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Emergent Polar Metal Phase in a Van der Waals Mott Magnet

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We report the emergence of a two-dimensional (2D) polar metal phase in van der Waals compound FePSe₃ under moderate pressures. This layered material is a Mott insulator with antiferromagnetic order under ambient conditions. We show that FePSe₃ uniquely allows tuning a 2D correlated insulator into an exotic metal state where a loss of inversion symmetry leads to periodic polar displacements of ions, within a conducting phase - a polar metal. Our combined synchrotron and neutron diffraction data allow us to present a long-sought, unambiguous high-pressure structural model and show the polar displacements of this new phase. We also observe the suppression of magnetic ordering at the insulator-to-metal transition correspondent with this structural change. Our work outlines a comprehensive temperature-pressure phase diagram of FePSe₃, combining detailed structural, magnetic and transport data. The high-pressure phase exhibits activated semiconductor behavior at high temperatures, a T^2 -dependence in its resistivity at lower temperatures - despite the conditions required for a ‘good metal’ Fermi-Liquid description not being met in this case - and a low-temperature resistivity upturn which is suppressed as the system is tuned away from the concomitant transitions. The realisation of a tunable 2D polar metal state in FePSe₃ due to the loss of its inversion symmetry combined with pressure-induced metallicity offers a promising new platform to investigate this exotic phase at accessible pressures.

I. INTRODUCTION

The study of layered van der Waals (vdW) compounds has become increasingly popular since the revolutionary discovery of graphene [1]. Magnetic vdW materials bring yet more functionality and fundamental physics to explore. In their atomically thin versions, they have attracted particular attention for applications in spintronics and magneto-optics [2, 3]. They also offer an excellent platform for studying strongly correlated electrons in low dimensions.

Many vdW magnets are also insulating, but undergo an insulator-to-metal transition (IMT) under the application of pressure [3, 4]. This makes them conceptually similar to the parent compound of cuprates: being insulating two-dimensional (2D) antiferromagnets with strongly

correlated electrons. In cuprates, varying stoichiometry drives an insulator to a ‘strange metal’, and eventually superconducting phase [5, 6]. Varying the pressure applied to layered vdW compounds offers a cleaner and more controllable way to tune physical properties, providing great insight into the underlying physics of the IMT and any exotic phases that may emerge [7–9].

One intriguing family is the transition metal chalcogenophosphates $TMPX_3$ ($TM = \text{Mn, Fe, Co, Ni, etc.}, X = \text{S, Se}$). These insulating compounds are antiferromagnets at ambient pressure, with the magnetic TM^{2+} ions forming a honeycomb structure within the vdW layers [10–12]. Previous studies have revealed pressure-induced transitions from insulating to metallic, and, in some reports, superconducting states [4, 13–18]. However, the critical underlying structural transitions remain elusive, prohibiting further investigation and engineering of this compound family. Up to this point, the literature has contained contradictory results and predictions - care must be taken in interpreting observed data, as demonstrated in this work and earlier studies [19, 20]

We selected FePSe₃ as the focus of this work, which

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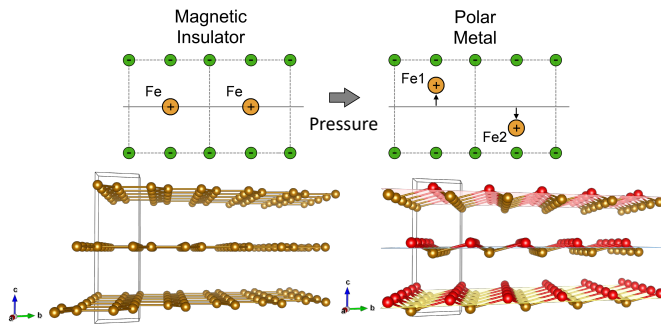


FIG. 1. **Tuning a Centrosymmetric Mott Insulator to a Correlated Polar Metal** This illustration demonstrates our key findings: tuning of a 2D material from a magnetic insulator (left) to a non-magnetic polar metal (right) via pressure. The latter is a special state that not only exhibits ordered displacements of polar ions, but also free carriers which rearrange so as to screen long-range electrical polarisation.

was recently reported to exhibit superconductivity at approximately 2.5 K and 9.0 GPa [13]. At ambient pressure, the crystal structure of FePSe_3 consists of two layers of Se on a triangular lattice, sandwiching one layer of Fe on a honeycomb lattice, with P_2 dimers located at the centre of each Fe-hexagon [10]. Both P_2 dimers and Fe ions are octahedrally coordinated with neighboring Se. The layers stack in an $ABCABC$ sequence, with Fe and P_2 aligned along axes parallel to the c -axis, which is normal to the vdW layers. FePSe_3 crystallizes in a centrosymmetric rhombohedral space group $R\bar{3}$. Below the reported T_N of 108 $\sim$ 119 K, it orders antiferromagnetically with a propagation vector of $(\frac{1}{2}, 0, \pm\frac{1}{2})$ and spin moments aligned parallel to the c -axis [12, 21].

In this article, we present a multi-probe study of FePSe_3 , combining synchrotron and neutron diffraction with resistivity measurements to investigate the effect of hydrostatic pressure on the crystalline structure, magnetic ordering, and electronic transport properties. We combine our results into a detailed phase diagram and report direct evidence for the emergence of a new polar metal phase linked to the loss of inversion symmetry in the high-pressure structure. Polar metals (Fig. 1) have become the focus of significant recent work due to their counterintuitive combination of both metallic conductivity and a non-centrosymmetric polar structure giving an ordered polarization (electric dipoles). Despite predictions in the 60s [22], only in the last decade or so have experimental realizations of polar metals been confirmed. There are few [23], if any, examples of such systems being tunable via a ‘clean’ parameter such as applied hydrostatic pressure. Thus, we present the first system in which both fundamental characteristics of the polar metal, the polar point group, and the metallicity, can be continuously and cleanly tuned in the same sample [24, 25]. The unexpected emergence of a polar metal state in FePSe_3 via pressure tuning offers a unique opportunity to study the evolution of this enigmatic state

- alongside the insulating state it is born from - in a 2D Mott magnet. We also discuss the loss of magnetic order as this state is entered, and discuss puzzles in the resistivity found in transport measurements.

