

Onset of non-linear IV transport with magnetic order in a Mott insulator

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MATERIALS AND METHODS:

Single Crystal Growth and Diffraction: The single crystals of EuMn_2P_2 were grown via Sn flux method as previously reported.¹ The single crystals had a hexagonal plate like morphology, 0.5–2 mm in width. Powder X-ray diffraction and X-ray energy dispersive spectroscopy (EDS) were used to confirm the phase purity and elemental composition of the single crystals. Single crystal X-ray diffraction data were collected using a SuperNova diffractometer equipped with an Atlas detector and a Mo $K\alpha$ source. The cuboid crystal, cut from a larger crystal piece, was mounted with Paratone-N oil. Data was analyzed and reduced using the CrysAlisPro software suite, version 1.171.36.32 (2013), Agilent Technologies. Initial structural models were developed using SIR92 and refinements of this model were done using SHELXL-97 (WinGX version, release 97-2).^{2,3} Real-time back reflection Laue X-ray diffraction was used to orient and align the crystals for measurement. Single crystals of EuZn_2P_2 for phonon subtractions were also synthesized using a Sn flux, via a similar method that will be reported elsewhere. The single crystals of EuZn_2P_2 and EuMn_2P_2 were found to be stable on the benchtop.

DFT Calculations: The bulk of the first-principle calculations were performed using the Vienna abinitio simulation package (VASP), and the GGA with the Perdew–Burke–Ernzerhof (PBE) type exchange correlation potential was adopted. For the self-consistent calculations a Monkhorst-Pack k-mesh of size $14 \times 14 \times 4$ was used to sample the Brillouin zone (BZ). The energy cutoff is chosen 1.5 times as large as the values recommended in relevant pseudopotentials. Spin-orbit coupling (SOC) was included self-consistently as described in the text. The Eu 4f states were treated by employing the GGA+U approach with the U value set to 5.0eV. A Hubbard $U = 5$ eV for Mn was used where described in the text.

Optical Spectroscopy: Transmissivity of EuMn_2P_2 single crystals was measured using a commercial FTIR spectrometer (Bruker Vertex 80V, Source: Globar, Detector: MCT) in the MIR (400 cm^{-1} to 6000 cm^{-1}) at room temperature. The complex conductivity of the material was determined using Kramers-Kronig (KK) constrained variational dielectric function (VDF) fitting as implemented in the freely available software RefFit.⁴ This fitting method is an alternative to the traditional approach of using a KK transform to determine the complex conductivity.

Bulk Magnetic Properties Measurements: Magnetization measurements were performed in a vibrating sample superconducting quantum interference device magnetometer (SQUID-VSM) from Quantum Design. All measurements were carried out after cooling in zero field. To reduce the remnant field of the superconducting magnet to less than 2 Oe before each measurement, we applied a magnetic field of 5 T at ambient temperature and then removed it in an oscillation mode. The magnetic field was applied parallel and perpendicular to the rod crystal direction of the single crystals that corresponds to the c-axis. Sample shape correction was accounted for in these measurements. In addition to the standard magnetization as a function of field and temperature, the oven option was used to measure the magnetization at high temperatures $T=300\text{-}700\text{K}$. The sample was mounted using the Zircar Cement for the oven option.

The Curie-Weiss law:

$$\chi(T) = \chi_0 + \frac{C}{T - \theta_{CW}} \quad (1)$$

where χ_0 is the temperature-independent susceptibility, C is the Curie constant, and θ_{CW} is the Weiss temperature, was used to analyze the high temperature behavior.

Thermodynamic and Electrical Transport Properties: All of the thermodynamic and electrical transport properties measurements were carried out in the Physical Properties Measurement System (PPMS-9), Quantum Design. The heat capacity measurements on the single crystals of EuMn_2P_2 and EuZn_2P_2 was collected from $T = 2\text{-}300 \text{ K}$ under applied fields of $\mu_0 H = 0.1 \text{ T}$. These measurements utilized

the heat capacity option in the PPMS-9 using the semi-adiabatic pulse technique with a 1% temperature rise and three repetitions at each temperature.

The resistivity option in the PPMS-9 was utilized to carry out the resistivity measurements. The resistivity was measured from $T = 200$ - 400 K using the four-probe technique. The leads were made out of Pt wire and the contacts were made using Dupont 4922N Ag paste. The Pt lead distance was 0.38 mm. The sample length was 1.3 mm longitudinally. The Hall resistivity data were antisymmetrized with respect to the applied magnetic field whereas the longitudinal resistivity data were symmetrized. For the temperature-dependent where $T=300$ K, two temperature sweeps were carried out, one with a positive and one with a negative field of the same magnitude. The data were subsequently antisymmetrized to subtract the contribution of the longitudinal resistivity.

$$\rho_{xx}(\mu_o H) = \frac{\rho(+\mu_o H) + \rho(-\mu_o H)}{2} \quad (2)$$

$$\rho_{xy}(\mu_o H) = \frac{\rho(+\mu_o H) - \rho(-\mu_o H)}{2} \left(\frac{L}{W} \right) \quad (3)$$

Where $\rho(+\mu_o H)$, $\rho(-\mu_o H)$ indicate the measured resistivity at positive and negative values of the magnetic field, respectively, while L and W designates the length and the width of the sample.

The phonons for EuZn_2P_2 were subtracted by first constructing an equivalent Debye model and then scaling for the small molecular mass difference between Zn and Mn. The two Debye model used is:

$$\frac{C_p}{T} = \frac{C_D(\theta_{D1}, s_1, T)}{T} + \frac{C_D(\theta_{D2}, s_2, T)}{T} \quad (4)$$

$$C_D(\theta_D, T) = 9sR \left(\frac{T}{\theta_D} \right)^3 \int_0^{\theta_D/T} \frac{(\theta/T)^4 e^{\theta/T}}{[e^{\theta/T} - 1]^2} d\frac{\theta}{T} \quad (5)$$

Where θ_{D1} and θ_{D2} are the Debye temperatures, s_1 and s_2 are the oscillator strengths, and R is the molar Boltzmann constant.⁵ The model parameters from the least-squares refinement to the data for $T > 25$ K are given in Table-SVII. The total oscillator strength $s_1 + s_2 = 5.18(6)$. This is in good agreement with the expected value of $1+2+2 = 5$, the total number of atoms per formula unit in EuZn_2P_2 .

For EuMn_2P_2 , once the phonons of EuZn_2P_2 were calculated, the phonons were normalized to EuMn_2P_2 using equation:

$$\frac{\theta_{LmYsZp}^3}{\theta_{XmYnZq}^3} = \frac{mM_X^{3/2} + nM_Y^{3/2} + qM_Z^{3/2}}{mM_L^{3/2} + sM_Y^{3/2} + pM_Z^{3/2}} \quad (6)$$

where θ_{LmYsZp}^3 and θ_{XmYnZq}^3 are the normalization factor of EuMn_2P_2 and EuZn_2P_2 , $mM_X^{3/2} + nM_Y^{3/2} + qM_Z^{3/2}$ and $mM_L^{3/2} + sM_Y^{3/2} + pM_Z^{3/2}$ are the molar masses of EuMn_2P_2 and EuZn_2P_2 (as $1\text{Eu}+2\text{Mn}+2\text{P}$ and $1\text{Eu}+2\text{Zn}+2\text{P}$). The normalization factor outputted is 1.026 respectively.^{5,6}

The electrical transport operation (ETO) option was utilized to carry out the electrical transport at low temperatures, $T=2$ - 100 K. All measurements were done using the inbuilt ETO option in the PPMS. This measurement was done with angle, magnetic field, and temperature dependence on voltage and current behavior. The input current was manually changed at every temperature in consideration of the insulating nature of EuMn_2P_2 . The applied magnetic field with 1 T intervals between $\mu_o H=0$ - 9 T and a temperature interval of $T=2$ - 20 K with 1 K intervals. These electronic resistivity measurements are performed using a two-probe configuration with the longitudinal and transverse resistivity probes connected to independent measurements channels. As the contacts are manually fabricated with silver epoxy, the measured data may exhibit effects of asymmetry with magnetic field due to slight misalignments of the silver contacts.

Data was analyzed with a two-channel model as described in the text. A real Schottky diode with a series resistance has a characteristic response function of:

$$I = I_s \left(e^{\left(\frac{V_{meas} - IR_s}{nV_t} \right)} - 1 \right) \quad (7)$$

Where I_s is the saturation current, nV_t is the characteristic voltage, and R_s is the series resistance. This has to then be put in parallel with a normal resistor to form the two-channel model. For fitting purposes, rewriting and symmetrizing is convenient:

$$V_{meas.} = \left(\frac{I_0}{|I_1|} \right) (nV_t \ln \left(\frac{|I_1|}{I_{s+1}} \right) + |I_1| R_s) \quad (8)$$

$$I_2 = \frac{V_{meas.}}{R_p} \quad (9)$$

$$I_{total} = I_1 + I_2 \quad (10)$$

Where I and $V_{meas.}$ are the raw data for at a given temperature, applied field, and sample angle.

Thermal Analysis Measurements: were performed utilizing a Netzsch DSC 404 C instrument. Approximately 20 mg of the sample in a glassy carbon crucible (L2.5 mm, l=5 mm) was sealed in a Nb ampule (L=5 mm, l=15 mm). The samples were heated under an Ar atmosphere with a heating rate of 10Kmin⁻¹ to T=450-580K and then cooled to 100 °C with a cooling rate of 10 Kmin⁻¹.

³¹P NMR: ³¹P MAS NMR spectrum was recorded on Bruker AVANCE-II spectrometer at 4.7 T magnetic field (³¹P NMR frequency of H₃PO₄ reference 80.987 MHz) using home built MAS probe for 1.8 mm od Si₃N₄ rotors. The spectrum was obtained with spin echo pulse sequence p/2 – t – p – t – rec, where p/2 = 1.7 ms, the echo delay was one sample rotation period t = t_r = 26 ms, and a 25 ms relaxation delay between the accumulations. 32k averages have been summed for a spectrum. Since the spectrum width is a bit larger than the excitation window, we used frequency sweep with 1a 50 kHz step of excitation and summed together for the total spectrum given in Fig. 1. The sample spinning rate was 42 kHz. The chemical shift is given respective to the resonance frequency of H₃PO₄. Spin-lattice relaxation of ³¹P was found to be exponential T₁ = 56 ms, as measured by inversion recovery pulse sequence. The ³¹P Knight shift temperature dependence was acquired at four temperatures from 295K down to 80K. Both the susceptibility and broadening of the spectra followed the Curie-Weiss behavior. The ³¹P Knight shift $K(T)$ follows perfectly the susceptibility curve $\chi(T)$. The χ vs K relation can be written as

$$K(T) = K_0 + \frac{H_{hf}}{N_A \mu_B} \chi \quad (11)$$

where K_0 is the temperature-independent shift and H_{hf} is the hyperfine field. From K vs χ fit we found a hyperfine field value of $H_{hf} = 4.0$ kOe/ μ_B .

