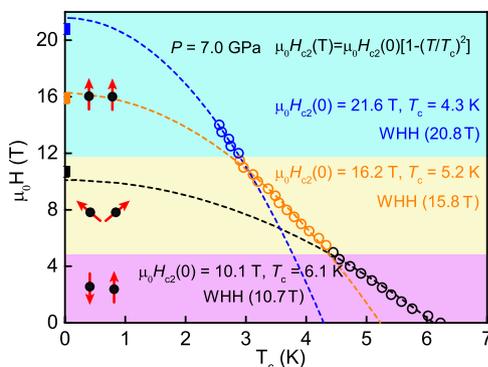


Fig. 5 Ginzburg-Landau (GL) fittings of the upper critical field $\mu_0 H_{c2}$ s at 7.0 GPa. The circles are superconducting transition temperatures T_c s of EuTe_2 determined from resistance at 7.0 GPa. The pink, yellow, and blue regions correspond to the antiferromagnetic order, spin flop, and spin flipped states. The black, orange, and blue dashed lines represent corresponding GL fittings. The T_c s, fitted $\mu_0 H_{c2}$ s using the GL formula, and the Werthamer-Helfand-Hohenberg formula for each magnetic state has been labeled.

$22\text{ T} - \mu_0 H_J = \mu_0 H_P$. The $\mu_0 H_J$ and $\mu_0 H_P$ with values of 13 and 9 T could be derived, respectively. If we assume the H_J is comparable with that of EuTe_2 , the $\mu_0 H_{c2}$ for the spin flipped state in the LP phase of EuTe_2 should be $10.1 + 13 \approx 23$ T, close to the fitted $\mu_0 H_{c2}$ of 21.6 T from the GL formula. Through DFT calculations, the localized Eu 4*f* electrons reside ~ 1.25 eV below the Fermi level for the LP phase, while the Te 5*p* electrons crossing the Fermi level involve SC cooper pairing (Supplementary Note 10), consistent with the Jaccarino-Peter mechanism.

In summary, we have studied the structural and electronic transport properties of EuTe_2 under pressure. EuTe_2 shows a SC transition above 5 GPa with a maximum T_c of 6.1 K at 7.0 GPa and a structural transition at 16 GPa. The transition temperature of the C-type AFM order is enhanced in compressed EuTe_2 due to the increase of the magnetic exchange interactions. In the LP phase, superconductivity coexists with the AFM, spin flop, and spin flipped states. However, the electronic states of Eu^{2+} are well below the Fermi level and do not involve cooper pairing directly. The local moments of Eu^{2+} in the spin flop and spin flipped states produce an exchange field, compensating with the external field and resulting in an ultra-high upper critical field that is larger than the Pauli limit. The HP phase is nonmagnetic and the $\mu_0 H_{c2} - T_c$ relation could be described by the GL formula with the $\mu_0 H_{c2}$ within the Pauli limit. Our results establish that EuTe_2 is a pressure-induced superconductor with a high upper critical field which could be understood by the Jaccarino-Peter mechanism.

Methods

Single-crystal growth and neutron diffraction. Bulk single crystals of EuTe_2 were grown by the self-flux method. Pure Eu and Te were combined in the molar ratio of 1:10 and sealed in an evacuated quartz ampoule. The ampoule was slowly heated to 850 $^\circ\text{C}$ in 100 h and held for 75 h, then slowly cooled to 450 $^\circ\text{C}$ in 300 h. The shiny black single crystals of EuTe_2 were separated from the Te flux at 450 $^\circ\text{C}$. The structure of EuTe_2 was confirmed by single-crystal XRD.

The powder neutron diffraction experiments were carried out on the Xingzhi triple-axis spectrometer at the China Advanced Research Reactor⁴⁰. Powder samples were stuck on an aluminum foil uniformly with a hydrogen-free glue to reduce the absorption of Eu, then sealed in a cylindrical vanadium container and loaded into a closed cycle refrigerator that regulates the sample temperature from 3.5 to 300 K. A neutron velocity selector was used upstream to cleanly remove higher order neutrons for the incident neutron energy fixed at 16 meV.

High-pressure XRD. The in situ high-pressure synchrotron powder XRD patterns of EuTe_2 were collected at 300 K with an x-ray wavelength of 0.6199 Å on the Beijing Synchrotron Radiation Facility, Institute of High Energy Physics, Chinese Academy of Sciences. A symmetric diamond anvil cell with a pair of 300 μm diameter culets was used. A sample chamber with a diameter of 120 μm was drilled by laser in a pre-indenting steel gasket. The EuTe_2 single crystals were ground into fine powders and compressed into a pellet with an 80 μm diameter and 20 μm thickness. The pellet was loaded into the middle of the sample chamber and silicone oil was used as a pressure-transmitting medium. A ruby sphere was also loaded into the sample chamber and pressure was determined by measuring the shift of its fluorescence wavelength. The data were initially processed using *Dioplas*⁴¹ (with a CeO_2 calibration) and the subsequent Rietveld refinements were managed using *TOPAS-Academic*⁴².

High-pressure magnetic and electrical property measurements. Magnetic and electrical measurements were taken on a physical property measurement system (PPMS, Quantum Design). High-pressure electrical transport measurements of EuTe_2 single crystals were carried out using a miniature diamond anvil cell made from a Be–Cu alloy on a PPMS. Diamond anvils with a 400 μm culet were used, and the corresponding sample chamber (with a diameter of 150 μm) was made in an insulating gasket achieved by cubic boron nitride and epoxy mixture. NaCl powders were employed as the pressure-transmitting medium, providing a quasi-hydrostatic environment. The pressure was also calibrated by measuring the shift of the fluorescence wavelength of the ruby sphere, which was loaded in the sample chamber. The standard four-probe technique was adopted for these measurements.

Theoretical calculations. Our structure searching simulations are performed by the swarm-intelligence-based CALYPSO (Crystal structure AnaLYsis by Particle Swarm Optimization) method, which enables global minimization of energy surfaces by merging ab initio total-energy calculations²². The structure searching was carried out at pressures of 5, 15, and 25 GPa which covers the experimental pressure range. The simulation cell sizes of 1–4 formula units were set. The underlying ab initio structural relaxations were carried out using density functional theory within the Perdew–Burke–Ernzerhof exchange–correlation⁴³ as implemented in the Vienna ab initio Simulation Package (VASP) code^{44,45}.

DFT calculations are performed using the VASP at the level of the generalized gradient approximation^{43,46}. We adopted the projector augmented wave pseudopotentials and a plane-wave cutoff energy of 500 eV⁴⁴. The experimentally measured lattice constants are used in our calculations and the positions of all atoms are fully relaxed until the force on each atom is less than 0.01 eVÅ⁻¹. We use $U = 4.4$ eV for Eu^{2+} ions because of the strong correlation among f electrons. The T_N of the pressurized EuTe_2 is obtained through parallel tempering Monte Carlo (MC) simulations^{47,48}.

Data availability

The source data and related supporting information are available upon reasonable request from the corresponding author.

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Author contributions

M.W. and H.S. proposed and designed the research. H.S. carried out the high-pressure measurements and data analysis with the help of Y.H., C.H., N.L., M.H., L.L., H.L., Z.L., and M.L. Single crystals were synthesized by C.H. Theoretical calculations were carried out by L.Q., Y.Z., W.W., D.Y., and Y.H. Neutron diffraction measurements were

conducted by P.C. with the support of H.Z., H.W., and L.H. M.W., H.S., and P.D. wrote the paper with input from all co-authors. M.W. oversaw the project.

Competing interests

The authors declare no competing interests.

Additional information

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