II. RESULTS

In this section we first present our powder and single-crystal X-ray diffraction data with varying pressure to unambiguously demonstrate a c axis collapse at the IMT after long controversy in the literature and combining datasets to solve the high-pressure structure. We then show through neutron diffraction that magnetic order is lost entirely in this phase, and finally present electrical transport data across the phase diagram. In the Discussion we build on this range of observations to demonstrate discovery of the new polar metal phase and the various puzzles uncovered in its transport data.

A. Synchrotron Diffraction and Crystalline Structures

Powder Diffraction The powder diffraction rings of FePSe_3 loaded in helium gas as the PTM showed no significant intensity variation about the Debye-Scherrer cone, indicating that our sample loading procedure was free from strong preferred orientation. The details and subsequent data reductions converting the original diffraction rings into integrated diffraction intensities as a function of 2θ are given in the SM [26].

Fig. 2 (a) presents an evolution of the post-processed powder diffraction patterns across a range of pressures from 0.7 to 34.1 GPa. The integrated intensities are normalized to the maximum intensity for each pressure point. All datasets are shifted vertically by an offset proportional to the pressure value.

The data show a dramatic shift in the 2θ position of a number of peaks between 7.6 and 8.1 GPa which indicates a distinct phase transition. The transition is sharp with little evidence of phase co-existence. There is no peak broadening associated with the transition, suggesting that a single-phase model with minimal distortion may be used to determine the crystal structure of the high-pressure phase. No further phase transition was evident up to the highest measured pressure (34.1 GPa). The low- and high-pressure phases will subsequently be referred to as LP and HP, respectively. Data assigned to these two phases are plotted in blue and orange in Fig. 2 (a) for a straightforward comparison.

The LP Bragg peak at the lowest 2θ is indexed in the refinement as 003, as marked in Fig. 2 (a). It is reasonable to assume that the crystal structure does not undergo a substantial reconstruction during the transition from LP to HP. Consequently, the position of this peak can be assumed to be due to the change of the interplanar distance at all pressures. The peak undergoes a drastic

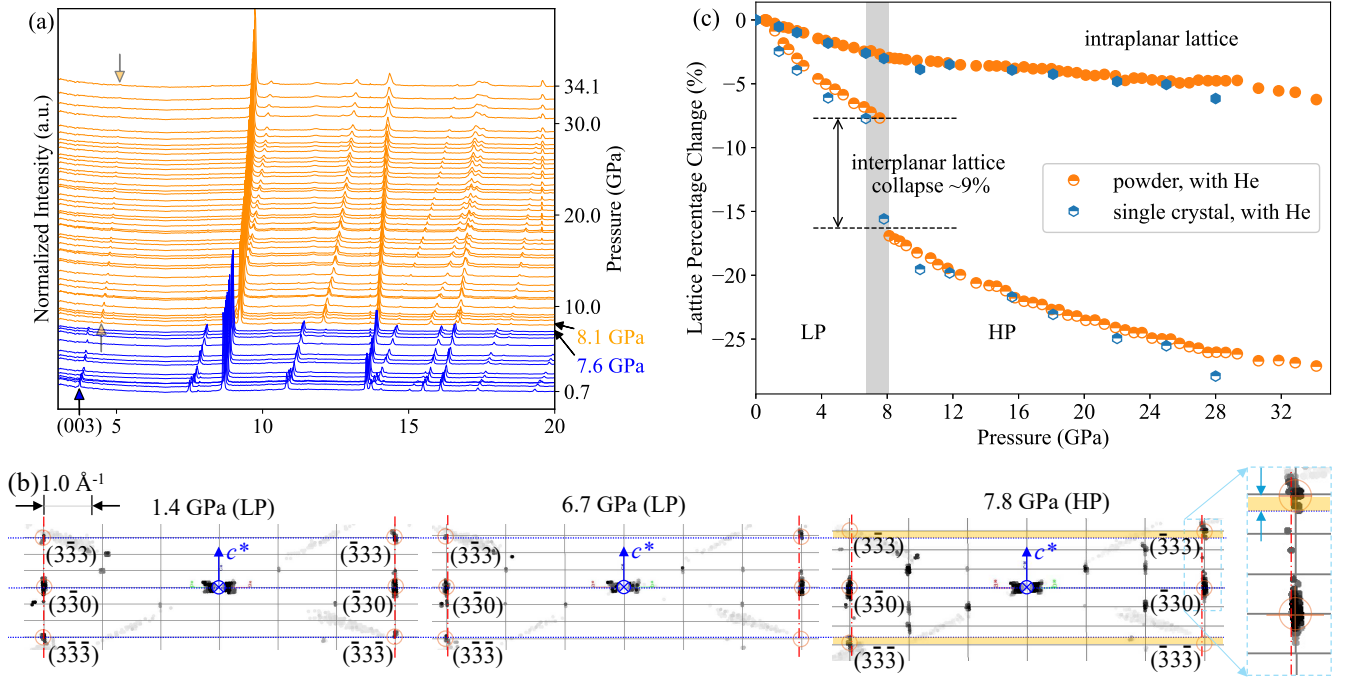


FIG. 2. Powder and Single-Crystal Synchrotron Diffraction Results and Lattice Parameters Evolution under Pressure (a) Integrated diffraction intensities as a function of 2θ for FePSe_3 powder sample in a DAC loaded with helium gas as a pressure-transmitting medium (PTM). For each pressure, the intensity was normalized to its maximum, and then an offset proportional to pressure values was added. Blue and orange represent data assigned to the LP and HP phases, respectively. (b) Slices of $(h\bar{h}l)$ planes at the lowest pressure (1.4 GPa, LP), before (6.7 GPa, LP) and after (7.8 GPa, LP) the phase transition pressure. The sub-pictures are all scaled in $\text{\AA}^{-1}$. The red and blue lines are given to guide the eye to show the separation distance at the 1.4 GPa, LP phase. The yellow color blocks highlight the difference in the separation of c^* . A zoom-in view is provided at 7.8 GPa to better illustrate the abrupt changes in c^* . (c) Pressure-induced relative changes of the lattice parameters in reference to the ambient-pressure structure. Orange and blue represent powder and single crystal cases, respectively. The grey-shaded region indicates the interplanar lattice collapse transition pressure.

change at the phase transition, indicating that the interplanar spacing collapses at the LP-HP transition.