¹⁵¹Eu ZF-NMR: ¹⁵¹Eu powder sample ZFNMR measurements of EuMn₂P₂ at T = 4.2 K were collected on a AVANCE II spectrometer with a home-built low temperature probe immersed in the liquid helium in dewar. The spin-5/2 nucleus shows 5 possible transitions in the spectra. When the magnetic moment of the nuclei is aligned with the quadrupolar field the gap between the peaks is uniform. Here, the middle transition has a different location, which refers to an angle between the two fields. In EuMn₂P₂ the electric field gradient (EFG) tensor is symmetric and perpendicular to the c -axis. Exact resonance frequencies can be calculated by the nuclear spin Hamiltonian

$$\mathcal{H} = -\gamma_n \hbar \mathbf{I} \cdot \mathbf{H}_{int} + \frac{h\nu_Q}{6} \left[3I_z^2 - I(I+1) + \frac{1}{2}\eta(I_+^2 + I_-^2) \right] \quad (12)$$

where the first term represents the Zeeman interaction between the nuclear magnetic moment $\mu_n = \gamma_n \hbar \mathbf{I}$ and the internal magnetic field \mathbf{H}_{int} . The second term represents the nuclear quadrupolar interaction between the EFG and the nuclear quadrupolar moment, where $\nu_Q = \frac{3e^2 Qq}{2I(2I-1)\hbar}$ is the nuclear quadrupolar frequency and η is a symmetry parameter of the EFG tensor. Here eQ is the electric quadrupole moment

of the nucleus and $eq = V_{zz}$ is the main principal value of the EFG tensor, $\eta = \frac{V_{xx}-V_{yy}}{V_{zz}}$. The operators are transformed from the crystal frame to the Laboratory frame using Euler angles in the ZYZ convention, after which the eigenvalues of eq. (12) are found. The differences of the eigenstates represent the transition energies, five of which have a probability to occur. The calculated results offer a good match with the experiment and we acquired values for the quadrupolar frequency, strength of the internal field and the angle of the quadrupolar field. The parameter errors were estimated with standard deviation in the error regions of the experimental data fittings.

¹⁵¹Eu Mössbauer spectroscopy: The ¹⁵¹Eu ME measurements at ambient pressure in the temperature range 300–1.75 K were carried out on these samples using a 100 mCi ¹⁵¹SmF₃ source. Both source and absorber were kept at the same temperature in a top-loading cryostat. Mössbauer spectra were analyzed with the NORMOS software package,⁷ to derive pertinent hyperfine interaction parameters, isomer shift S, magnetic hyperfine field B_{eff}, and absorption areas (abundances) of spectral components. All isomer shift values are quoted relative to the SmF₃ source from here onwards.

Table SI. Quantification of the non-linear behavior in a two-channel model with i⊥c at $\mu_o H=0$ T as described by the combination of equation (8), (9), and (10) in the SI.

T (K)	nV_i (V)	R_s (Ω)	I_s (A)	R_p (Ω)
2	1.46263	13453.01	1.10×10^{-5}	4.93×10^{20}
3	0.98622	11132.51	1.78×10^{-5}	4.79×10^{19}
4	0.90182	8309.404	3.92×10^{-5}	4.84×10^{10}
5	1.50788	5090.016	1.38×10^{-4}	2.70×10^{10}
6	1.52124	4050.199	2.29×10^{-4}	2.70×10^9
7	1.24701	3899.045	2.22×10^{-4}	41228.02
8	0.99289	3352.601	2.99×10^{-4}	54326.12
9	1.68091	2459.097	6.80×10^{-4}	63239.63
10	1.28865	2261.202	7.66×10^{-4}	118603.8
11	0.92313	2544.819	4.48×10^{-4}	11069.33
12	1.28358	2597.081	4.79×10^{-4}	5849.598
13	0.61154	3434.207	1.47×10^{-4}	3637.912
14	0.69954	4146.982	1.03×10^{-4}	2742.348
15	0.65008	3543.032	1.14×10^{-4}	2536.328
16	0.50423	6332.828	1.79×10^{-5}	1876.715
17	0.47924	6386.381	1.88×10^{-5}	1713.698
18	0.36378	7733.925	7.27×10^{-6}	1534.904
19	0.36783	7573.468	7.24×10^{-6}	1416.138
20	0.36179	7234.724	8.36×10^{-6}	1320.561

Fig. S1 ^{151}Eu Mössbauer spectra of EuMn_2P_2 single crystals showing the evolution of the spectra with temperature. The solid lines are fits derived from either a full Hamiltonian solution (red line: $T = 13.75\text{K}$) or from the dynamic model (magenta lines).

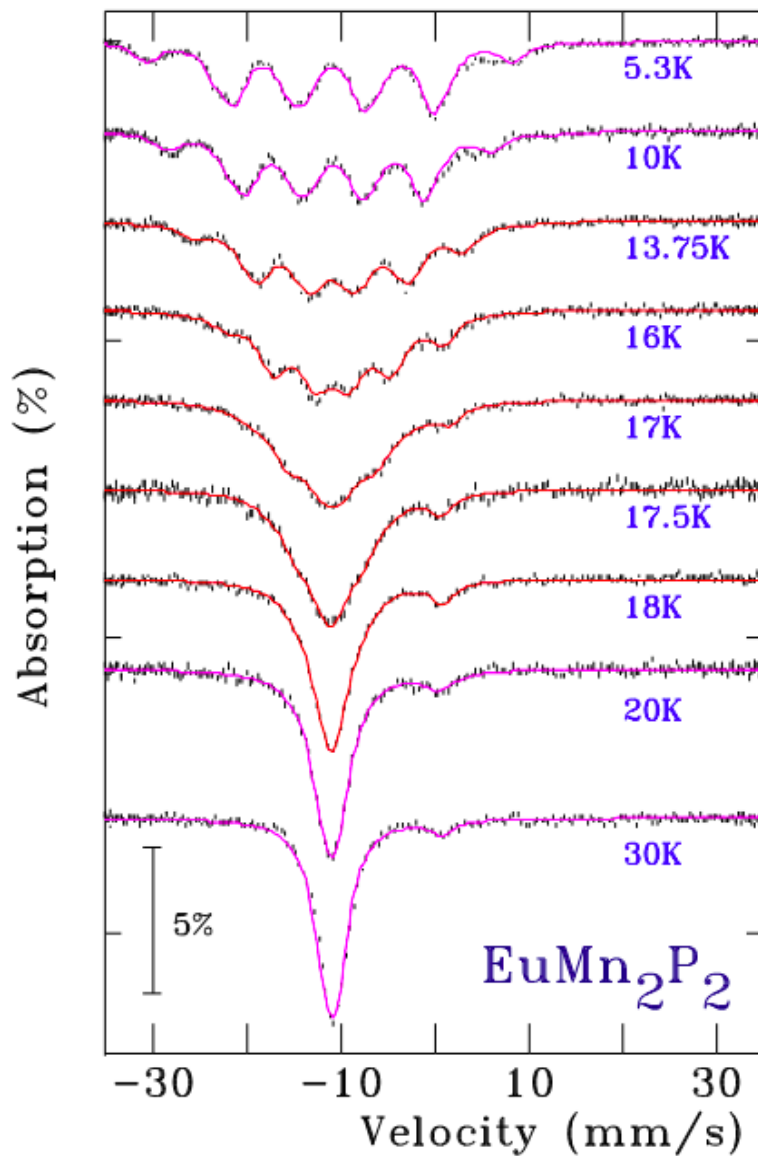


Fig. S2 Temperature dependence of the fitted hyperfine field contributions in EuMn_2P_2 derived from the modulated model. **(a)** The average hyperfine field (B_{avg}) is plotted as solid round symbols with a dotted line showing a fit yielding a transition temperature of 18 K. **(b)** fluctuation rate derived from dynamic fits derived from the dynamic distribution fits.

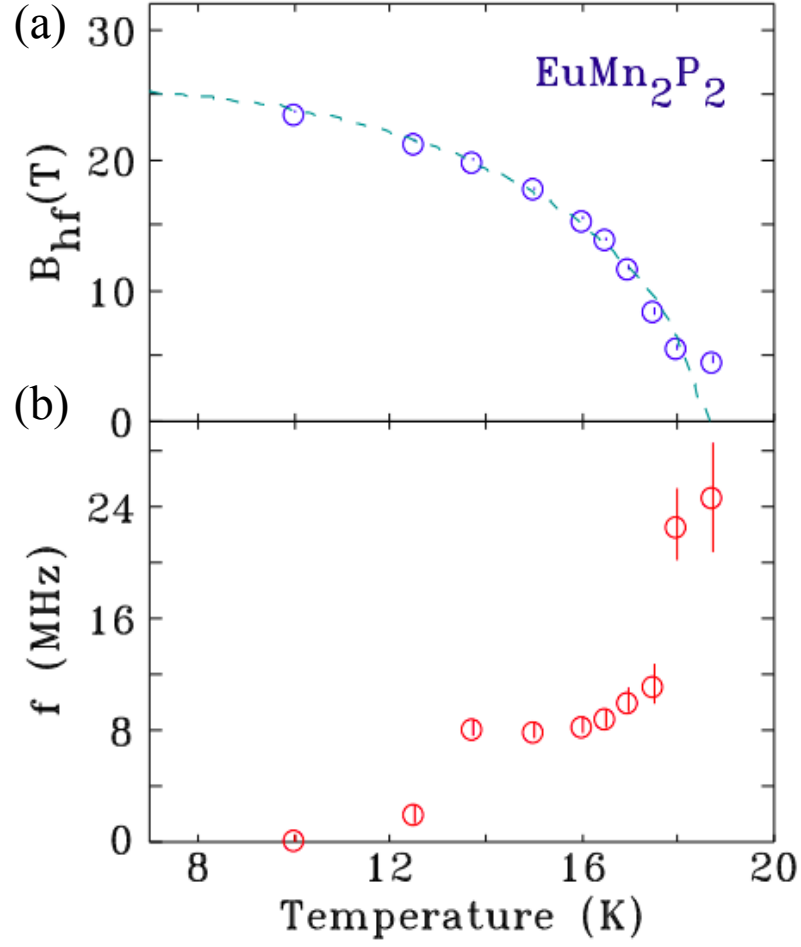


Fig. S3 Curie-Weiss Analysis of EuMn_2P_2 for $\mu_0 H \parallel c$ and $\mu_0 H \perp c$ shows a cusp indicative of magnetic order at $T_N = 17.5(1)$ K. The effective magnetic moment is higher than the theoretical value for Eu^{2+} ($S=7/2$), and consistent with a small contribution from Mn ions. (a) $\mu_0 H \perp c$ for $\mu_0 H = 0.1$ T in the range $T = 2$ -300 K, (b) Curie-Weiss analysis $\mu_0 H \parallel c$ for $\mu_0 H = 0.1$ and $T = 2$ -300 K. The measurements of the crystal sample show an excellent agreement with the previous study by Payne *et. al.*¹

