

Enhanced Thermopower in a Magnetic Semiconductor EuTe_4 with Multiple Charge-Density-Wave Instabilities

Hidefumi Takahashi^{1,2,*}, Kiichiro Yoshida¹, Akitoshi Nakano³, and Shintaro Ishiwata^{1,2,†}

¹*Division of Materials Physics and Center for Spintronics Research Network, Graduate School of Engineering Science, The University of Osaka, Osaka 560-8531, Japan*

²*Spintronics Research Network Division, Institute for Open and Transdisciplinary Research Initiatives, The University of Osaka, Yamadaoka 2-1, Suita, Osaka, 565-0871, Japan*

³*Department of Physics, Nagoya University, Nagoya 464-8602, Japan*

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We investigate the layered magnetic semiconductor EuTe_4 , focusing on its intricate charge-density-wave (CDW) states near and above room temperature through single-crystal x-ray diffraction (XRD), magnetic, and thermoelectric measurements. The XRD measurement revealed that the CDW state inducing the polar lattice distortion persists even at 650 K, demonstrating its remarkable thermal stability. Notably, the Seebeck coefficient near room temperature reaches values exceeding $500 \mu\text{V K}^{-1}$. This large Seebeck coefficient, not fully captured by a simple band calculation, is comparable to those observed in heavy-electron semiconducting oxides, suggesting the importance of electron correlation and spin- and lattice instabilities. Furthermore, potentially reflecting the competition of two types of CDW state, the thermal conductivity near room temperature is as low as $0.02 \text{ W cm}^{-1} \text{ K}^{-1}$. As a result, the thermoelectric figure of merit zT reaches 0.22 at 460 K. These findings establish EuTe_4 as a compelling platform to explore novel types of thermoelectric materials with multiple electronic instability.

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I. INTRODUCTION

Low-dimensional materials exhibiting charge-density-wave (CDW) states have attracted considerable interest due to their rich interplay of electronic and structural properties, as well as their potential for advanced device applications [1]. Among these systems, transition-metal chalcogenides such as $1T\text{-TaS}_2$, $1T\text{-TiSe}_2$, and NbSe_3 have been studied as prototypical platforms for investigating CDW physics [1–5]. These materials exhibit a variety of intriguing phenomena, including metal-insulator transitions, nonlinear transport, and collective electronic behaviors driven by strong electron-lattice coupling. Furthermore, their tunability via external stimuli such as temperature, pressure, and chemical doping has opened avenues for potential applications in memory devices and sensors [6–10].

Recently, rare-earth tritellurides (RTe_3) have been extensively studied because of rich CDW states [11–13], which can be tuned through external perturbations such as chemical intercalation and applied pressure [14]. Importantly, increasing the atomic number of the rare-earth element in RTe_3 modifies the Fermi surface nesting, leading to the emergence of a second CDW state [15]. The coexistence or competition between distinct CDW states enriches the phase diagram, which includes a superconducting phase [16,17]. Additionally, the rare-earth elements induce complex spin textures through global and local mirror-symmetry breaking associated with the CDW state [15,18].

A europium tetratelluride EuTe_4 also exhibits unique CDW orders as shown in Fig. 1(a). Unlike its tritelluride counterparts, where partial Fermi surface nesting allows metallic conduction, EuTe_4 exhibits a fully gapped Fermi surface, rendering it semiconducting below the CDW transition temperature [19–22]. X-ray diffraction (XRD) measurements have confirmed the incommensurate nature of the CDW, characterized by a modulation vector $\mathbf{q} \sim 0.67\mathbf{b}^*$, while some experiments suggest the presence of multiple CDW states [22–26]. One of the remarkable features of EuTe_4 is an exceptionally large thermal hysteresis in a temperature range including room temperature, likely arising from the complexity of the CDW orders.

*Contact author: takahashi.hidefumi.es@osaka-u.ac.jp

†Contact author: ishiwata.shintaro.es@osaka-u.ac.jp

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Consequently, EuTe_4 demonstrates nonvolatile, optically and electrically tunable polar order in the CDW state at room temperature, highlighting its potential for applications in ultrafast electronic and photonic devices [27,28]. In addition, the magnetic Eu^{2+} ions provide an opportunity to study the coupling between the complex CDW states and magnetism [29,30].