The LP data were analyzed using the ambient pressure structure model, which had the hexagonal $R\bar{3}$ space group [11]. The refined lattice parameters at the lowest pressure point (0.7 GPa) were determined to be $a = b = 6.238(2) \text{ \AA}$ and $c = 19.49(1) \text{ \AA}$, which are in reasonable agreement with the literature values at ambient pressure ($a = b = 6.265(6) \text{ \AA}$, $c = 19.80(2) \text{ \AA}$, and $\gamma = 120^\circ$). The HP data were analyzed using the structural model we obtained from our single-crystal synchrotron studies.

Single-Crystal Diffraction In the single-crystal experiment, we collected high-quality diffraction data from 1.4 to 28.0 GPa. Slices of the $(h\bar{h}l)$ scattering plane exhibit strong-intensity peaks which are indexed as $3\bar{3}l$ and $3\bar{3}l$ where $l = 0, \pm 3$, as shown in Fig. 2 (b). The slices are plotted to scale in $\text{\AA}^{-1}$. The intensity of the peaks is consistent with the structure factor analysis. The separation between reflection index points peaks along the $[001]$ and $[1\bar{1}0]$ directions unambiguously reflects the changes of the inter- and intraplanar lattice parameters,

respectively. An abrupt increase in separation along the $[001]$ direction is clearly seen from 6.7 to 7.8 GPa (highlighted with yellow color blocks), while changes along the $[1\bar{1}0]$ direction remain negligible throughout the entire pressure range. This observation in the raw data, independent of any structural model, provides definitive and model-independent proof of the interplanar collapse with applied hydrostatic pressure in FePSe_3 .

The lattice parameters were initially refined based on indexed reflections. We used the ambient-pressure lattice parameters of LP as the starting point to refine the cell at the lowest pressure (1.4 GPa). No space group symmetry was forced on the refinement at this step. The well-indexed reflection peaks and refined parameters ($a = 6.185(4) \text{ \AA}$, $b = 6.176(3) \text{ \AA}$, $c = 18.89(3) \text{ \AA}$, $\alpha = 90.28(1)^\circ$, $\beta = 89.90(1)^\circ$ and $\gamma = 119.99(7)^\circ$) are consistent with the $R\bar{3}$ symmetry of LP with tolerable deviations. We consistently refined the lattice parameters for all pressures using the cell parameters at the previous pressure point. The results are summarized in the SM [26].

Fig. 2 (c) summarizes the relative changes of lattice parameters as a function of pressure, resulting from our

comprehensive synchrotron studies. It can be seen that the intraplanar lattice parameters decrease smoothly at an average rate of $\sim 0.17\%$ per GPa. The interplanar distance, by contrast, declines more rapidly at an average rate of $\sim 0.86\%$ per GPa, followed by an abrupt drop of approximately 9% across the LP-HP transition. After the transition, c continues to decrease at a rate of $\sim 0.50\%$ per GPa up to the highest pressure. The LP-HP transition region is shaded to guide the eye.

Thorough analysis of the single-crystal diffraction is the most reliable way to assign the correct space group and determine the structural model. Data below 6.7 GPa were well-refined with the LP structure model of the $R\bar{3}$ space group. Refinements of the data above 7.8 GPa with the $R\bar{3}$ space group gave poor quality parameters. As can be seen from Fig 2 (b), there is no substantial change in a and b , and merely a sudden change of interplanar spacing in c . Hence it is sensible to keep using the hexagonal cell description and check among the subgroups of $R\bar{3}$ with k -index=1, namely $R\bar{3}$, $P\bar{1}$ and $P1$. After careful inspection, we find that there are no new peaks at new d spacings, but peaks with equivalent d spacing present different structure factors. Further analysis determined that this is due to a Friedel Pair not showing the same observed structure factor, namely $I(hkl) \neq I(\bar{h}\bar{k}\bar{l})$. This can only be explained by the loss of centrosymmetric symmetry, leaving $R\bar{3}$ and $P1$ as the only possible solutions. Meanwhile, we still only observe intensities at $-h + k + l = 3n$, suggesting that the hexagonal setting remains. Consequently, we demonstrate the sole possible solution for the HP phase to be the $R\bar{3}$ space group. All peaks could be indexed and refined with the $R\bar{3}$ space group. Two representative sets of crystallographic information for the LP and HP phases, including refinement parameters, are given in Table I. The atomic position information, observed versus calculated structure factors, as well as the analysis of Friedel pairs graphically are given in the SM [26].

The solved $R\bar{3}$ model of the HP phase can be used equally successfully to refine powder data, providing an unambiguous HP phase structure model.

TABLE I. **Reduced crystallographic data in LP (4.4 GPa) and HP (10.0, 28.0 GPa) phases.** The parameters are obtained from the single-crystal synchrotron diffraction experiment. Here we use the primitive hexagon cell description for the lattice parameters.

		4.4 GPa	10.0 GPa	28.0 GPa
Lattice parameters	Space group	$R\bar{3}$	$R\bar{3}$	$R\bar{3}$
	$a = b$ (Å)	6.0971(6)	6.0155(7)	5.895(3)
	c (Å)	18.279(19)	15.34(6)	14.1(2)
	Volume (Å ³)	588.5 (6)	480.8 (18)	423(7)
Refinement parameters	$R[F^2 > 2\sigma(F^2)]$	0.0038	0.090	0.0922
	$wR(F^2)$	0.095	0.221	0.2401
	S	0.88	2.1	2.3
	Reflections	36	125	134

Key Structural Differences Fig. 3 exhibits the crystalline structure of LP and HP FePSe₃ side by side. (a) and (d) provide an isometric view of the bulk structures. Fe, P and Se atoms are shown in brown, gray and green respectively. We represent the Fe-hexagon network with solid brown lines. The interplanar distances shrink substantially from LP to HP, while the stacking geometry $ABCABC$ remains unchanged. Normal to the vdW layers, as shown in (b) and (e), the Fe-hexagon network remains undistorted from LP to HP. The $C3$ rotation symmetry is thus preserved across the transition.