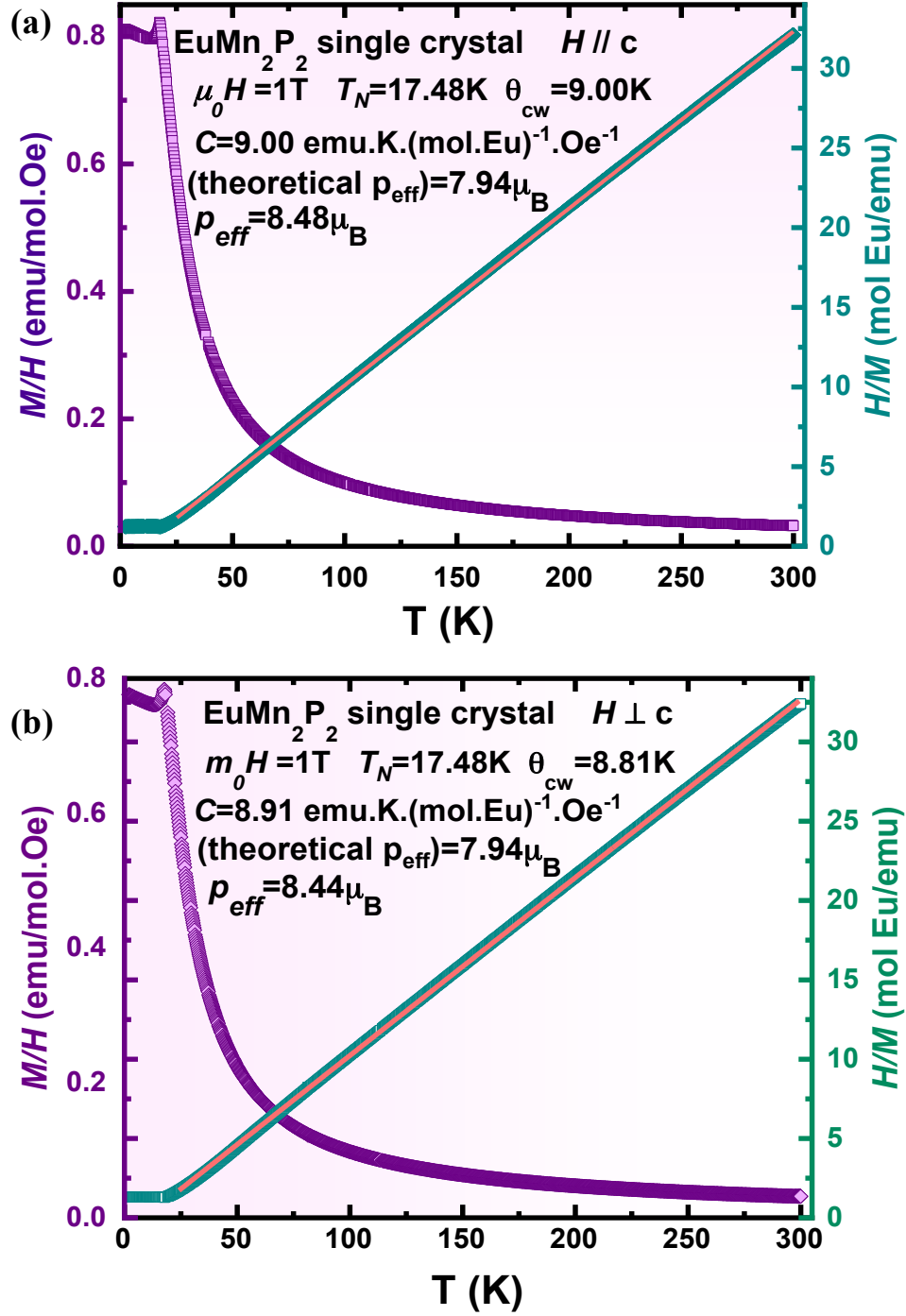


Fig. S4 The magnetic behavior is isotropic above T_N , with the development of a weak anisotropy ($\sim 5\%$) below T_N . **(a)** Comparison in the magnetization as a function of temperature for $\mu_o H \parallel c$ and $\mu_o H \perp c$, $T = 2\text{--}300\text{K}$, **(b)** anisotropy in the $\mu_o H \parallel c$ and $\mu_o H \perp c$ below T_N , and **(c)** ratio of magnetization $\mu_o H \parallel c$ and $\mu_o H \perp c$ at $\mu_o H = 0.1\text{ T}$ and $T = 2\text{--}300\text{ K}$ displaying the anisotropy below T_N . (Ref 1: $\chi(T_N) = 0.63\text{ emu/mole}$, any thoughts?)

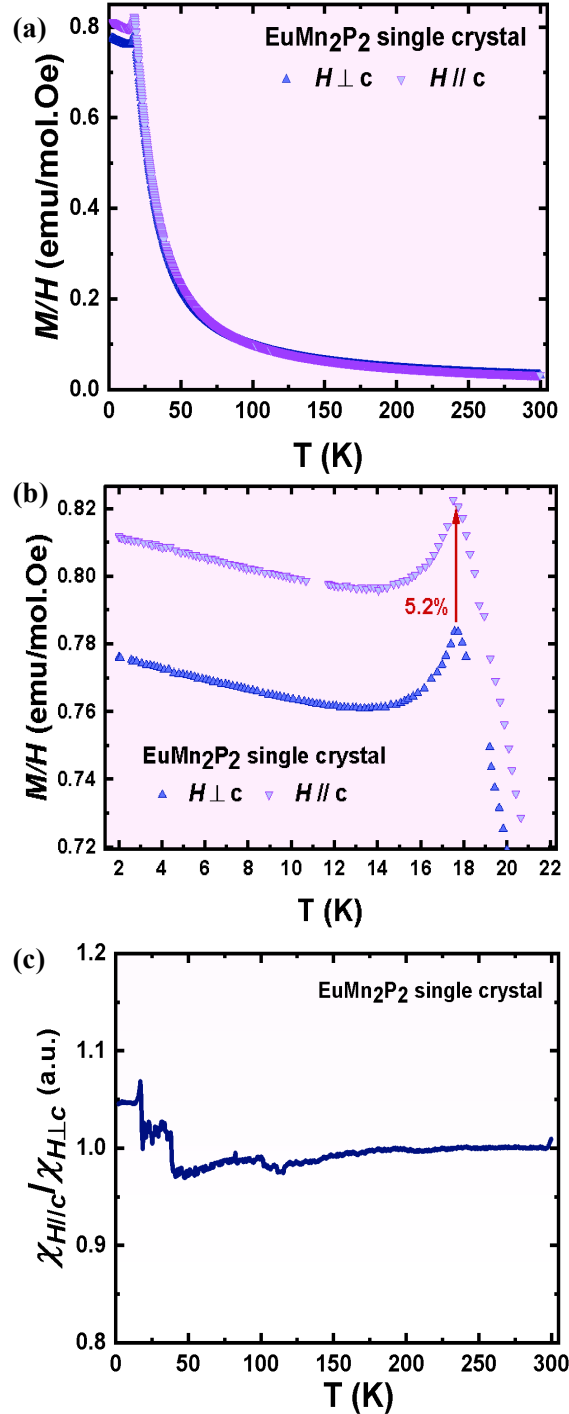


Fig. S5 (a) Magnetization as a function of magnetic field with $\mu_0 H \parallel c$ from $\mu_0 H = -7$ to 7 T and $T = 2$ -25 K, showing a linear response below 3 T, as seen in most Eu compounds. Full saturation at $7 \mu_B/\text{Eu}^{2+}$ is observed around $\mu_0 H = 4$ T. **(b)** There are small, offset, hysteresis loops observed in the magnetization as a function of magnetic field with $\mu_0 H \parallel c$ at intermediate temperatures below T_N , e.g. $T=8$ K. **(c)** Magnetic phase diagram of EuMn_2P_2 derived from Magnetization as a function of magnetic field at with $\mu_0 H \parallel c$, constructed using the data from (a) and (b).

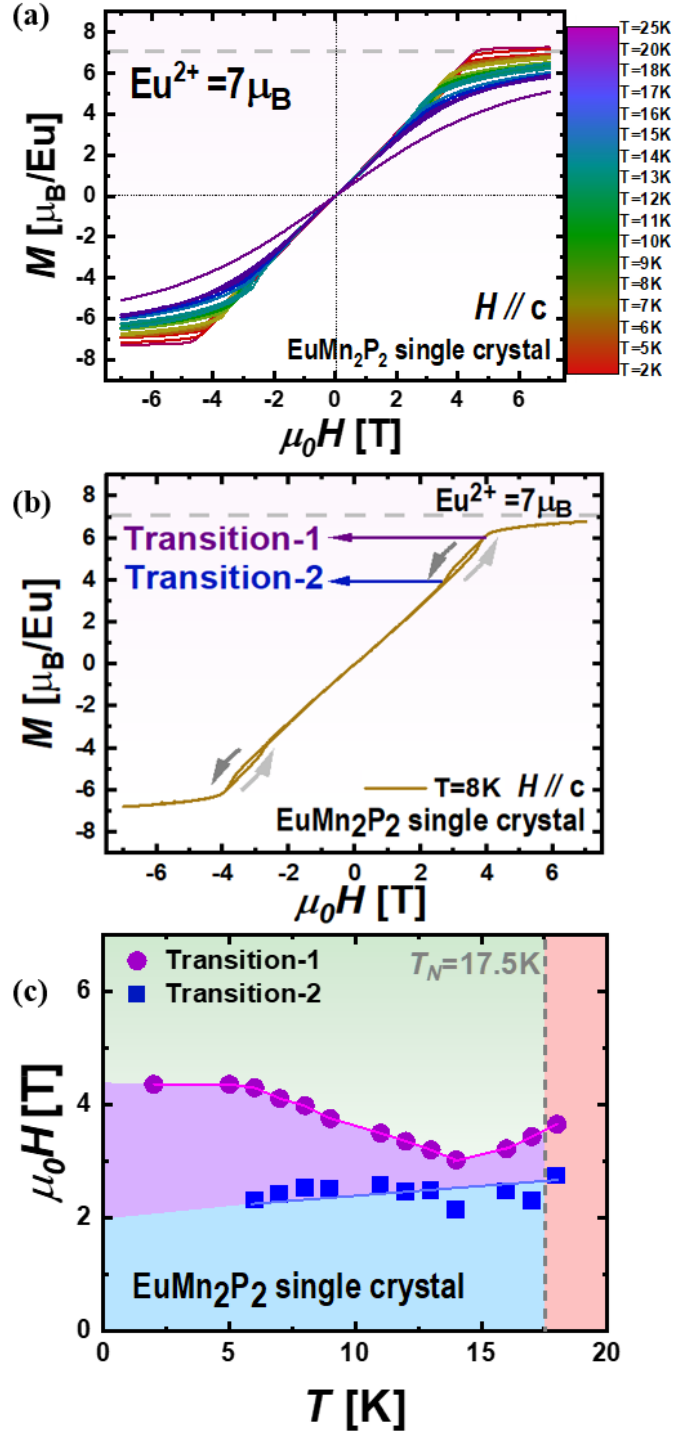


Fig. S6 ^{151}Eu NMR measurement in zero applied field represented with black squares. The blue line represents a fitting done with the five transition frequencies acquired from the Hamiltonian with the provided parameters.

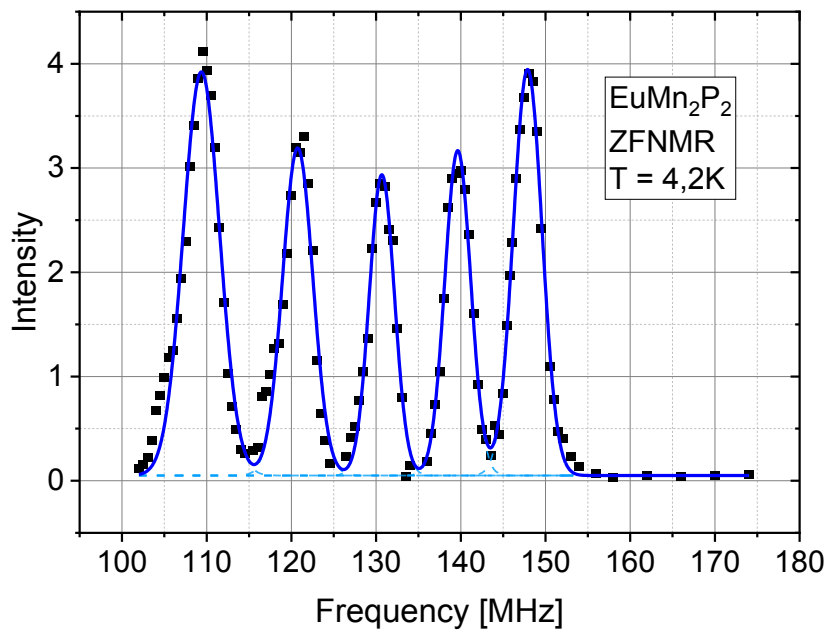


Table SII. Experimental and calculated transition frequencies in the spectrum of ^{151}Eu ZFNMR of EuMn_2P_2 , at $T = 4.2\text{K}$. The calculation parameters: $\nu_Q = 19.47 \pm 0.03\text{ MHz}$, asymmetry of the EFG tensor: $\eta = 0$, Larmor frequency $\nu_L = 129.28 \pm 0.13\text{ MHz}$, corresponds to the internal field at Eu nucleus $H_{\text{int}} = 12.32\text{ T}$, and angle between the magnetic moment and quadrupolar tensor $\theta = 87.12 \pm 1.07$ degrees.