Here we report on the structural, magnetic, and thermoelectric properties of EuTe_4 single crystals. XRD experiments reveal that the CDW state in EuTe_4 persists even above 400 K. Magnetic measurements indicate significant in-plane anisotropy, which reflects the lattice distortion with the polar lattice displacement along the a axis. Both electrical resistivity and thermoelectric power exhibit pronounced hysteresis over a broad temperature range, with the thermoelectric power reaching an exceptional value of $500 \mu\text{V K}^{-1}$ near and above room temperature. This behavior cannot be fully captured by simplified first-principles calculations, suggesting the pivotal role of many-body effects, including electron-electron interactions and magnetism. These results establish EuTe_4 as a compelling platform for studying the interplay between CDW order, magnetism, and thermoelectric phenomena.

II. METHOD

A. Crystal growth and transport measurement

Single crystals of EuTe_4 were grown by the Te flux method [19]. Eu lumps (99.9%) and Te granules (99.9999%) were mixed in a ratio of 1:15. The total weighed starting materials were sealed in an evacuated fused silica tube in a vacuum, followed by heating at 850°C for 2 days in a muffle furnace. The furnace was then slowly cooled to 430°C in 24 h, and was held at this temperature for 2 weeks, with subsequent decanting with the use of a centrifuge. The large single crystal shown at the bottom of Fig. 1(a) was used for the transport measurements. The crystal structure was identified by the synchrotron XRD measurements for single-crystal samples at BL02B1 in SPring-8. Magnetic properties, electrical resistivity, and the Seebeck coefficient (thermopower) below 400 K were measured with a magnetic property measurement system (MPMS; Quantum Design, Inc.). The resistivity was measured by a standard four-terminal method. Resistivity between 290 and 490 K was measured on a hot plate in air. The Seebeck coefficient was measured by a conventional steady-state method. Thermal conductivity was measured by a conventional four-terminal steady-state method with the use of a He closed-cycle refrigerator (from 10 to 470 K). The Seebeck coefficient was measured between 250 and 460 K with the use of the same refrigerator. Because of limitations in the temperature-control stability of the refrigerator, reliable data could be acquired only on the warming branch for $T \lesssim 300$ K and on the cooling branch for $T \gtrsim 300$ K consequently, the Seebeck

coefficient and thermal conductivity measured with use of the refrigerator are shown only for the warming run below approximately 300 K and for the cooling run above approximately 300 K. The composition of the single crystals was determined by energy-dispersive X-ray (EDX) spectroscopy using a JEOL JCM-7000 scanning electron microscope. EDX measurements were performed at different positions on the crystal surfaces within an instrument error of 1%–2% (for details, see Fig. S1 and Table S1, respectively, in Supplemental Material [31]). Eu tends to be deficient by less than 10% from the nominal composition (Eu:Te ratio 1:4). Thermogravimetry–differential thermal analysis (TG-DTA; STA-2500 Regulus made by NETZSCH) was performed with 8.2 mg of crushed single crystals in Al_2O_3 capsules.

B. Band calculations

The nonrelativistic bulk electronic structure calculations for EuTe_4 were performed within density functional theory with use of the Perdew-Burke-Ernzerhof exchange-correlation functional as implemented in the QUANTUM ESPRESSO code [38–41]. The projector augmented wave method was used to account for the treatment of core electrons [41]. The cutoff energy for plane waves forming the basis set was set to 50 Ry. The lattice parameters and atomic positions were taken from the single-crystal XRD experiment. The corresponding Brillouin zone was sampled with the use of a $11 \times 11 \times 5$ k -point mesh to calculate the plane-wave self-consistent field (PWscf), and a $30 \times 30 \times 30$ k -point mesh for normal states and a $10 \times 20 \times 30$ k -point mesh for the high-temperature CDW-I phase, which will be discussed later, as observed by single-crystal X-ray diffraction states to perform Fermi surface visualization by the FermiSurfer package [42]. We ignore the $\text{Eu } f$ orbitals since they are predicted to be more than 1.5 eV below the Fermi surface [32]. The Seebeck coefficient was calculated with BoltzTraP [43].

III. RESULTS AND DISCUSSION

A. Structural analysis and magnetic properties

Figure 1(c) shows the XRD patterns for the $(0KL)$ plane and Table I summarizes the structure parameters at 650 K [the XRD pattern for the $(HK0)$ plane at 650 K is shown in Fig. S2 in Supplemental Material [31]]. The analyzed structure is shown in Fig. 1(a) as the CDW-I state. It is noted that the space group of the CDW-I phase, $P2_1cn$ (no. 33), is a polar structure with no inversion symmetry. While previous studies have suggested a transition from the CDW state to the normal state near room temperature [19], the present measurement confirmed a nearly commensurate superlattice peak at around $(1, \pm 1/3, 0)$ corresponding to the previously reported threefold CDW state even at 650 K, as denoted by the green arrows.