The HP phase differs from the LP phase primarily in loss of the inversion symmetry, resulting from the buckling of the Fe-hexagons along the c -axis and the collective displacement of Se sites. The LP phase only possesses one Wyckoff site for Se (18f), thus forming perfect octahedral coordination with Fe or P₂ positioned in the center, as is shown in Fig. 3 (c). Within each vdW layer, Fe atoms and the center of the P₂ dimers define one plane at $z = \frac{1}{6} + \frac{n}{3}$, n : integral). Se atoms at $z \pm \Delta$ positions form two sandwiching planes, represented by light and dark green colors together with + and - signs for $z + \Delta$ and $z - \Delta$, respectively. We also show a normal view to the ab plane of one Se-octahedron on the top-right corner of Fig. 3 (c). The bonds between Fe or P₂ and the three Se atoms at the same plane ($z + \Delta$ or $z - \Delta$) are shown by contrasting colors.

In the HP phase, we observe that the Fe planes buckle slightly, resulting from Fe₁(3a) moving upward while Fe₂(3a) move downward by $\sim 1\%$ in lattice unit along the c -axis, in reference to their center position. The Se atoms also split into two distinct Wyckoff sites, Se₁ (9b) forming one Se plane and Se₂ (9b) forming the other, as highlighted in Fig. 3 (d-f) by light and dark green colors respectively. The relative displacement of the two Se planes to the center of the Fe plane differs slightly. Within each [FeSe₆] octahedron, Se₁ and Se₂ are rotated coherently within the plane but in opposite directions between the planes. Meanwhile, the [P₂Se₆] octahedron remains nearly undistorted. We indicate the local in-plane collective rotations of Se atoms with orange and blue along one axis parallel to the c -axis in Fig. 3 (f). In addition, Fig. 3 (g) exhibits how the octahedron's local distortion propagates within and across the vdW planes in a bulk structure. We use red, blue and gold hexagon networks to present the Fe-hexagon in ABC stacking sequence. It can be seen that each neighboring Se-triangle rotates in the opposite direction within one Se plane. These structural distortions break the inversion symmetry of the crystalline structure, leaving FePSe₃ HP phase in the $R\bar{3}$ space group with a polar point group 3, and also contribute to a net dipole moment along the c -axis.

B. Neutron Scattering and Magnetic Properties

Fig. 4 (a) shows the powder neutron diffraction patterns after background subtraction at temperatures

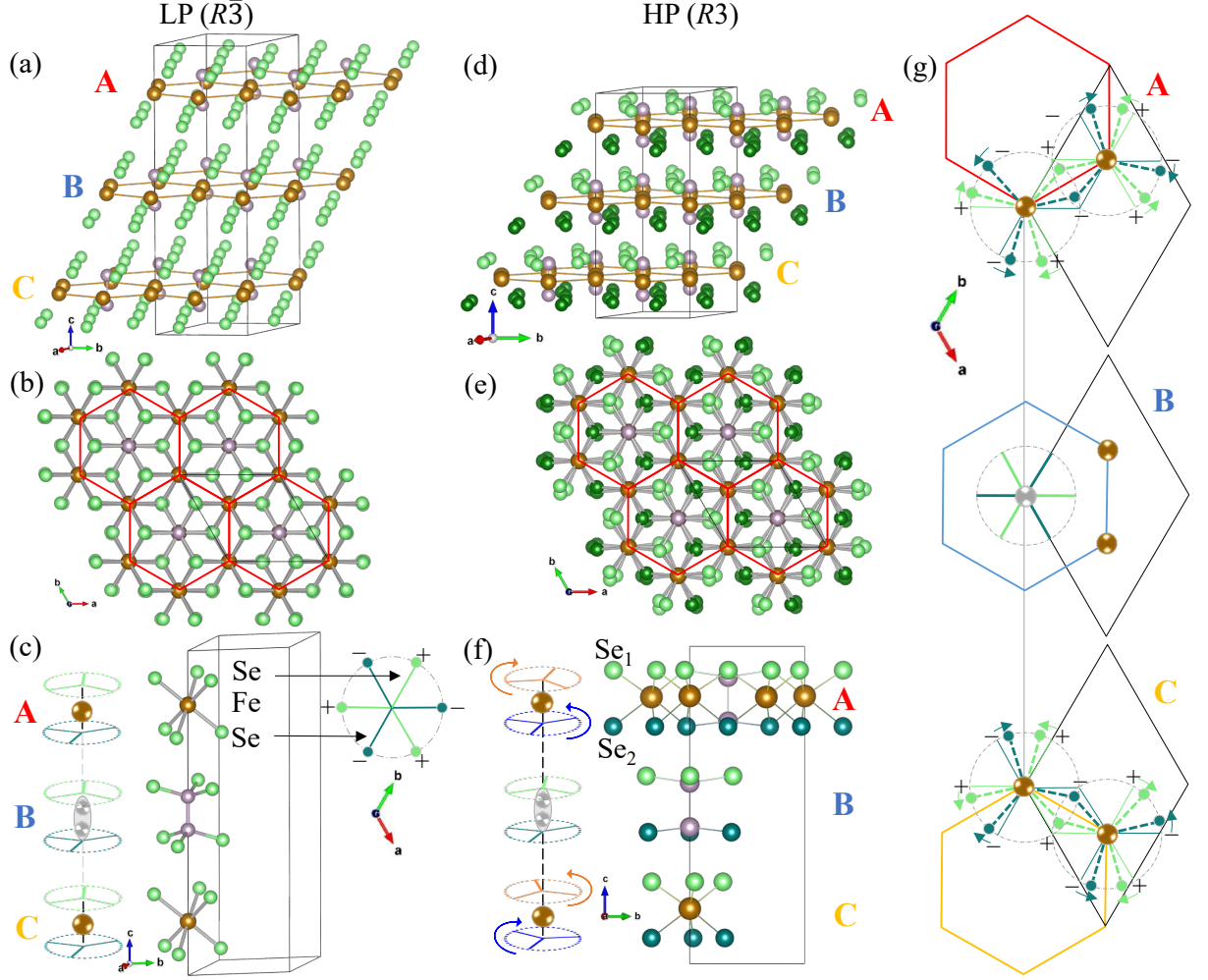


FIG. 3. **Crystalline structures of the ambient-/low-pressure (LP) and high-pressure (HP) phases of FePSe₃** Fe: brown; P: grey; Se: green. (a, b) are the structural isometric view with a minimal stacking unit of ABC triple layers; (b, e) display the view normal to the vdW planes, with the red solid lines indicating the Fe-honeycomb network; (c, f) present the FeSe₆ and P₂Se₆ octahedra along the axis parallel to c . The cartoon aside depicts Se planes in both phases. The light green and dark green with + and - signs represent the Se atoms in the plane above or below the plane defined by Fe or P₂ dimer within each vdW layer. The collective displacement of Se atoms in the HP phase is demonstrated with different colors and arrows. (g) shows the normal view of the three individual stacking layers in ABC ordering. The collective rotation of each Se plane is indicated with arrows in different colors.

above (HT) and below (LT) the magnetic transition in red and blue respectively, for pressure values of 1.2, 4.1 and 8.3 GPa. In addition to the sample signals, there were visible peaks originating from the diamond anvils, as marked with green dotted lines in the graph. More details about background subtraction and diamond peak identification can be found in the SM [26].