Transition $m =$	Exp (MHz)	Standard dev (MHz)	Calc (MHz)
$-5/2 \leftrightarrow -3/2$	147.884	0.0576	147.890
$-3/2 \leftrightarrow -1/2$	139.663	0.0664	139.642
$-1/2 \leftrightarrow +1/2$	130.656	0.0707	130.681
$+1/2 \leftrightarrow +3/2$	120.758	0.0714	120.746
$+3/2 \leftrightarrow +5/2$	109.374	0.0634	109.376

Fig. S7 Evidence for a magnetic field independent non-linear V-I characteristic at $T=2\text{K}$ at (a) $\mu_0 H \parallel c$ and (b) $\mu_0 H \perp c$ for various magnetic fields. (c) The $\mu_0 H \parallel c$ and $\mu_0 H \perp c$ display identical behavior at $T=2\text{K}$ which is below T_N at $i \perp c$. (d)-(f) are magnified versions of (a)-(c).

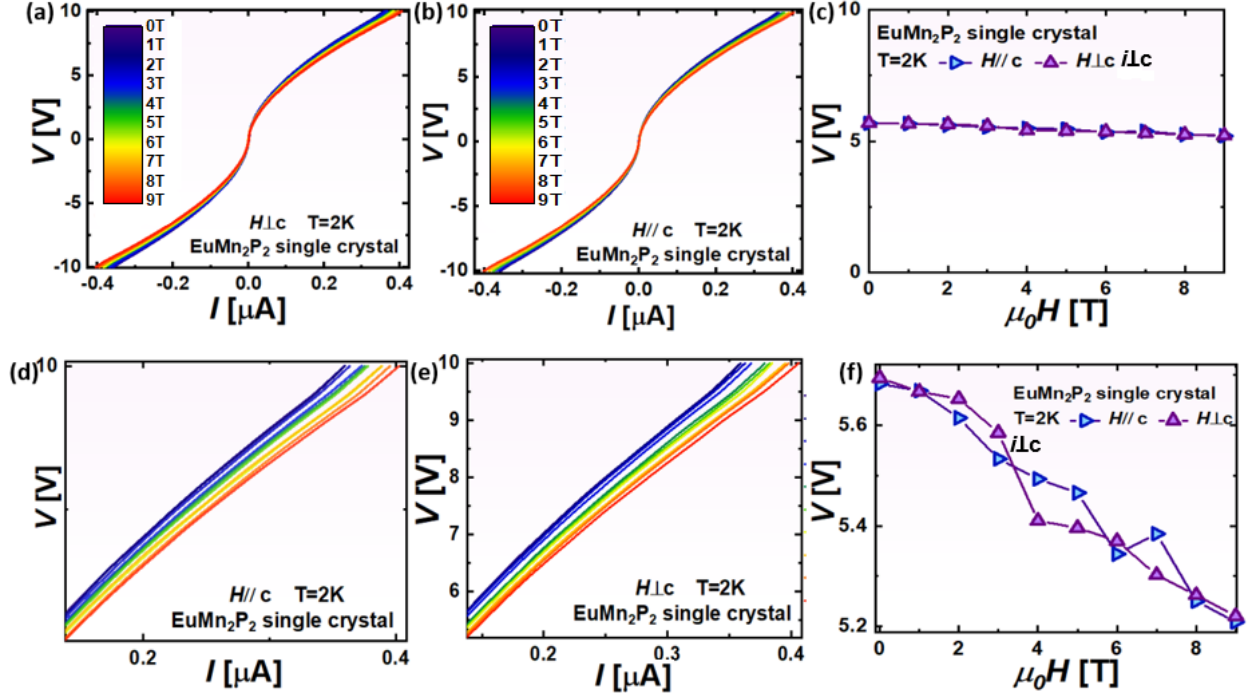


Fig. S8 Alternative view of the I-V behavior: differential resistance, dV/dI , for $\mu_0 H \parallel c$ as a function of voltage at (a) T=2K, (b) T=5K, and (c) T=10K.

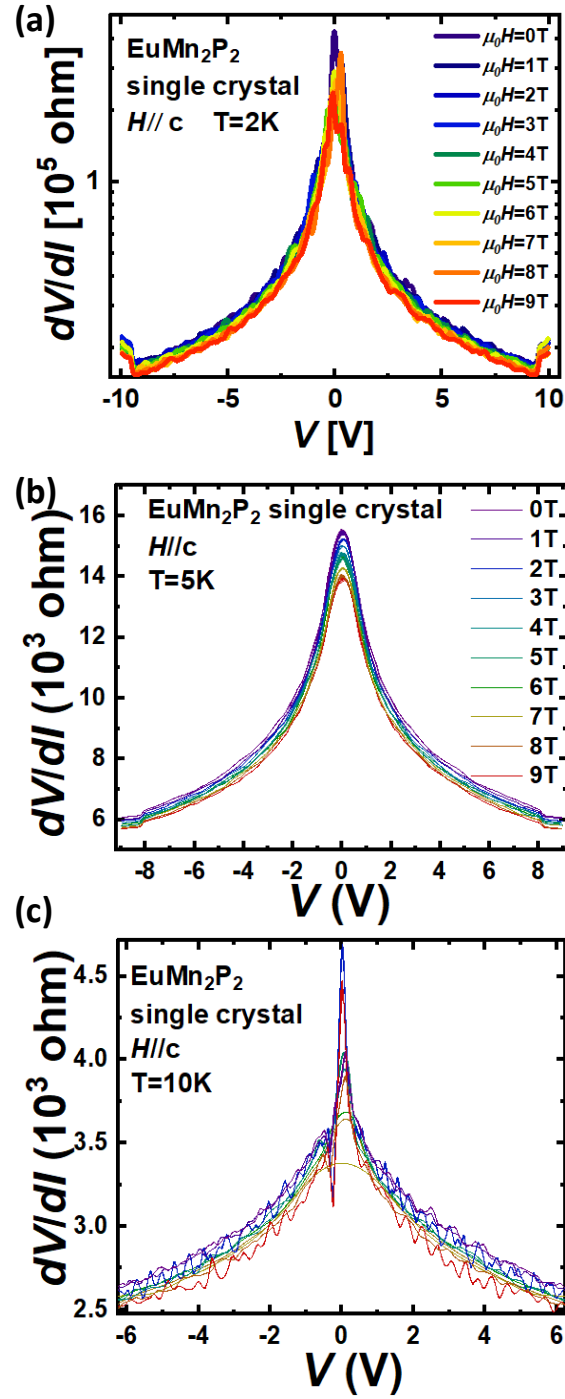


Table SIII The table below is a derived from the combination of equation (7), (8), and (9) in the supplementary text at $\mu_o H \perp c$ and $\mu_o H = 9T$.

T (K)	nV_i (V)	R_s (Ω)	I_s (A)	R_p (Ω)
2	1.38938	12481.17	1.21×10^{-5}	8.85×10^{17}
3	0.99292	10143.23	2.10×10^{-5}	1.22×10^{14}
4	0.98668	7782.008	3.63×10^{-5}	105087.0
5	0.99215	8418.093	9.84×10^{-6}	14616.88
6	0.97453	7089.717	2.34×10^{-5}	11659.76
7	1.29734	4621.303	1.28×10^{-4}	13314.81
8	0.79225	6511.687	3.10×10^{-5}	6492.985
9	1.18612	3633.853	1.81×10^{-4}	7871.676
10	1.02598	3327.602	2.43×10^{-4}	6970.228
11	1.17927	2943.889	3.48×10^{-4}	6000.555
12	1.01141	2664.900	3.96×10^{-4}	5491.424
13	0.97802	2442.924	4.88×10^{-4}	5016.348
14	0.95192	2222.205	5.79×10^{-4}	4530.668
15	0.97223	2244.757	5.46×10^{-4}	3511.671
16	0.96730	2408.785	4.56×10^{-4}	2734.356
17	0.89172	3007.707	2.23×10^{-4}	2019.601
18	0.95455	1695.951	8.97×10^{-4}	3016.451
19	0.90409	2363.399	4.30×10^{-4}	1914.135
20	0.84149	2516.778	1.11×10^{-5}	1240.173

Table SIV Single crystal x-ray diffraction (SXRD) parameters and refinement statistics.

Formula	EuMn ₂ P ₂
Crystal system	Trigonal
Space Group	<i>P</i> -3 <i>m</i> 1 (No. 164)
a (Å)	4.136
b (Å)	4.136
c (Å)	7.005
V (Å ³)	103.776704
Z	1
M/gmol ⁻¹	323.78
ρ ₀ /gcm ⁻³	5.181
μ/mm ⁻¹	21.452
Radiation	Mo Kα, λ = 0.71073 Å
Temperature (K)	293 K
Reflections collected	2632
Unique Reflections	186
Refined Parameters	9
Goodness-of-fit	1.323
R[F] ^a	0.0076
R _w (F _o ²) ^b	0.021

$$^a R(F) = \frac{\sum ||F_o| - |F_c||}{\sum |F_o|}$$

$$^b R_w(F_o^2) = [\sum w(F_o^2 - F_c^2)^2 / \sum w(F_o^2)^2]^{1/2}$$

Table SV Atomic coordinates for EuMn₂P₂ determined by SXRD

	Occ.	Wyckoff Positions	x (Å)	y (Å)	z (Å)	U _{eq} (Å ²)
Eu	1	1a	0	0	0	0.00329(6)
Mn	1	2d	2/3	1/3	0.62080(6)	0.00401(9)
P	1	2d	2/3	1/3	0.26921(10)	0.00412(13)

Table SVI Anisotropic displacement parameters for EuMn₂P₂ determined by SXRD

	U(1,1)	U(2,2)	U(3,3)	U(1,2)	U(1,3)	U(2,3)
Eu	0.00323(7)	0.00323(7)	0.00340(9)	0.00161(4)	0	0
Mn	0.00412(12)	0.00412(12)	0.0380(17)	0.00206(6)	0	0
P	0.0041(2)	0.0041(2)	0.0042(3)	0.00203(10)	0	0

Fig. S9 Band structure and density of states of EuMn_2P_2 within GGA+U. For the Eu atoms we consider an A-type antiferromagnetic order, with $U=5\text{eV}$. For Mn we consider **(a)** a C-type antiferromagnetic order with $U=0\text{eV}$, or **(b)** ferromagnetic order with $U=5\text{eV}$. In both cases DFT predicts metallicity for EuMn_2P_2 and illustrates that antiferromagnetic order or Hubbard U alone are not sufficient to explain the observed semiconducting behavior.

