Supplementary Material: Field-induced Metal-Insulator Transition in β -EuP₃

Guangqiang Wang,¹ Guoqing Chang,² Huibin Zhou,¹ Wenlong Ma,¹

Hsin Lin,³ M. Zahid Hasan,^{2,4} Su-Yang Xu,⁵ and Shuang Jia^{1,6,7,8,*}

¹International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, China

²Laboratory of Topological Quantum Matter and Advanced Spectroscopy (B7),

Department of Physics, Princeton University, Princeton, NJ, USA

³Institute of Physics, Academia Sinica, Taipei 11529, China

⁴Lawrence Berkeley National Laboratory, Berkeley, CA, USA

⁵Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

 $^{6}Collaborative$ Innovation Center of Quantum Matter, Beijing 100871, China

⁷CAS Center for Excellence in Topological Quantum Computation,

University of Chinese Academy of Sciences, Beijing 100190, China

⁸Beijing Academy of Quantum Information Sciences, Beijing 100193, China

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Identification of the β -EuP₃ phase by using powder X-ray diffraction

The low-temperature phase of β -EuP₃ was grown by cooling down the melted mixed raw materials. In order to confirm the resultant crystals, Cu- K_{α} powder X-ray diffraction (XRD) measurement was conducted at room temperature in a Rigaku MiniFlex-600 diffractometer. Figure S1 shows the XRD pattern for β -EuP₃ in which the peaks obtained through Rietveld refinement are labeled. The XRD pattern matches well with the one in earlier experiment [1].



Fig. S1. The powder XRD pattern for β -EuP₃ in which the peaks are indexed with (*hkl*).

Isothermal M(H) curves for β -EuP₃

Figure S2 shows the M(H) curves at different temperatures for β -EuP₃. The saturated magnetization is above 7.2 μ_B /formula unit (f.u.) at 2 K, very close to the value for the $4f^7$ spin configuration of the ${}^8S_{7/2}$ Hund's ground state of Eu²⁺ cation.



Fig. S2. M(H) for β -EuP₃ at different temperatures when the field is perpendicular to the plane.

High resistance measurement

The PPMS's capacity for measuring resistance in a four-point fashion is limited to mega ohms. In order to delineate the resistance change in the AFM state at low temperatures, we adapted a DC-IV method by using Keithley 6221 and 6430 for measuring high resistance. The current and voltage chosen in the resistance measurements are listed in Table S1.

Cable S1. The setting details for Keithle	y 6221 and 6430	when recording high	resistance data
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Circumstances in which	Temperature	The Current sourced	The Voltage range
high resistance exists	or field range	by 6221	chosen for 6430
RT in 0 T	2–8 K	10 nA	200 V
RT in 0.5 T	2-4 K	50 nA	20 V
MR at 2 K	0–0.6 T	10 nA	200 V
	0.6–0.8 T	10 nA	20 V
MR at 3 K	0–0.6 T	10 nA	200 V
MR at 4 K	0–0.5 T	10 nA	200 V
MR at 5 K	0–0.4 T	10 nA	200 V
MR at 6 K	0–0.3 T	10 nA	200 V
MR at 7 K	0–0.2 T	10 nA	200 V

Magnetoresistance for α and β -EuP₃

Figure S3 shows the resistivity with respect to the applied field at different temperatures for α and β -EuP₃ in left and right panel, respectively. α -EuP₃ shows a negative MR as long as the spin is polarized below 50 K. This negative MR likely comes from the anisotropic magnetoresistance which exists in ferromagnetic metal in general. As comparison, the resistivity for β -EuP₃ shows a colossal change in magnetic field at base temperature. Obviously the profile and magnitude of the MR are very different between the α and β -phases.



Fig. S3. Resistivity with respect to the applied field for α and β -EuP₃ when the field is perpendicular to the plane.

Hysteresis measurement

As shown is Fig. S4, the MR in β -EuP₃ shows no apparent hysteresis when the magnetic field is parallel along the stacking direction. This feature is essentially different from the large hysteresis in manganites due to the spatial separation in the first order MIT transition [2].