The HT data (red) contained only the nuclear peaks. The indexed crystal structures were consistent with our synchrotron results. The pressure values for the neutron measurements were then determined using the structure parameters resulting from the synchrotron data. The LT data (blue) included both nuclear and magnetic peaks. Since the nuclear peaks remained nearly unchanged with varying temperatures at each pressure point, we sub-

tracted the HT data from the LT data to separate the magnetic peaks. The resulting temperature subtractions are shown in Fig. 4 (b).

At 1.2 and 4.1 GPa, pressures below the LP to HP crystalline transition, magnetic peaks can be seen clearly and well-indexed with the ambient-pressure magnetic structure model by Wiedenmann *et al.* [12] assuming no substantial changes in magnetic structures. The Bragg peaks corresponding to the $(\frac{1}{2}, 0, \frac{1}{2})$ and $(\frac{1}{2}, 0, \frac{1}{2})$ antiferromagnetic propagation k -vectors are marked in Fig. 4 (b).

The magnetic unit cell doubles along a and c compared to the nuclear one, as is shown in Fig. 4 (c). Only Fe²⁺ atoms with opposing spin moments in black and white arrows are shown. The nuclear unit cell is displayed with solid black lines as a reference. We demonstrate the vdW

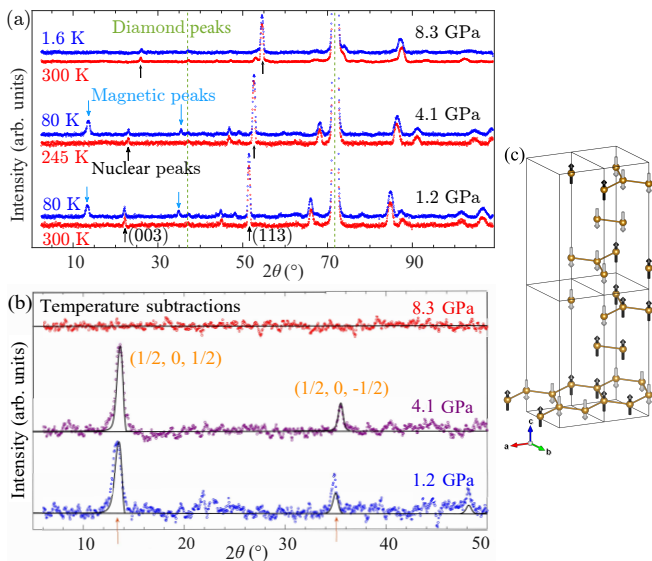


FIG. 4. Powder Neutron Diffraction Patterns and Magnetic Structure under Pressure (a) Diffraction data taken at temperatures well above (red) and below (blue) the magnetic transition temperature at pressures of 1.2, 4.1 and 8.3 GPa after background subtraction. Nuclear, magnetic peaks for FePS₃, and peaks originating from the diamond anvils are indicated with black, cyan and green, respectively. (b) Resulting diffraction intensity obtained by subtracting the intensity at high temperature (red) from that at low temperature (blue) for each pressure point. Magnetic peaks consistent with the ambient-pressure $(\frac{1}{2}, 0, \pm\frac{1}{2})$ magnetic propagation vectors are marked with orange arrows. Solid black lines represent refinements of the data. (c) Magnetic ordering of Fe atoms in the LP phase with the nuclear unit cell, in solid black lines as a reference. The black and white arrows on Fe²⁺ atoms show opposing spin moments, parallel to the *c*-axis. Not all magnetic atoms are shown in the magnetic cell, which are four times the nuclear cell.

layer in the bottom layer while only showing the magnetic atoms within one cell for the remaining layers. The moment direction is normal to the *ab* plane, consistent with the ambient-pressure data. The FM zigzag chains are along the *b* direction, and are coupled with one another antiferromagnetically within the plane.

Data at a constant counting time were also collected while warming from the base temperature. We were thus able to extract the temperature dependence of the magnetic peak intensity at each pressure. At 1.2 GPa, the intensity evolution was fitted to a honeycomb-lattice 2D Ising model [27, 28], with *S* set to 2. This fit yields an estimated Néel temperature T_N of approximately 128 K (details in the SM [26]), slightly higher than the previously reported value of 108 ~ 119 K at ambient pressure [12, 21]. Data at 4.1 GPa were too sparse for a meaningful fit but still consistent with the trend of increased magnetic transition temperature with elevating pressure.

The magnetic moment resulting from refinements was

aligned parallel to the *c*-axis with a magnitude of $3.8(2) \mu_B$. The magnitude is lower than that reported in Ref. [12] ($m_0 = 4.9 \mu_B$). We recognized that some of the difference may be due to peak asymmetry, as the refinements in Fig. 4 do not fully capture the integrated intensities of the magnetic Bragg peaks. It is possible that the magnetic structure developed some disorder under pressure and that some Bragg intensity was lost into diffuse scattering which was subsequently removed using the background subtraction method. Fig. 7 in the SM [26] shows some evidence for diffuse scattering in the LP phase.