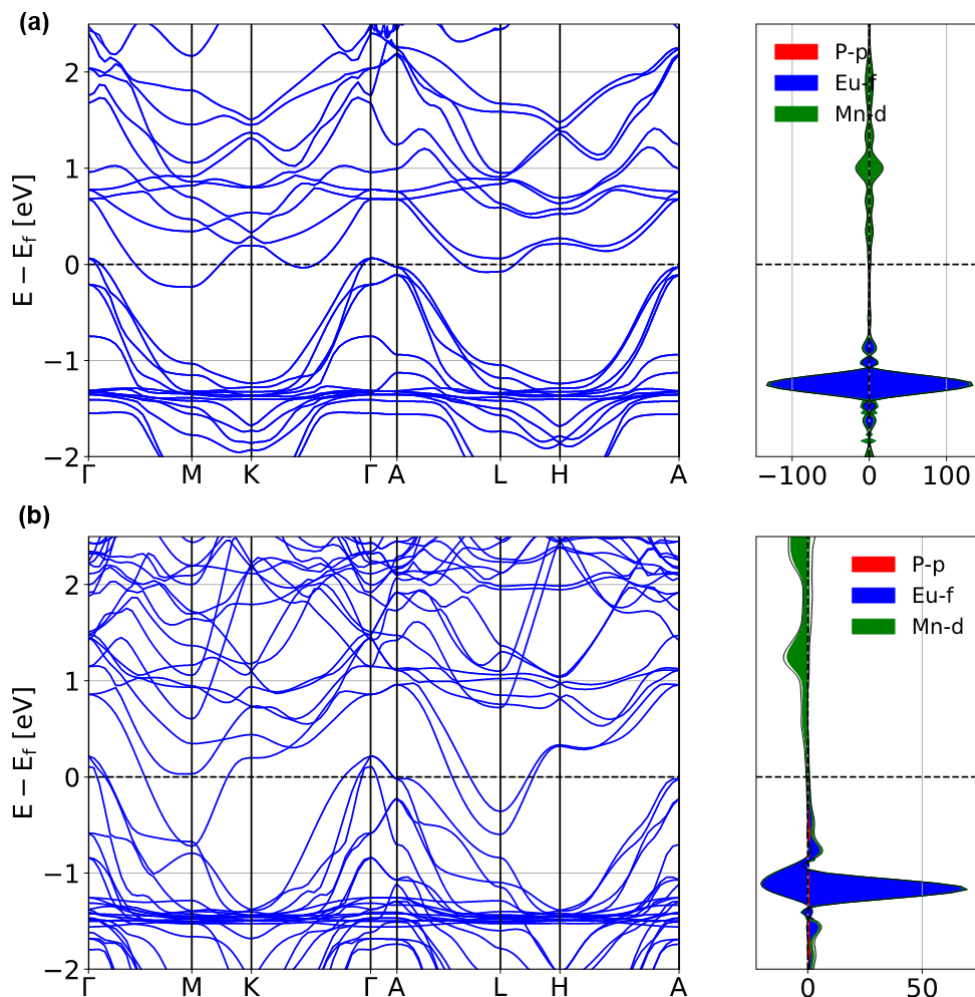


Fig. S10 (a) Band structure and density of states of EuMn_2P_2 within GGA+U considering an A-type antiferromagnetic order with $U = 5\text{eV}$ on the Eu atoms and non-magnetic Mn atoms. A metallic behavior is predicted in agreement with the expectation that a non-spin polarized d-shell should not have an energy gap. (b) Band structure and density of states for EuMn_2P_2 within GGA+U with A-type antiferromagnetic order and $U = 5\text{eV}$ for Eu and C-type antiferromagnetic and $U = 5\text{eV}$ for Mn. These contributions result in a prediction of semiconducting behavior for EuMn_2P_2 . The physical mechanism is the antiferromagnetic order in Eu suppresses spin up to spin down hoppings t , where $t < U$ and thus resulting in a Mott insulator. Further, within the crystal structure of EuMn_2P_2 we notice that the Mn and P units are bonded covalently in tetrahedra, whereas Eu layers behave as Van der Waals layers with weak J coupling with one another. These bonding interactions signify that the close bonding in the Mn-P units corresponds to the density of states close to the Fermi level and the weak Eu bonding is further away from the Fermi level.

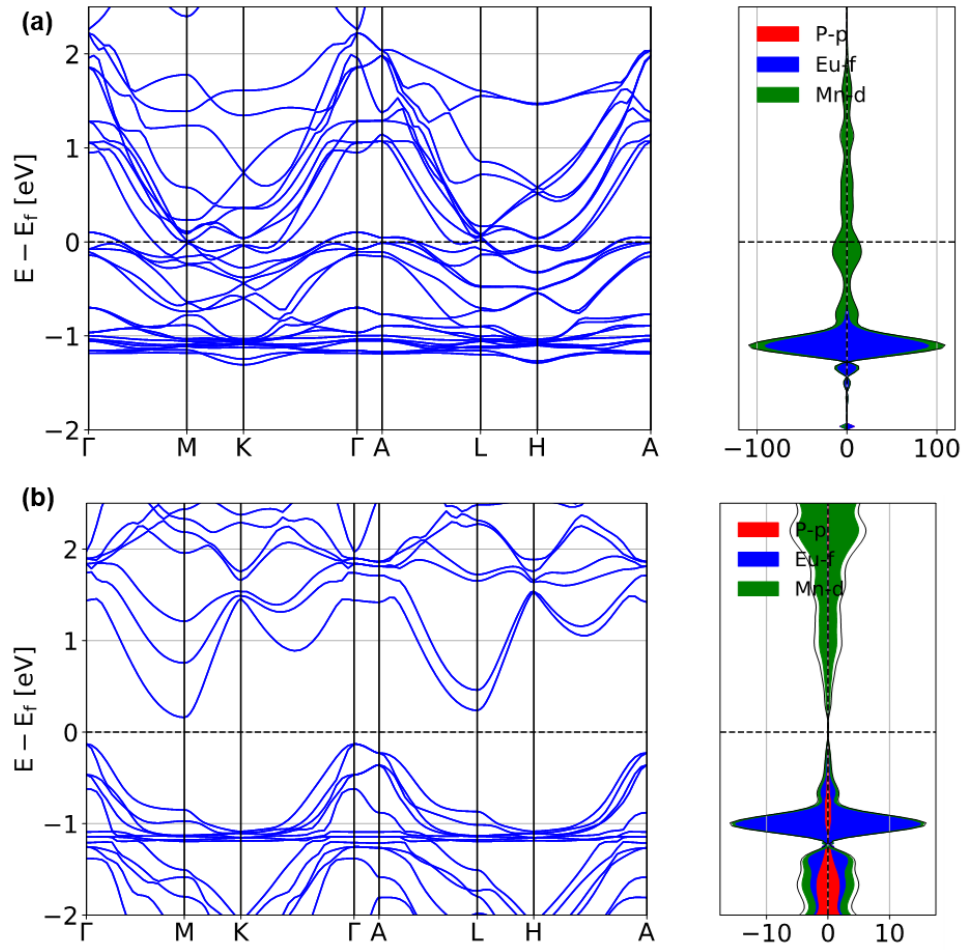


Fig. S11 Band structure for the predicted magnetic configuration (a) without (b) with spin orbit coupling (SOC) shows Evidence that the effect of SOC is negligible. Also, the parity criterion confirms that it is topologically trivial. In addition, we used the Fu-Kane parity criterion to confirm that the EuMn_2P_2 is a topologically trivial insulator.

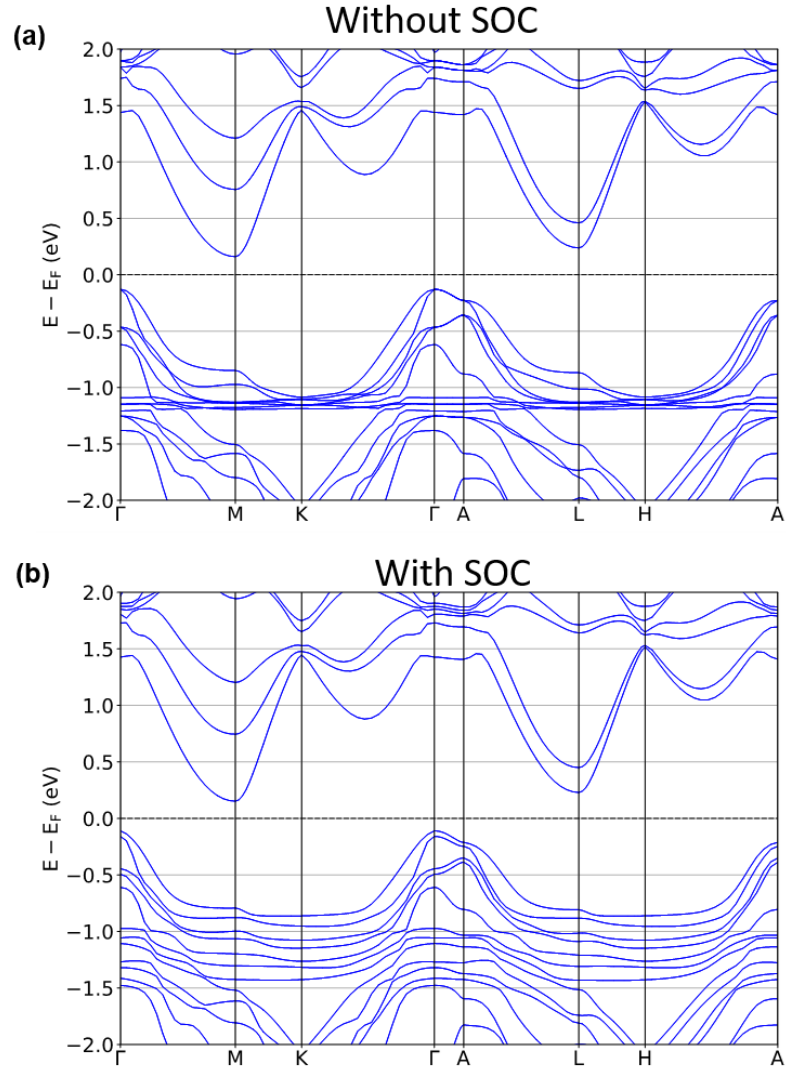


Fig. S12 Comparison between different putative antiferromagnetic orders for the Mn atoms. **(a)** Within GGA+U, the C-type antiferromagnetic order is predicted to be the lowest in energy compared to other putative antiferromagnetic orders. **(b)** This agrees with the observation of a sharp phase transition when Eu atoms magnetically order at 17K due to lowering of the symmetry. **(b)** For a G-type antiferromagnetic order, a sharp phase transition is not expected when Eu atoms magnetically order.