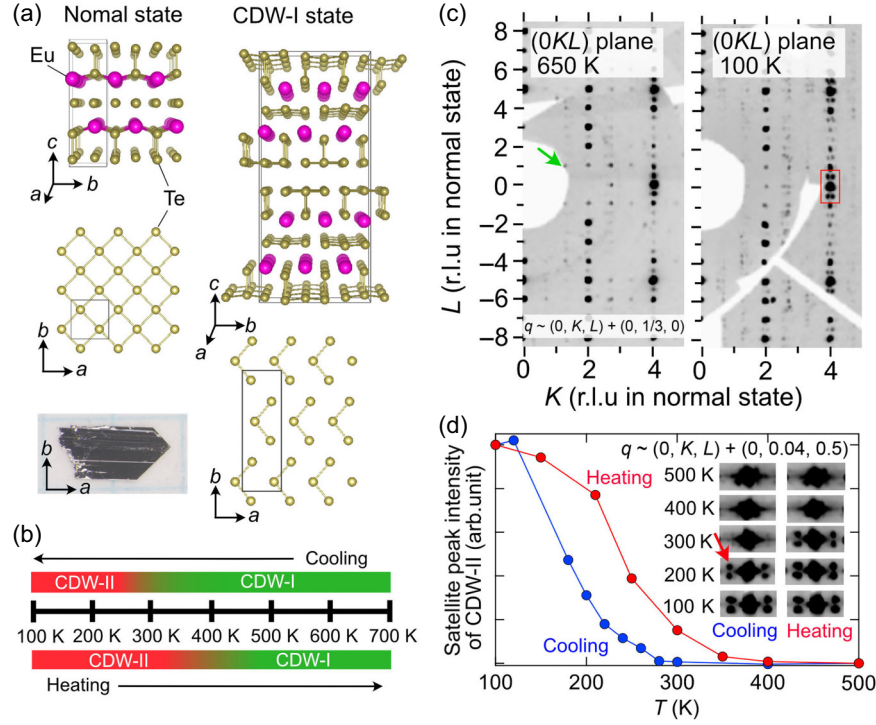


FIG. 1. (a) Crystal structures of EuTe₄ of the normal state and the CDW state reported in Ref. [19]. (b) Temperature-dependent stability of CDW states. (c) Single-crystal x-ray diffraction patterns for the (0KL) plane measured at 650 K (left panel) and 100 K (right panel). The green arrow identifies the CDW-I superlattice reflection, whereas a new peak ascribed to the CDW-II phase appears within the red-boxed region. (d) Enlarged view of the red-boxed region in (c) and the temperature dependence of the CDW-II superlattice reflection at $\mathbf{q} \sim (0, 4 \pm 0.04, 0.5)$.

This result indicates the realization of the nearly threefold CDW state at far above room temperature. Although the modulation is incommensurate, we performed structural

refinement assuming a commensurate threefold modulation along the b axis as an average structure, which effectively captures the lattice distortion associated with

TABLE I. Refined structural parameters for EuTe₄ at 650 K determined by single-crystal synchrotron X-ray diffraction. The space group is $P2_1cn$, and the lattice parameters obtained are $a = 4.5861(3)$ Å, $b = 13.9979(11)$ Å, and $c = 31.653(13)$ Å. The number of unique reflections for the refinement within the resolution limit $d > 0.5$ Å is 4774. The R factor and goodness of fit obtained are 6.87% and 1.017, respectively.

	Site	x	y	z	Occupancy	U_{iso} (Å ²)
Eu1	4a	0.5794(4)	-0.74725(7)	0.16522(3)	1	0.0392(2)
Eu2	4a	-0.4215(4)	-0.41333(5)	0.16603(4)	1	0.0390(3)
Eu3	4a	0.0720(4)	0.08217(6)	-0.16550(4)	1	0.0399(3)
Te1	4a	0.0317(10)	0.07782(8)	-0.05331(5)	1	0.0490(6)
Te2	4a	0.0603(9)	-0.25063(12)	-0.05371(4)	1	0.0552(4)
Te3	4a	0.6303(9)	-0.41740(7)	0.05376(5)	1	0.0415(4)
Te4	4a	0.5778(6)	-0.08540(6)	-0.14340(5)	1	0.0359(3)
Te5	4a	0.5775(8)	-0.24707(9)	0.25046(3)	1	0.0519(4)
Te6	4a	0.0880(8)	-0.41452(7)	0.25013(5)	1	0.0504(6)
Te7	4a	0.0768(6)	-0.24750(9)	0.14213(3)	1	0.0362(2)
Te8	4a	0.0784(5)	-0.58053(6)	0.14320(5)	1	0.0350(3)
Te9	4a	0.5680(9)	-0.58143(8)	0.25013(5)	1	0.0526(5)
Te10	4a	0.0324(10)	-0.57764(8)	0.05485(5)	1	0.0492(5)
Te11	4a	0.6310(10)	-0.08181(8)	-0.05497(5)	1	0.0415(4)
Te12	4a	0.0613(9)	-0.24929(11)	0.05289(4)	1	0.0518(4)