At 8.3 GPa, at which point the crystal structure has already changed to the HP phase with *R*3 symmetry, there was no signature of any magnetic ordering at low temperatures, down to 1.6 K. Calculating the magnitude of scattering expected from typical Fe moments and comparing with the size of signal observed in our previous FePS₃ work [29], we can be confident that any signal would be visible above the level of noise in the data. We cannot rule out the presence of magnetic diffuse scattering as this might appear as background. However, no clear diffuse signal with an obvious *Q*-dependence was observed at any temperature in the HP phase meaning that, as shown in the SM [26], any diffuse scattering at the 8.7 GPa must be very weak and featureless. This implies that any surviving magnetic moments must be essentially uncorrelated, leading to a paramagnetic phase. We note that we cannot distinguish a paramagnetic from a non-magnetic phase where Fe local moments are lost. However, we deem it unlikely for magnetism to suddenly turn disordered down to 1.6 K in the HP phase.

C. Transport Measurements and Electronic Properties

Fig. 5 (a) presents the temperature dependence of resistivity for a single-crystal FePS₃ from 6.0 up to 13.5 GPa. The magnitude of the resistivity decreased by four orders of magnitude at room temperature from 6.0 to 7.0 GPa, indicating an insulator-to-metal transition (IMT). Another single-crystal sample measured from 2.0 up to 14.0 GPa, shown in the SM [26], exhibited a consistent IMT between 6.0 and 8.0 GPa.

. Before the transition Up to 6.0 GPa, FePS₃ exhibits an insulating character. In the high-temperature region ($T > 250$ K), where the conduction is dominated by thermally activated behavior, the data can be well-described by an Arrhenius-type exponential expression, $\rho(T) \sim e^{E_a/(k_B T)}$. Plotting $\ln(\rho)$ against $1/T$ allows us to perform a polynomial fit (detailed in the SM [26]), which yields an extracted activation energy E_a of 0.24, 0.21 and 0.11(3) eV for 2.0, 4.0 and 6.0 GPa.

At 6 GPa, the resistivity gradient shows abnormal changes with a change of sign in the intermediate temperature range which could potentially result from underly-

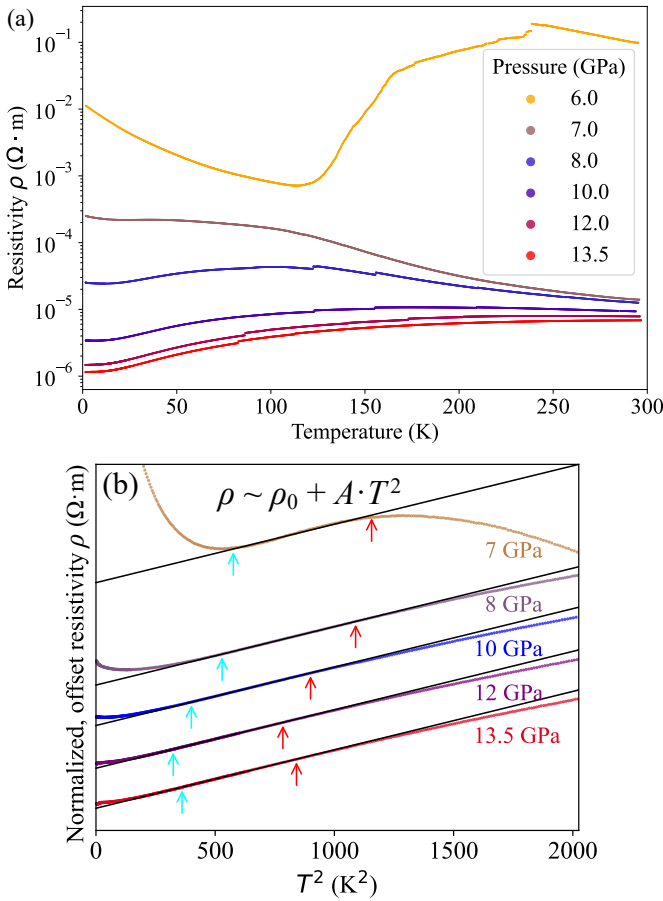


FIG. 5. **Single-Crystal Resistivity Measurements under Pressure** (a) The resistivity ρ of a FePSe₃ single crystal, loaded in a Cubic Anvil Cell with glycerin as the pressure-transmitting medium. (b) Normalized and offset resistivity as a function of T^2 , a Fermi-liquid temperature power law, below ~ 40 K for FePSe₃. The fitted curves are plotted in solid black lines with the best-fitting T^2 region marked with arrows for each pressure.

ing magnetic transitions or Fermi-surface reconstruction in proximity to the LP to HP transition. Further analysis and repeated thermal cycling would be needed to uncover more details. We also observed hysteresis in the 6.0 GPa data while cooling down and warming up, suggesting the IMT to be strongly first-order. Such hysteresis was not observed in data collected at other pressures.

. **After the transition** Above 6 GPa up to the highest pressure point, we did not observe superconductivity in two independent measurements using different samples from the same batch for pressures up to 13.5–14.0 GPa and temperatures down to ~ 1.6 K. This was in contrast to previous report of superconductivity with a T_c of 2.5 K above 8 GPa [13]. Our crystallographic data showed our sample to be of good crystalline quality and the structural model to be correctly interpreted. Our samples have equivalent resistivity magnitudes (and

hence inferred cleanliness) to those in the reference. We make further comments in the SM [26]. In the main text, we focus on the newly uncovered exotic transport behaviors enriching the T - P phase diagram.

In the 7.0, 8.0, and 10.0 GPa data, we find that the resistivity increases initially upon cooling from 300 K, similar to an activated behavior of an insulator or semiconductor. The resistivity shows a broad maximum at approximately 40, 100, and 190 K for the 7.0, 8.0, and 10.0 GPa data respectively. Upon further cooling, the resistivity decreases showing metallic behaviors. At higher pressure points (12.0 and 13.5 GPa) such a trend was not discernible within the measured temperature range. The resistivity decreases upon cooling from 300 K.

Further analysis of the conducting phase finds that the resistivity can be uniquely described by $\rho(T) = \rho_0 + A \cdot T^2$ at temperatures below ~ 40 K, as shown in Fig. 5 (b). We note that the A -coefficient of the T^2 term decreases significantly with increasing pressure. The T^2 fits are detailed in the SM [26].

This behavior, however, terminates with an upturn in the resistivity at even lower temperatures. The low-temperature resistivity upturn is observed around 20 K, 10 K and 6 K in the 7.0, 8.0 and 10.0 GPa data, respectively. The upturn is quickly suppressed with elevated pressure, becoming completely absent above 10.0 GPa.