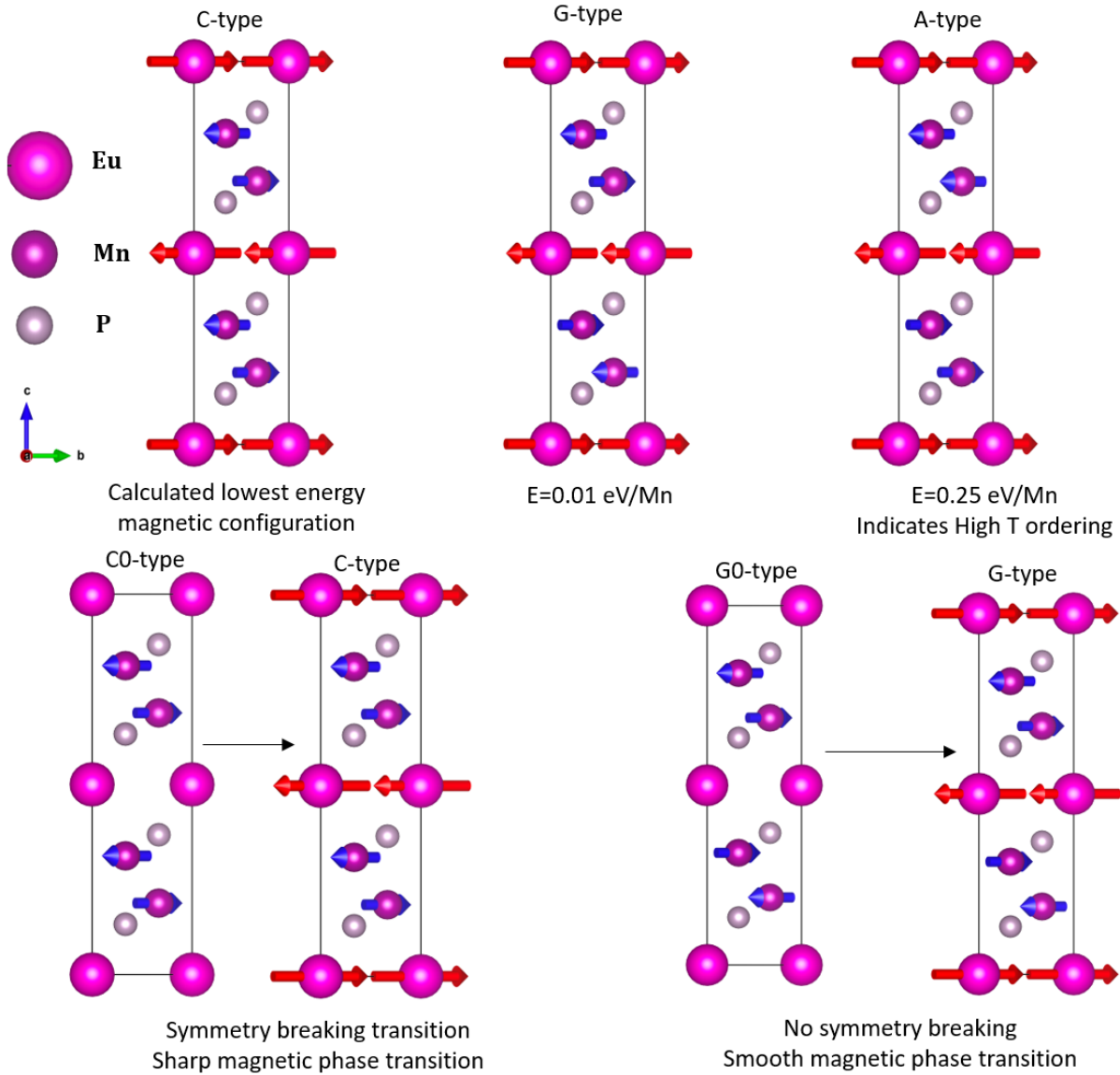


Fig. S13 (a) Four probe resistivity of EuMn_2P_2 single crystals at $T=200\text{-}400\text{K}$. **(b)** Natural logarithm of normalized resistivity as a function of $\frac{1}{T}$ measured on a single crystal of EuMn_2P_2 along the c-axis. The bandgap extrapolated is $E_g=0.2\text{eV}$ and is fit to $\ln\left(\frac{\rho}{\rho_{T=400\text{K}}}\right) = \frac{E_g}{k_B} \frac{1}{T}$.

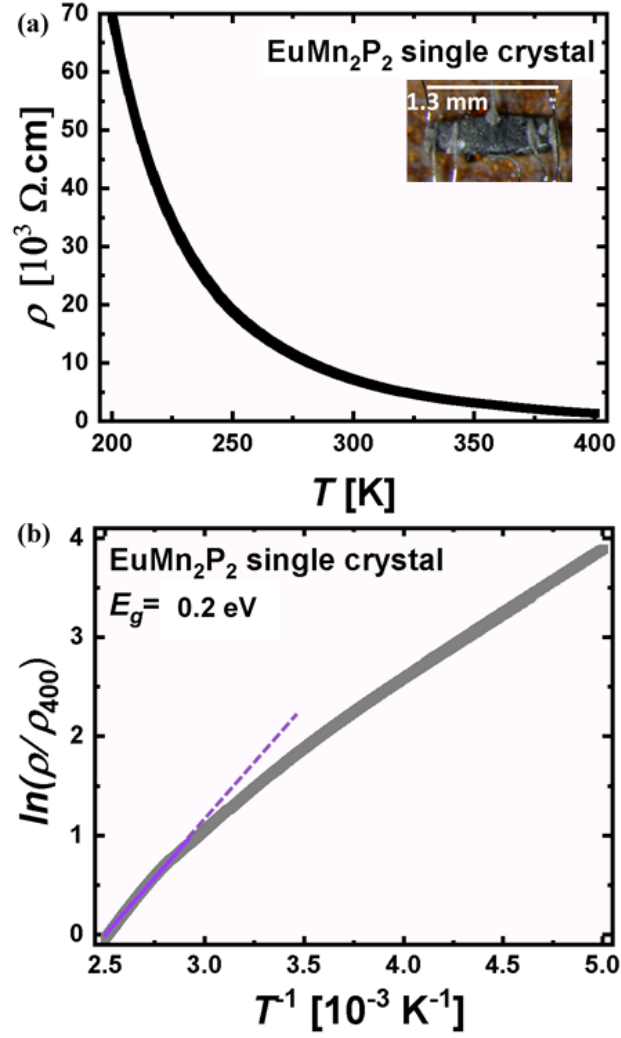


Fig. S14 Optical spectroscopy to determine the (a) transmission and (b) conductivity of EuMn_2P_2 single crystals. These data are consistent with a sharp band edge at 0.68 eV, with a tail of states extending down from the band edge to $\sim 2000 \text{ cm}^{-1}$ ($\sim 0.2 \text{ eV}$), in agreement with transport measurements. The sharp absorption features in the 500-1800 cm^{-1} range are IR active phonon modes.

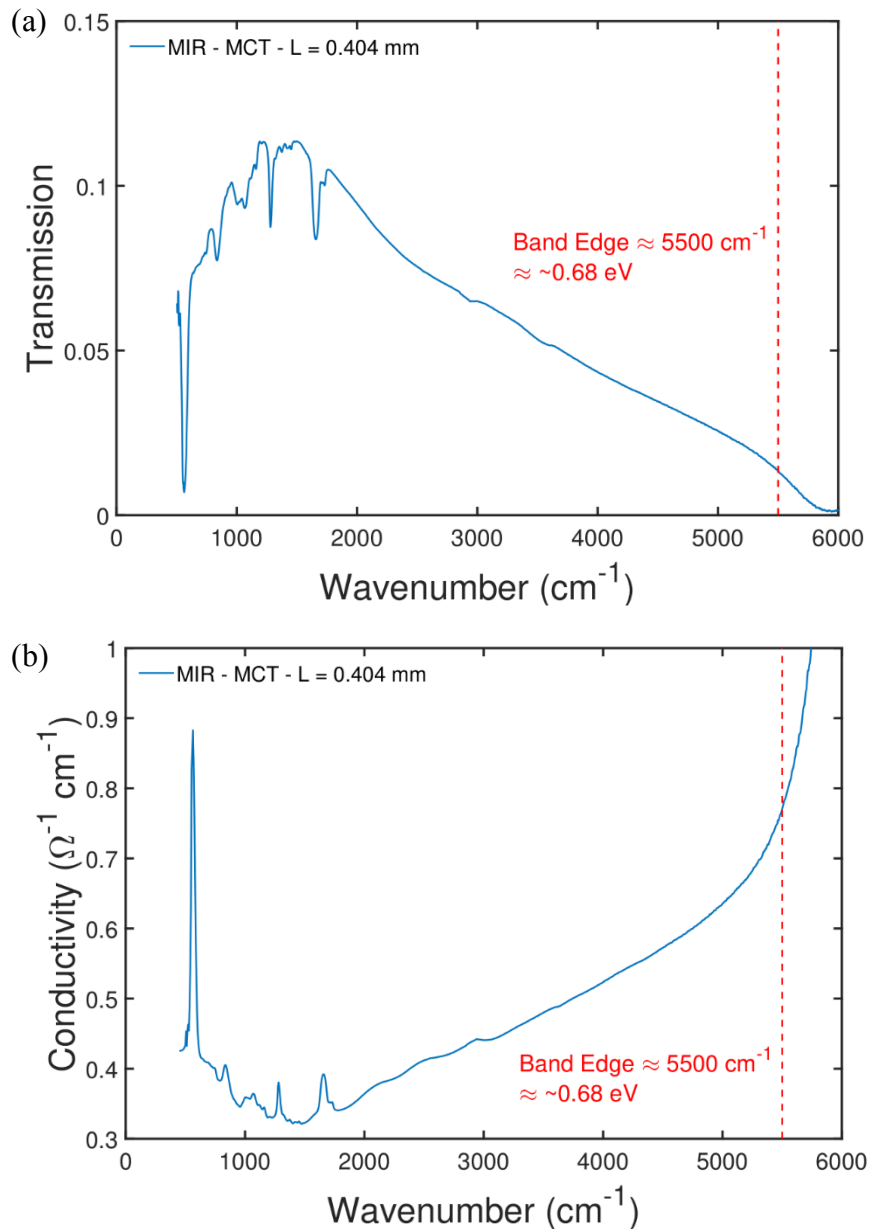


Fig. S15 High temperature magnetization of EuMn_2P_2 single crystals shows no evident magnetic phase transitions corresponding to Mn^{2+} .

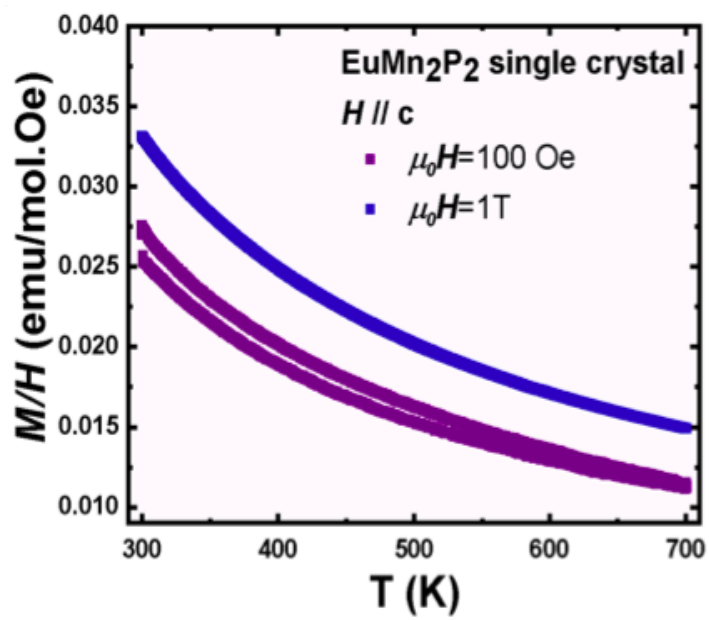


Fig. S16 Differential scanning calorimetry measurements of EuMn_2P_2 single crystals for $T=450\text{-}580\text{K}$. At $T=510\text{K}$ there is a phase transition attributed to the melting of incompletely removed Sn flux.