the high-temperature CDW phase. Some previous studies also suggest that CDW transition occurs above 600 K [24], and the incommensurate modulation wave vector $\mathbf{q} \sim 0.67\mathbf{b}^*$ persists at 300 K [20,21,23,42]. Angle-resolved photoemission spectroscopy (ARPES) measurements also show that the CDW gap is already open at around 400 K or even higher [20]. We have also measured XRD patterns below 400 K and obtained complex diffraction patterns, reflecting the influence of different CDW phases [22,24,25] or surface domains [20]. Therefore, the precise structural analysis has not been achieved below 400 K. Figure 1(c) compares the single-crystal X-ray diffraction patterns for the (0KL) plane recorded at 650 and 100 K. The superlattice reflection corresponding to CDW-I indicated by the green arrow at 650 K becomes substantially more intricate at 100 K, most likely owing to domain formation and the coexistence of competing CDW orders; in addition, a new, well-defined reflection appears within the region delineated by the red rectangle. Figure 1(d) shows an enlarged view of this region and the temperature evolution of the associated superlattice peak at $\mathbf{q} \sim (0, K, L) + (0, 0.04, 0.5)$. Upon both cooling and subsequent heating, this peak develops a pronounced hysteresis that mirrors the hysteresis observed in the temperature dependences of the electrical resistivity and Seebeck coefficient discussed below. These observations indicate that the lattice modulation in the CDW-II phase can be associated with this superlattice reflection.

Figure 1(b) illustrates the temperature evolution of the CDW states. The high-temperature CDW-I state [$\mathbf{q}_{\text{CDW}} \sim (0, 1/3, 0)$] changes to the low-temperature CDW-II state [$\mathbf{q}_{\text{CDW}} \sim (0, 0.04, 0.5)$] at around 300 K. Here we define the CDW modulation vectors $\mathbf{q}_{\text{CDW}}$ with respect to the unit cell of the normal state shown in Fig. 1(a). A previous study suggests the possibility of a transition between the normal state and the CDW-I state at around 726 K on heating and at around 652 K on cooling [24]. To determine the stability of the crystals at high temperatures, we performed TG-DTA and measured XRD before and after the process, as shown in Fig. S3 in Supplemental Material [31]. The results revealed that the crystals decomposed at around 700 K during the heating process, and an anomaly was observed at around 660 K during the cooling process, probably due to crystallization into Eu_3Te_7 [33]. In addition, the normal state structure could not be observed even with the single-crystal XRD measurements at temperatures above 650 K.

Figure 2(a) shows the temperature dependence of the magnetic susceptibility χ for H parallel to the a axis ($H\parallel a$), H parallel to the b axis ($H\parallel b$), and H parallel to the c axis ($H\parallel c$) at $H = 0.1$ T. The compound undergoes an antiferromagnetic (AFM) transition at 7 K (T_N), as observed in a previous study [19]. The magnetic susceptibility for $H\parallel a$ is significantly reduced below T_N as compared with that for $H\parallel b$ and $H\parallel c$ [20], indicating that