III. DISCUSSION

Drawing together our results from multiple experimental probes, we are able to complete the comprehensive temperature-pressure phase diagram for FePSe₃ - Fig. 6. The color blocks represent different phases, with gradient-shaded boundaries to guide the eye.

Our synchrotron studies establish that FePSe₃ at 300 K undergoes a $R\bar{3}$ (LP) to $R3$ (HP) structural transition at 7–8 GPa of hydrostatic pressure. We show this to A significant interplanar collapse along the c -axis occurs during this LP-HP transition. The HP phase ($R3$ space group) loses the unit cell's inversion center, leading to out-of plane displacements of the Fe atoms in the metallic phase. The unusual transition from a higher to lower symmetry space group under pressure is unique in FePSe₃, making it different from other compounds in this family.

Our resistivity measurements reveal exotic transport properties changes in the phase diagram. The LP phase remains insulating up to 6 GPa, with the activated band gap shrinking in application of pressure, compared to the ambient-pressure literature value, which agrees qualitatively with the previous report [13]. We also observe an insulator-to-metal transition (IMT) between 6 and 8 GPa, as marked with the yellow dotted arrow in Fig. 6. The IMT transition pressure corresponds to the LP-HP structural transition pressure. We note, however, the high-temperature region of the HP phase still shows activated semiconducting behavior.

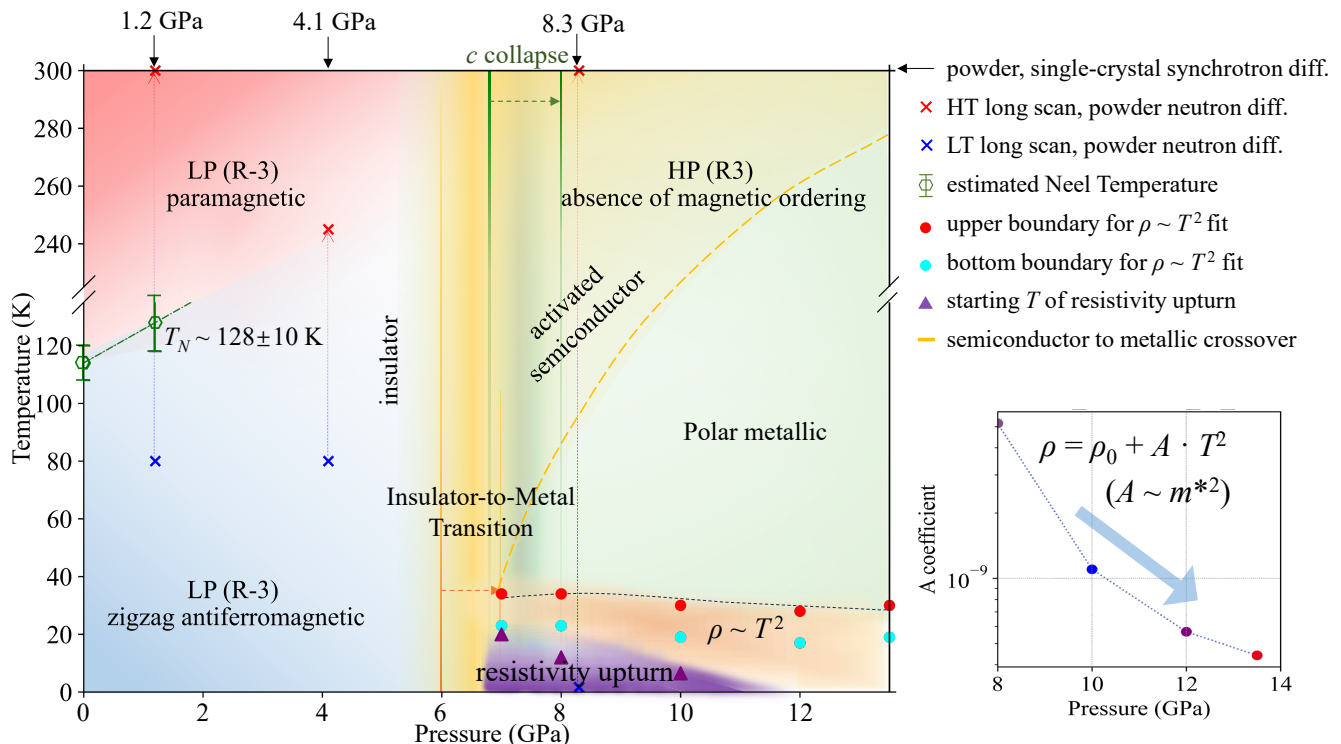


FIG. 6. T - P phase diagram of FePSe_3 constructed from multiple probing techniques. The green open circles correspond to the antiferromagnetic transition temperature. The one at ambient pressure is from literature (magnetic susceptibility measurement), and the one at 1.2 GPa is derived from the intensity versus temperature change in our neutron diffraction data (fitted curve in the supplementary). The shaded light orange region refers to where the resistivity follows the $\rho \sim T^2$ behavior with the upper boundary in red, and the lower boundary in cyan. The region under the purple triangles is where the resistivity upturn was observed. The inset at the bottom right shows the fitted coefficient A of the T^2 term as a function of pressure for the shaded orange region.

At lower temperatures of the HP phase, FePSe_3 presents an unexpected T^2 resistivity temperature dependence. The fitted boundary of this temperature-pressure region is marked with dots in Fig. 6. should be underlined that the A -coefficient of the T^2 term decreases with increasing pressure, indicating a substantial decrease in the effective mass of the charge carriers.

An upturn in the resistivity, deviating from this state, emerges at even lower temperatures, marked with purple. Such deviation is more discernible in proximity to the AFM transition and is fully suppressed at 11~12 GPa. No bulk superconductivity was observed in the HP phase up to the highest pressure (~14 GPa) in two independent measurements of FePSe_3 samples from the same batch.

A. Discovery of Polar Metal Phase

We identify that FePSe_3 undergoes an IMT concomitant with the LP-HP structural transition. One aspect is that the associated interplanar c -collapse coincides with the IMT. Similar features were reported in MnPS_3 , FePS_3 and NiPS_3 previously [4, 14, 30], but should not be considered as universal across this family, as recent cases

of VPS_3 [31] and NiPSe_3 [15] exhibit Mott transitions without structural changes.