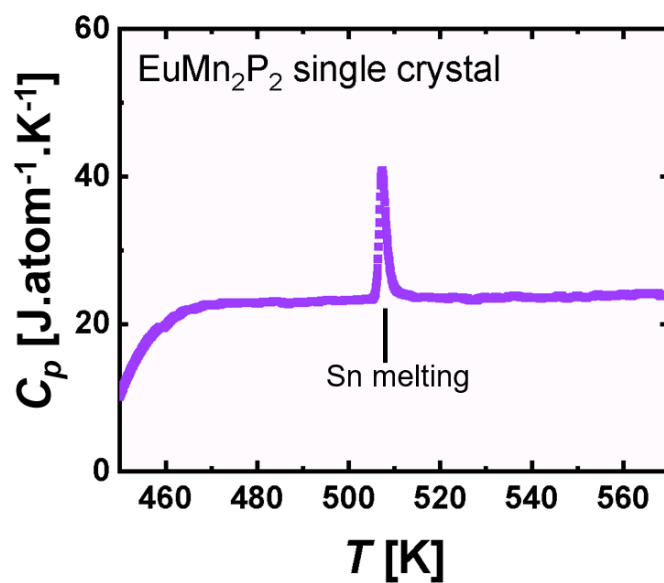


Fig. S17 Temperature dependence of magnetic susceptibility of EuMn_2P_2 single crystal **(a)** and powder sample **(b)**. The cyan-colored lines represent Curie-Weiss fitting, measured under the magnetic field used in ^{31}P NMR experiments. The blue dots in graph (b) show the Knight shift temperature dependence. The inset shows a linear relation of χ vs K which corresponds to a Hyperfine field value of $H_{\text{hf}} = 4.0 \text{ kOe}/\mu_{\text{B}}$.

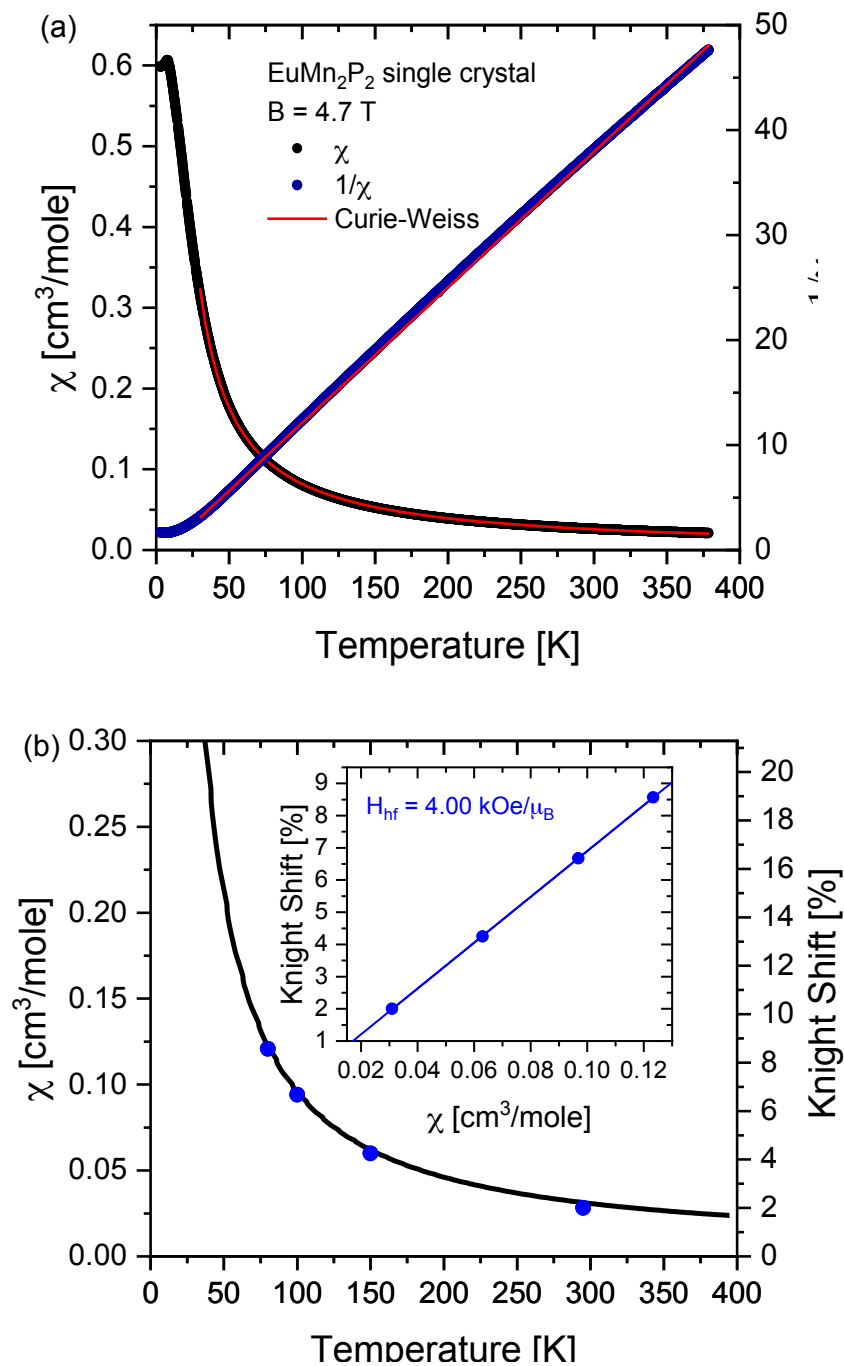


Fig. S18 ^{31}P NMR spin-lattice relaxation (Blue) and spin-spin relaxation (red) time temperature (in) dependence of EuMn_2P_2 . In range of the error bars both relaxations show almost constant behavior.

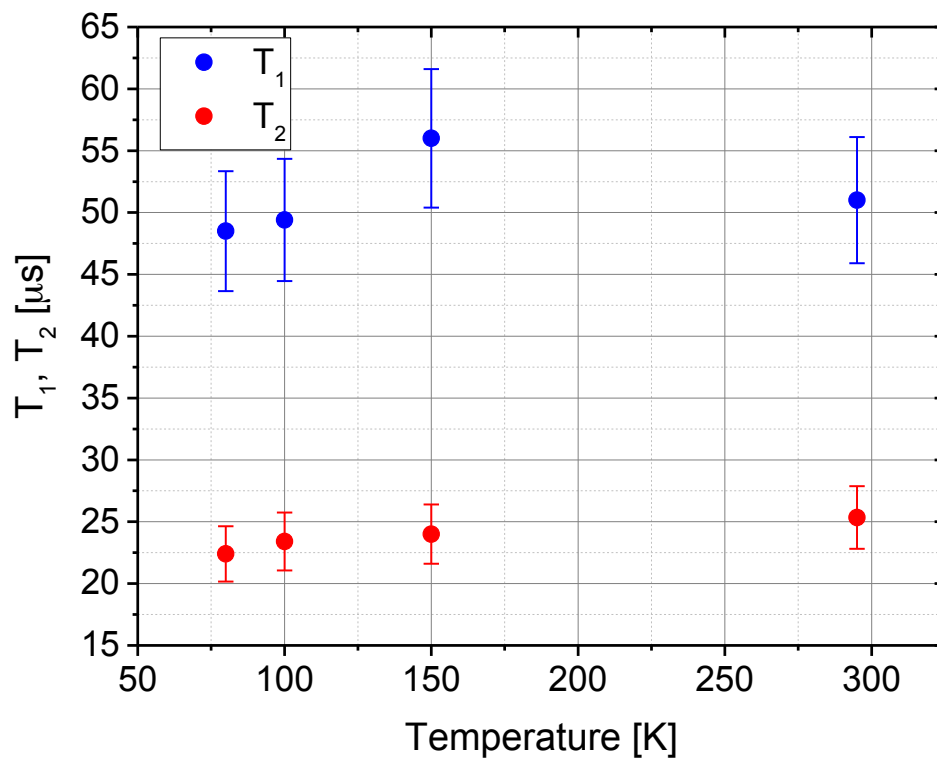


Fig. S19 Heat capacity divided by temperature as a function of temperature for EuMn_2P_2 single crystals at $\mu_0 H = 0.1 \text{ T}$ from $T = 2\text{--}300 \text{ K}$. The navy blue is the data points, and the light blue is the phonons modeled using the 2-debye models from $T = 25\text{--}300 \text{ K}$ from EuZn_2P_2 and scaled to account for the molecular mass difference of Mn and Zn. The sharp transition at $T_N = 17 \text{ K}$ is attributed to the Eu antiferromagnetic phase transition.

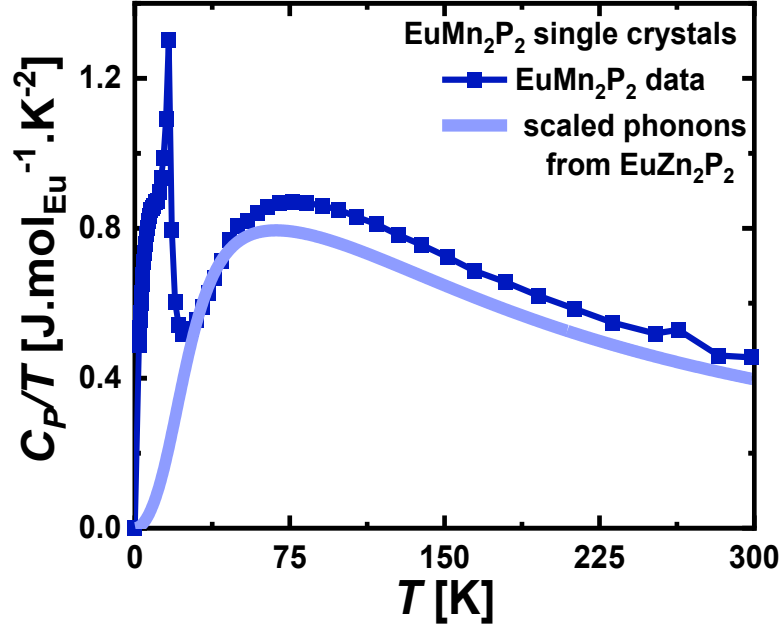


Table SVII Two-Debye model parameters used to describe the EuMn_2P_2 phonons. The oscillator strengths were fixed to those found for EuZn_2P_2 , and the Debye temperatures adjusted to account for the molecular mass difference between Mn and Zn.

Single crystals	s_{D1} (oscillator strength/formula unit)	s_{D2} (oscillator strength/formula unit)	θ_{D1} (K)	θ_{D2} (K)
EuMn_2P_2	2.8	2.83	506(2)	182.3(6)

Table SVIII. The table below lists average Mn-P bond distances (Å) in known materials and a comparison to the magnetic and conducting properties.