Fig. S4. The resistivity change in β -EuP₃ in a close loop from -9 T to +9 T to -9 T at 2 K.

Fitting of the $\rho_{xx}(T)$ in different magnetic field

In order to address the gap closing for β -EuP₃ in a magnetic field, we fit the $\rho(T)$ curves from 100 K to 300 K with an activated form ($\rho = \rho_0 e^{E_g/2k_BT}$). Figure S5 shows that the $\rho(T)$ curves in different magnetic fields approximately follow the activated form and the yielded band gap E_g is shown in Fig. 3 (d) in the main text.



Fig. S5. The resistivity change in β -EuP₃ in a close loop from -9 T to +9 T to -9 T at 2 K.

Longitudinal MR when the direction of the external field is in the stacking plane

Figure S6 shows the magnetization and electrical properties of β -EuP₃ when the magnetic field is in the stacking plane. Because the moments align along in the stacking plane in the sequence of two-in-two-out, the magnetic field parallel to this direction tends to induce a spin-flop transition. Figure S6 b shows the spin-flop transition occurs at 1 T and then a continuous rotation of the magnetic moment occurs upon increasing H, leading to a saturated magnetization at 2 T at based temperature. Such spin-flop trastion induces a steep change of resistivity in three orders of magnetude (Fig. S6 d). This feature is absent in the transversal MR when the magnetic field is parallel along the hard axis. Figure S6 e and f show that the $\rho(T)$ curves also manifest a hump-like feature which distincts a high-temperature semi-conductive and low-temperature metallic phases in the external fields higher than 2 T. When $\mu_0 H \leq 2.2 T$, the $\rho(T)$ curves show a reentry to the insulator state a low temperature. The magnetization and resistivity measurements reveal that the reentry point overlap with the AFM transition points. There is no phase separation region apparently in the vicinity of the phase boundary (Fig. S7).



Fig. S6. Magnetic and electrical properties of β -EuP₃ when H is in the stacking plane. (a): magnetization (M) under different external magnetic fields ($\mu_0 H$) versus temperature. (b): M versus $\mu_0 H$ at different temperatures. (c): ρ_{xx} under several representative H versus temperature. (d): ρ_{xx} versus H at different temperatures. (e): Zoom-in of the panel (c) demonstrates a broad peak on the $\rho(T)$ curves for $\mu_0 H \ge 2T$. (f): Zoom-in of the panel (c) for 2 K $\le T \le 20$ K demonstrates detailed features on the $\rho(T)$ curves including an MIT and an reentry of the insulating state overlapped with T_N when 2.2 T $\ge \mu_0 H \ge 1.1$ T.



Fig. S7. Contour map in the H-T plane of for resistivity in β -EuP₃ when the field is in the stacking plane. The color bar at the side of the map represent the magnitude of the resistivity. The dots, squares and triangles denote the transition temperatures. PM, PI and AFI stand for the paramagnetic metal, paramagnetic insulator, and AFM insulator phase, respectively.(a): a full diagram for $\mu_0 H < 9 T$ and T < 50 K. The magenta dashed line, determined by magnetization, is the boundary between AFI and PM/PI ; the red dashed line is the boundary between PI and PM. The black dashed line, determined by resistivity, is the boundary between AFI and PM. The black dashed line is overlapped the magenta. (b) Zoom-in of the phase diagram for $1.1 T < \mu_0 H < 2.4 T$ and 2 K < T < 20 K show no phase separation of AFM and insulator states.

Resistance for another piece of β -EuP₃

We have measured several pieces of samples. All the samples in the experiment, which were obtained from the crystal growth described in the draft, behave similar in the R(T) and R(H) dependence. Figure S8 shows another piece of β -EuP₃ which has CMR at 10 K as well. Because its resistance is beyond the measurement upper limit of PPMS, we did not measure the MR below 10 K. Yet the profile of R(T) and R(H) is same as that for the piece of the sample shown in the text.



Fig. S8. Left panel: R(T) curve for another piece of β -EuP₃. Right panel: Resistance versus magnetic field at 10 K.

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^{*} Electronic address: gwljiashuang@pku.edu.cn