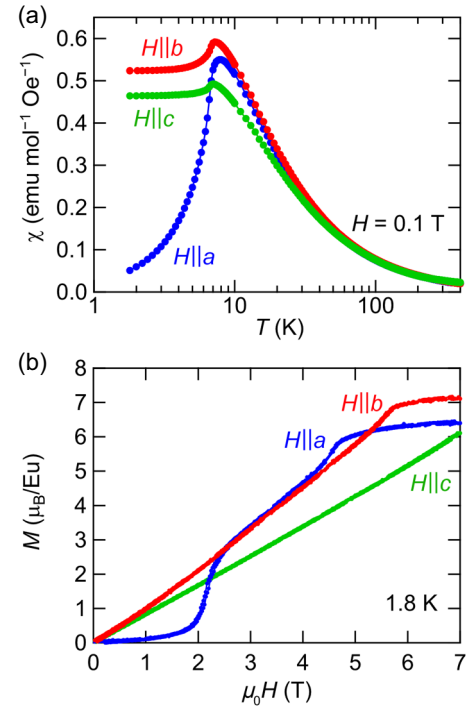


FIG. 2. (a) Temperature dependence of the magnetic susceptibility measured on zero-field cooling for $H\parallel a$, $H\parallel b$, and $H\parallel c$ at 0.1 T. (b) Magnetization as a function of the magnetic field for $H\parallel a$, $H\parallel b$, and $H\parallel c$ at 1.8 K, where negligible hysteresis is observed for all field orientations.

the spin moments are parallel or antiparallel to the a axis in the AFM phase. The spin anisotropy within the a - b plane in the AFM phase of EuTe_4 can be associated with the orthorhombic distortion caused by the CDW transition. In the case of the similar layered compound EuTe_2 , the spins are oriented along the c axis, consistent with the tetragonal symmetry [44]. In addition, the spin orientation in EuTe_4 can be influenced by the Dzyaloshinskii-Moriya interactions resulting from an inversion symmetry breaking with polarity along the a axis. The AFM nature is also confirmed by the Curie-Weiss fits for $H\parallel a$ in the range between 10 and 350 K, as shown in Fig. S4 in Supplemental Material [31], which resulted in a Weiss temperature θ_w of -2.5 K. The estimated effective magnetic moment, $7.32 \mu_B$, is close to the theoretical value for EuTe_4 with Eu^{2+} ($7.94 \mu_B$), as previously confirmed by an x-ray absorption experiment [23]. Here we assume that the valence of the Te ions in the Eu-Te slab is -2 , and that the Te monolayers and bilayers are neutral [23].

To further investigate the anisotropy in the magnetically ordered states, we measured the magnetic field dependence of the magnetization M for $H\parallel a$, $H\parallel b$, and $H\parallel c$ at 1.8 K, as shown in Fig. 2(b). For $H\parallel b$ and $H\parallel c$, the magnetization curves are linear to the field, and the former shows saturation at approximately 5.7 T. On the other

hand, the magnetization curve for $H \parallel a$ exhibits a sudden jump at approximately 2 T, which implies a spin-flop transition, and shows saturation at 4.5 T. The saturation field for $H \parallel a$ is lower than the others, indicating that the localized moment tends to align along the a -axis direction as suggested by the temperature-dependent magnetic susceptibility below T_N . Furthermore, although the magnetization exhibits negligible hysteresis over the entire field range, the magnetoresistance displays a pronounced hysteresis loop (see Fig. S5 in Supplemental Material [31]). We attribute this discrepancy to a field-induced subtle lattice distortion by the spin reconfiguration, which leaves the net magnetization almost unchanged yet produces the hysteresis of magnetoresistance, potentially associated with the multidomain state discussed below. These observations provide direct evidence for strong coupling among the spin, lattice, and charge modulation.

B. Thermoelectric properties

Figure 3(a) shows the temperature dependence of the electrical resistivity ρ for $I \parallel a$ and $I \parallel b$. A hysteresis is observed over a wide temperature range between 200 and 400 K measured with the MPMS, consistent with the findings of prior studies [19,20]. We repeated the measurement several times and consistently reproduced the same loop, indicating that extrinsic factors such as cracks in the crystal have a negligible influence on the result. The broad hysteresis loop suggests the emergence of distinct CDW states at lower temperature. Furthermore, the hysteresis loop remains unclosed even when the highest measured temperature is increased to 490 K by use of a hot plate in air, suggesting that the low-temperature CDW-II phase partially remains up to rather high temperatures, where the CDW-I phase is expected to be stable [23]. On the other hand, small hysteretic behavior can be found at around 100–200 K, indicating persistent effects of CDW domains at lower temperatures. In contrast to previous reports, we do not observe a pronounced increase in electrical resistivity at low temperatures characteristic of a semiconducting electronic structure. This discrepancy may arise from the different impurity levels in the Eu source: we used 99.9% pure Eu, whereas the earlier studies used 99.999% pure Eu. A 0.1% deviation in stoichiometry results in a change in carrier density of roughly 10^{18} – 10^{19} cm^{-3} , which is large enough to affect the low-temperature transport of narrow-gap semiconductors [45]. EDX measurements also suggest a few percent deficiency of Eu, which introduces hole carriers to the system. While resistivity exhibits minimal in-plane anisotropy near room temperature, a slight anisotropy is found at low temperatures, potentially reflecting a quasi-one-dimensional band structure, as previously indicated by ARPES measurements [20–22]. Additionally, a subtle anomaly near 70 K for the $I \parallel a$ orientation may correspond to the emergence of another distinct CDW phase,