Avg. Mn-P bond (Å)	Material	Magnetism	Conductivity	Reference
2.27873	γ MnP ₄	AFM	semiconducting	8
2.35900	LaMnCuP ₂	weak magnetic AFM	metallic	9
2.50380	CoMnP	FM	metallic	10
2.35767	MnP	FM	metallic	11
2.51350	SrMn ₂ P ₂	magnetically-frustrated AFM	semiconducting	12
2.49050	CaMn ₂ P ₂	magnetically-frustrated AFM	semiconducting	12
2.49375	EuMn ₂ P ₂	AFM	insulating	1, This work
2.93400	Mn ₃ Ni ₂₀ P ₆	FM	metallic	13
2.38700	CsMn ₂ P ₂	AFM	metallic	14
2.27317	MnP ₄	diamagnetic	semiconducting	15
2.46400	MnSiP ₂	AFM	semiconducting	16
2.42425	ZrMnP	FM	metallic	17
2.39775	Hf _{1.04} Mn _{1.06} P _{0.90}	FM	metallic	17
2.58500	Eu ₁₄ MnP ₁₁	FM	semiconducting	18
2.37256	Mn ₃ P	helical magnetism	metallic	19
2.42678	Mn ₂ P	AFM	metallic	20
2.43800	BaMn ₂ P ₂	AFM	semiconducting	21

Fig. S20 (a) Schematic of switch on and off in regards to magnetic ordering driven non-linear current voltage response, **(b)** Key data that supports the non-linear current voltage response at temperatures below $T_N = 17\text{K}$

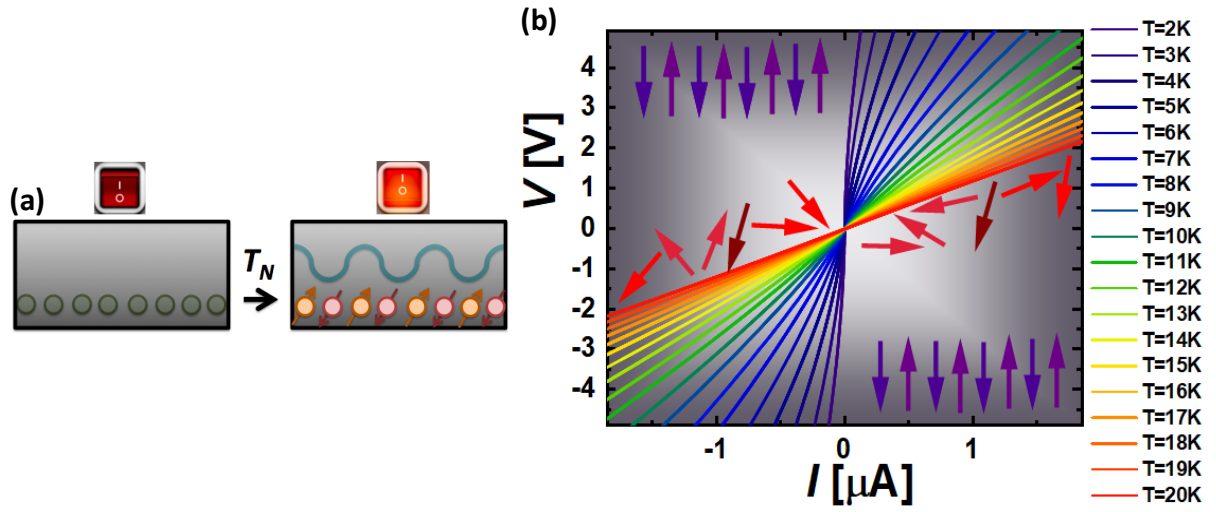


Fig. S21 (a) Resistance, dV/dI at $\mu_0 H \parallel c$ as a function of voltage $T=2K$ at $\mu_0 H=0T$. The differential resistance is broken up in channels, reflecting the low and high voltage responses respectively. Channel-1 is denoted at the low voltage and channel-2 is denoted as the high voltage response. (b) The temperature dependence of channel-1. (c) The temperature dependence of channel-2. As expected for an insulator, the decrease in resistance as the temperature is increased in (b) and (c). (d) The same normalized resistivity in (b), but plotted vs. $\frac{1}{T^{1/2}}$. A $T^{1/2}$ scaling is suggestive of 1-dimensional variable-range-hopping (VRH) in the low voltage region. (e) The same normalized resistivity in (c), but plotted vs. $\frac{1}{T^{1/4}}$. A $T^{1/4}$ scaling is suggestive of 3-dimensional variable-range-hopping (VRH) in the high voltage region.

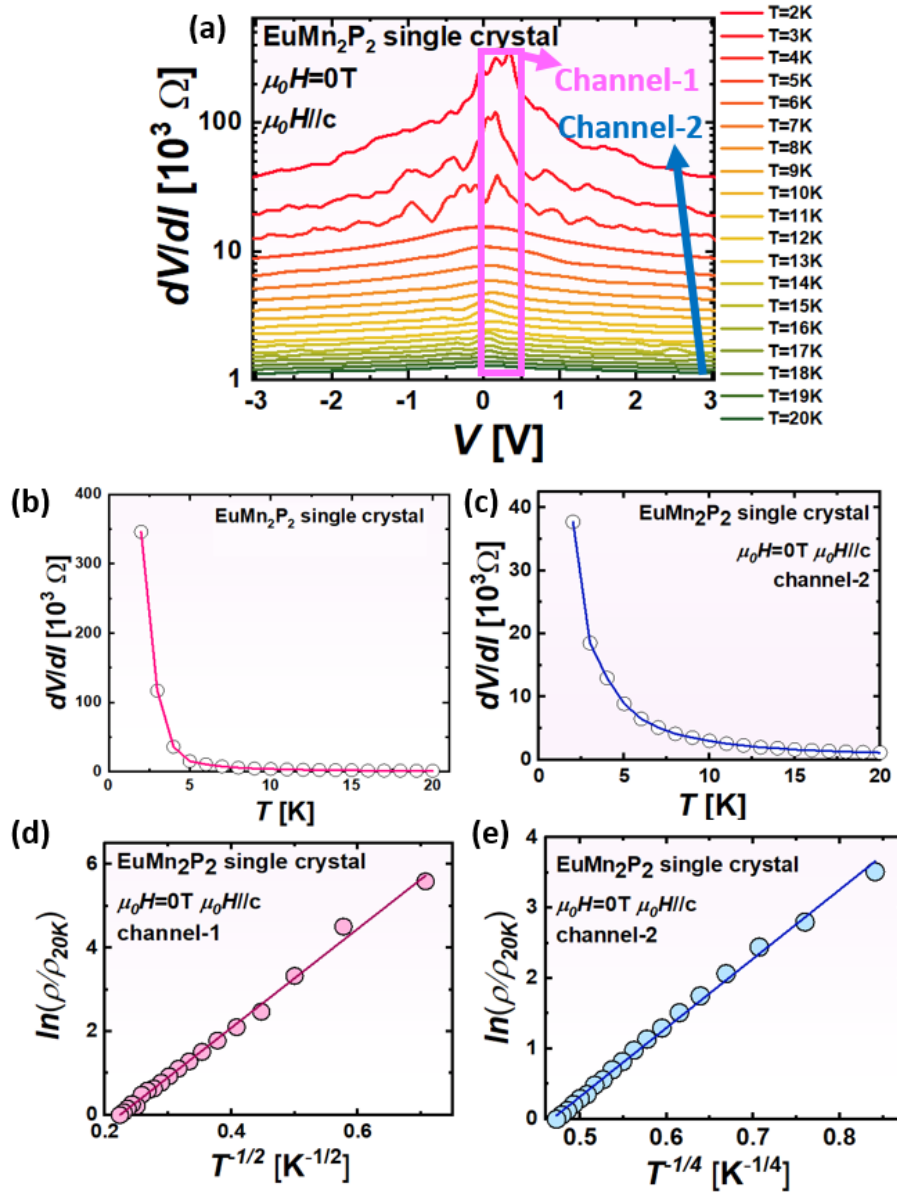
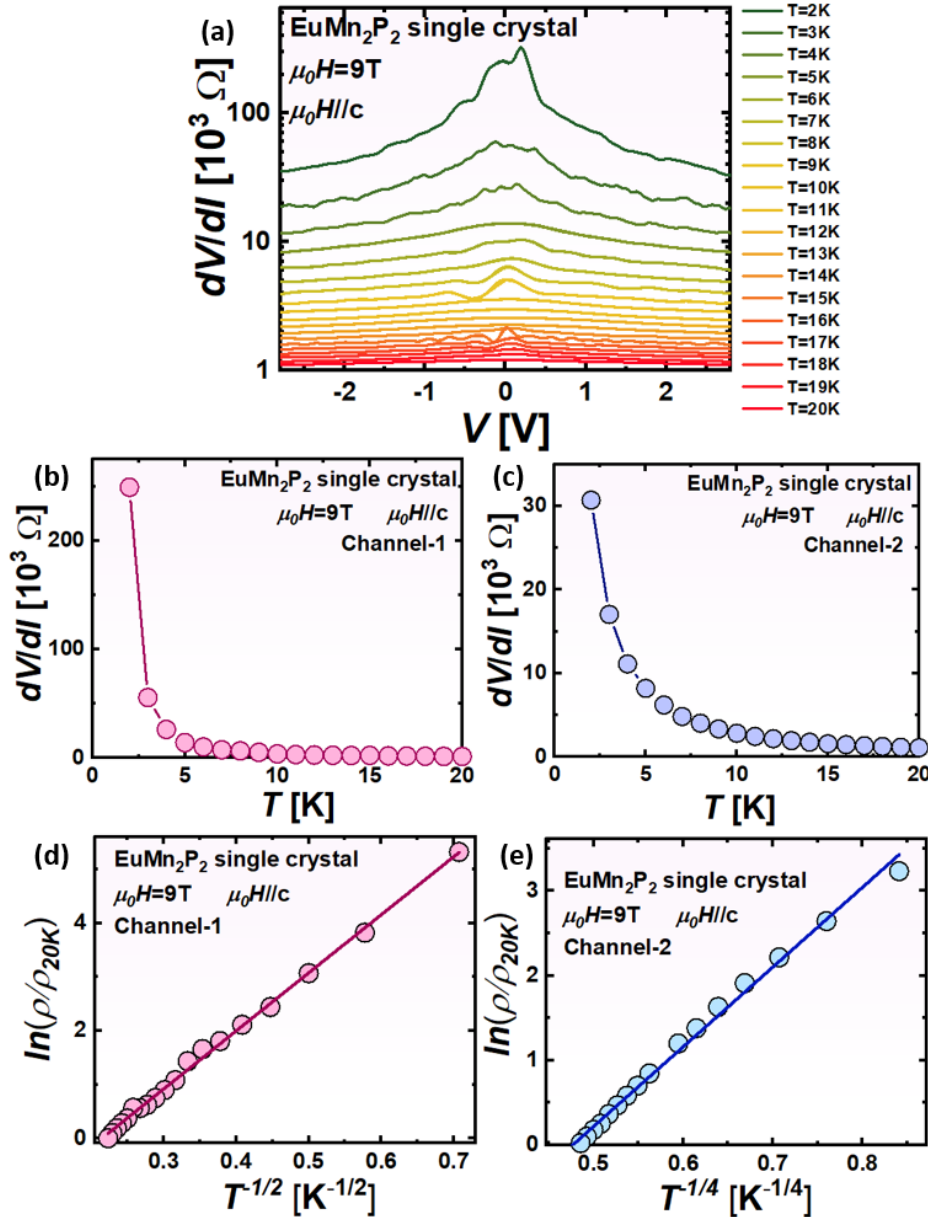


Fig. S22 (a) Resistance, dV/dI at $\mu_0 H \parallel c$ as a function of voltage $T=2K$ at $\mu_0 H=9T$. The differential resistance is broken up in channels, reflecting the low and high voltage responses respectively. Channel-1 is denoted at the low voltage and channel-2 is denoted as the high voltage response. (b) The temperature dependence of channel-1. (c) The temperature dependence of channel-2. As expected for an insulator, the decrease in resistance as the temperature is increased in (b) and (c). (d) The same normalized resistivity in (b), but plotted vs. $\frac{1}{T^{1/2}}$. A $T^{1/2}$ scaling is suggestive of 1-dimensional variable-range-hopping (VRH) in the low voltage region. (e) The same normalized resistivity in (c), but plotted vs. $\frac{1}{T^{1/4}}$. A $T^{1/4}$ scaling is suggestive of 3-dimensional variable-range-hopping (VRH) in the high voltage region.



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