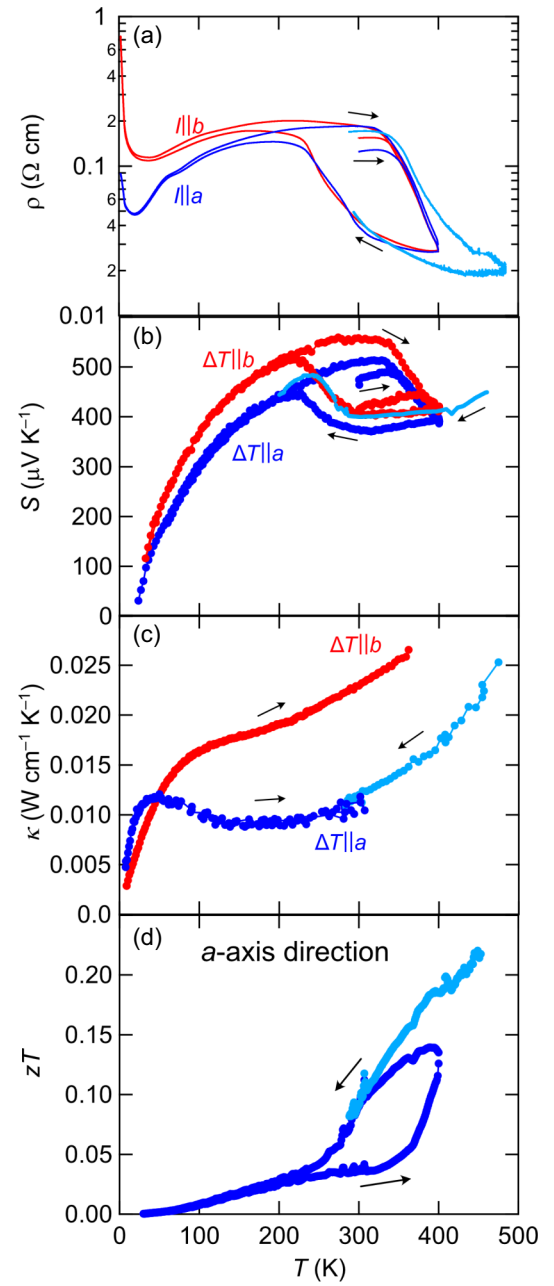


FIG. 3. Temperature dependence of (a) electrical resistivity for $I \parallel a$ and $I \parallel b$, (b) Seebeck coefficient for $\Delta T \parallel a$ and $\Delta T \parallel b$, (c) thermoelectric conductivity for $\Delta T \parallel a$ and $\Delta T \parallel b$, and (d) dimensionless figure of merit zT for $I \parallel a$ and $\Delta T \parallel a$.

as recently suggested in scanning tunneling microscopy studies [25].

To elucidate the impact of CDWs on the electronic properties, we investigated the temperature dependence of thermoelectric properties, as shown in Fig. 3(b). Whereas a similar pronounced hysteresis is observed over a broad temperature range, the hysteresis loop closes at around 230 K in the temperature dependence of the Seebeck

coefficient. Since the Seebeck coefficient is less sensitive to electron scattering as compared with electrical resistivity, the thermal hysteresis below 230 K in the resistivity can be ascribed to the domain dynamics, and the large thermal hysteresis at around 300 K in the Seebeck coefficient suggests a first-order transition between distinct electronic states with different CDW modulations; i.e., high-temperature CDW-I phase and low-temperature CDW-II phase. While previous studies have indicated the presence of multiple CDWs, the detailed structural nature of these phases remains unresolved.

Remarkably, the Seebeck coefficient at room temperature (approximately 300 K) reaches exceptionally high values of 550 and 400 $\mu\text{V K}^{-1}$ during heating and cooling processes, respectively. In the temperature range from room temperature to 460 K, where a large Seebeck coefficient is observed, electrical resistivity becomes as low as approximately 0.02 $\Omega\text{ cm}$ during the cooling process. Consequently, the present compound exhibits a relatively high power factor (PF; equivalent to S^2/ρ), reaching up to 10 $\mu\text{W K}^{-2}\text{ cm}^{-1}$ at 460 K, as shown in Fig. S6 in Supplemental Material [31]. This value is comparable to that of $\text{La}(\text{Fe}, \text{Co})_4\text{Sb}_{12}$, a well-known thermoelectric material, which exhibits a PF of approximately 6.66 $\mu\text{W K}^{-2}\text{ cm}^{-1}$ at 300 K [46]. Such a high PF suggests that EuTe_4 has potential as an efficient thermoelectric material in the temperature range above room temperature. To evaluate the dimensionless figure of merit zT , the temperature dependence of the thermal conductivity was measured, as shown in Fig. 3(c). The thermal conductivity is relatively low, approximately 0.01 $\text{W cm}^{-1}\text{ K}^{-1}$ at 300 K, especially for the temperature gradient along the a axis, and the peaky structure typically observed for clean semiconductors is absent as in the case of a glass. In sharp contrast, the rare-earth tritellurides RTe_3 ($\text{R} = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$) with CDW orders display a much larger lattice thermal conductivity of 0.04–0.08 $\text{W cm}^{-1}\text{ K}^{-1}$ (total $\kappa = 0.16\text{--}0.25\text{ W cm}^{-1}\text{ K}^{-1}$) and the conventional low-temperature upturn characteristic of crystalline solids [47]. The glasslike behavior found in EuTe_4 is therefore most plausibly attributed to a multidomain state generated by the competition of nearly degenerate CDW orders. This scenario is supported by the broad hysteresis observed in the temperature dependence of the resistivity and by scanning tunneling microscopy measurements that reveal pronounced CDW competition and domain formation at low temperatures [20,25].

The temperature dependence of zT for the a -axis direction is presented in Fig. 3(d). zT increases significantly above 250 K, reflecting the rapid decrease in electrical resistivity, and reaches a value of 0.22 at 460 K. This value is comparable to that of filled skutterudite antimonides and the transition-metal chalcogenide 1T-TiS_2 [46,48]. One of the key factors contributing to the observed high thermoelectric performance is the large thermopower of

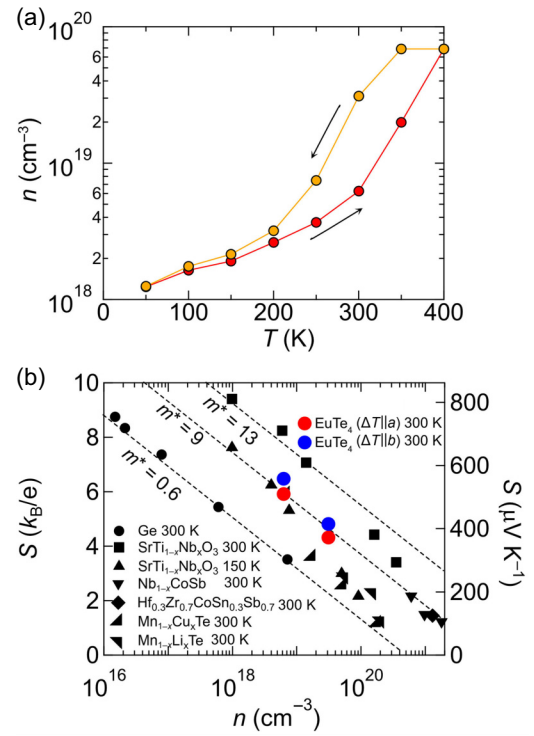


FIG. 4. (a) Temperature dependence of carrier concentrations n estimated from the Hall resistivity ($I||a, V||b, H||c$). (b) Seebeck coefficient S of EuTe_4 , p -type Ge, Nb-doped strontium titanate, half Heusler compounds, and (Cu,Li)-doped MnTe as a function of carrier concentration n at selected temperatures [50–54]. In all systems, S follows a $\ln(n^{-1})$ dependence, as expected by Eq. (4) in Ref. [50].

the system. This significantly large value (approximately 500 $\mu\text{V K}^{-1}$) suggests the presence of a gaplike electronic structure, as reported in ARPES studies [20], which is inconsistent with the first-principles band calculations as shown in Fig. S8 in Supplemental Material [31]. This large Seebeck coefficient is comparable to that of CoSb_3 (approximately 450 $\mu\text{V K}^{-1}$), studied as a promising thermoelectric material [49], where the large Seebeck coefficient is attributed to a low carrier concentration of approximately $4 \times 10^{17}\text{ cm}^{-3}$. However, as shown in Fig. 4(a), the carrier concentration n in EuTe_4 , derived from Hall resistivity measurements (see Fig. S7 in Supplemental Material [31]), reveals a much higher carrier concentration of approximately 10^{19} cm^{-3} at 300 K. For materials with similar carrier concentrations, typical Seebeck coefficients range from 200 to 300 $\mu\text{V K}^{-1}$ [50], suggesting that the large Seebeck coefficient of EuTe_4 cannot be explained by the conventional one-electron approximation.

In nondegenerate semiconductors with a carrier concentration less than 10^{20} cm^{-3} , the Seebeck coefficient S is generally expressed as a logarithmic function of the carrier concentration, as $S \sim \ln(n^{-1})$ [55]. The logarithmic function in Ref. [55] arises from the dependence of both the Seebeck coefficient and the carrier concentration

on the energy gap Δ in nondegenerate semiconductors, where the Seebeck coefficient scales as $S \sim \Delta/k_B T$, and the carrier concentration n follows an exponential behavior, $n \sim \exp(-\Delta/k_B T)$. Notably, the $S\text{-}\ln(n^{-1})$ trend observed in EuTe_4 deviates significantly from that of a typical semiconductor, Ge, shifting towards higher S values as shown in Fig. 4(b). This deviation suggests additional contributions such as increased electron mass, multiple Fermi pockets, magnetism, and structural fluctuations [34–37,56–60]. In particular, a moiré-like CDW structure has been proposed as the driver of the emergent semiconducting band structure [26]. If the observed deviations of the $S\text{-}\ln(n^{-1})$ trend from that of Ge were caused solely by an enhanced carrier mass, the effective mass (m^*) would need to be an order of magnitude larger than the free electron mass (m_0).

To understand the origin of this deviation, we compare EuTe_4 with other semiconductors exhibiting heavy carrier mass at room temperature, such as the SrTiO_3 system ($m^* \sim 9\text{--}13 m_0$) [55], several half Heusler compounds ($m^* \sim 4\text{--}6 m_0$) [51,52], and the (Cu, Li)-doped MnTe system ($m^* \sim 2\text{--}7 m_0$) [53,54]. These compounds fall along nearly the same straight line in Fig. 4(b), and their power factors lie in the same range ($1\text{--}30 \mu\text{W K}^{-2} \text{cm}^{-1}$) as that of EuTe_4 . Furthermore, the Fermi surface obtained from first-principles calculations in the CDW-I phase along with the band dispersions in Figs. S8 and S10 in Supplemental Material [31] reveals an essentially two-dimensional, multipocket band structure. ARPES confirms that EuTe_4 is semiconducting, with no Fermi surface at the Fermi level; nevertheless, at an energy of about -0.2 eV, the spectra display a two-dimensional, multipocket structure that closely matches the calculated Fermi surface [20,21]. This characteristic band structure just below the Fermi level may therefore be crucial for producing the unusually large Seebeck coefficient of EuTe_4 . In the case of MnTe, the power factor is likely enhanced further by paramagnon drag far above the Néel temperature ($T_N \sim 315$ K), reflecting the persistence of short-range antiferromagnetic correlations up to approximately 600 K. Because EuTe_4 orders antiferromagnetically only below $T_N \sim 7$ K, a comparable paramagnon contribution near room temperature is unlikely; instead, the material has competing CDW instabilities well above room temperature, and thus a phason-drag mechanism—the CDW analogue of magnon drag—may boost the Seebeck coefficient [61]. To confirm these possibilities as the origin of the exceptionally large thermopower of EuTe_4 , a thorough investigation of these competing CDW states is essential.

IV. CONCLUSIONS

In conclusion, we have successfully synthesized single crystals of EuTe_4 and conducted comprehensive investigations of its structural, magnetic, and thermoelectric

properties. Structural analysis revealed that the crystal exhibits a CDW state at a temperature as high as 650 K. Measurements of the magnetic and thermoelectric properties revealed in-plane anisotropy, associated with the polar lattice distortion and a large zT value of 0.22 at 460 K, reflecting a large Seebeck coefficient of $550 \mu\text{V K}^{-1}$ and a low thermal conductivity of $0.02 \text{ W cm}^{-1} \text{K}^{-1}$. Band structure calculations based on the CDW structure failed to reproduce the semiconducting gap and the observed large Seebeck coefficient. This discrepancy underscores the need for further studies on EuTe_4 , including detailed structural investigations of the CDW state and theoretical approaches incorporating many-body effects, such as magnetic interactions and electron-electron correlations, to fully elucidate the enhanced thermoelectric performance.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [62], embargo periods may apply.

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