Our key result is that, due to a periodic displacement of the Fe sites as the system enters the HP phase, FePSe_3 becomes a polar metal at readily accessible pressures (less than 10 GPa). As the state can be easily and cleanly tuned, the community now has a new handle on the little-understood physics of these rare systems. That is, the material can be studied in both polar and non-polar configurations, at all temperatures, and direct comparisons can thus be made. In effect, this allows a control to any tests, and lets us understand the initial state that the new physics emerges from. We discuss more details in the following two aspects.

. Broken Symmetry and Formation of Net Dipole in the HP phase We identify the broken inversion symmetry and a polar point group symmetry in the HP structure from the synchrotron diffraction data. In the initial LP phase, there is a single symmetry-constrained Fe site in the unit cell, and so all Fe atoms sit exactly on ab planes.

In the HP phase however, the Fe-hexagon buckling has been observed in our results, where two separate Fe sites

are present in the structure, as a result of an unusual lowering of symmetry under compression. As demonstrated in Fig. 7, the two sets of irons move out-of-plane in opposing directions while the z -centre of the ab plane remains as that in the LP phase. We demonstrate the relative movement of one set of Fe moving up and one moving down in different colors.

Our detailed analysis of the atomic displacement suggests local net dipole moments to form in the HP phase along the c -axis due to the Fe buckling. Additionally, we observed relative displacement of the Se and P atoms but the amplitude is rather subtle. We believe further investigations are needed to identify whether the system stabilizes in an anti-ferroelectric state or could potentially allow for a net dipole moment forming as a result of ligand atom displacement. To date, polar metals can be divided into those that are essentially ferroelectric semiconductors and have significant carrier densities doped into the system to give a weakly metallic state or metals where no bulk polarization (effectively capacitance) is measurable as the free carriers move and reconfigure to screen any microscopic dipoles. The latter case results in essentially a periodic modulation of electron clouds or potentials conceptually similar to the perturbations to electronic transport seen in a charge density wave (CDW), but built on totally different foundations. The electric dipoles in polar metals arise as driven by lattice change, whereas a CDW arises from electronic Fermi surface nesting. FePSe₃ is unique in being tuned from insulator to semiconductor to metal with pressure, whilst at the same time the polar point group develops with the onset of the semiconducting state. This presents an opportunity to understand how the screening develops as a function of carrier concentration, free from disorder effects in cases of chemical doping. Importantly, partially screened dipoles are an intriguing candidate for unconventional superconductivity in the doped polar metallic state of SrTiO₃ [23]. Indeed, the possibility of strong electron-electron correlations mediated by unscreened or under-screened dipoles (static or fluctuating) presents an exciting opportunity for new superconducting materials.

Polar metals, combining two phenomena usually considered mutually exclusive, are increasingly viewed as topologically significant systems too [23]. The physics of these materials is yet in their infancy and holds great promise.

Though polar metals are promising multifunctional materials [9], predicted back in the 60s by Anderson [22] they have only recently started to be demonstrated in very few materials - 2D examples are rarer still. So far there are only a few known examples of polar metals: CePt₃Si [32], UIr [33], LiOsO₃ [34], WTe₂ [35], doped SrTiO₃ [7] and pressurized Hg₃Te₂X₂ [8]. FePSe₃ is an exciting new addition to this nascent field, worthy of further exploration into the polarization and charge distributions at moderate pressure values. We note that NiPS₃ has been recently suggested to be another physical system under high pressure (above ~ 23 GPa) [36].

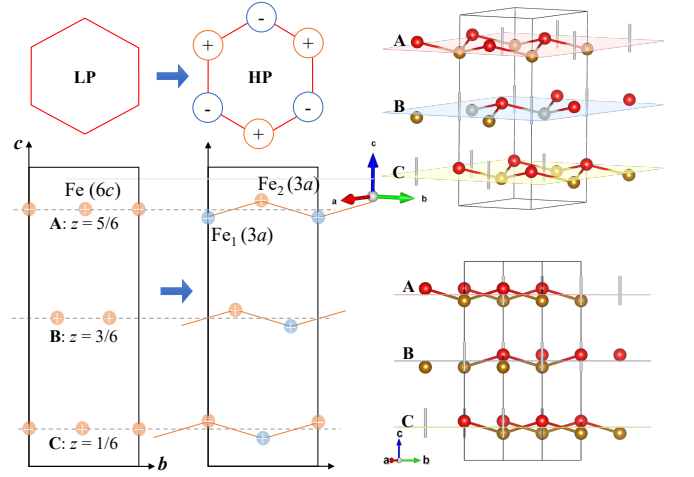


FIG. 7. **Cartoon of the Fe-hexagon buckling in the HP phase.** The amplitude of the relative movement of Fe₁ and Fe₂ in reference to the z -centre of each plane has been increased for better eye guidance. Only Fe ions are shown here.

However, the limited access to room-temperature measurements makes it difficult to go beyond speculating the possibility of such phase.

As the polar metal state can be induced and then tuned with pressure, FePSe₃ gifts the community a new handle on the little-understood physics of these exotic systems. That is, the material can be studied in both polar and non-polar configurations at all temperatures, including intermediate phases showing nontrivial transport properties, thus allowing for direct comparisons. Our unambiguous structural model of this clean and simple system lays a solid foundation for further experiments and density functional theory calculations, and we identify multiple questions raised by the anomalous transport data in this new phase for ongoing focused study in the field.

Exotic Electronic Transport Properties The HP polar phase involves rich phases in the T - P phase diagram, far from being a simple metal. In the high-temperature region of the polar HP phase, FePSe₃ shows activated semiconductor behavior instead of turning metallic immediately, with the resistivity value in the order of $10^{-4} \Omega \cdot m$. The crossover temperature between the high-temperature non-metallic and lower temperature metallic region at varying pressure values forms a fan-shaped region around the IMT. It suggests another interesting intermediate state for further exploration.

Below ~ 40 K, we observe that the resistivity of FePSe₃ demonstrates a T^2 region, which distinguishes it from other compounds of the family. We note that the fitted A -coefficients of the T^2 term are not constant in the HP phase but rather decrease with increasing pressure (Fig. 6). If we were to take the quasi-particle description, where the A -coefficient reflects the effective mass of electrons, such a trend suggests that the electrons are

turning less correlated and electron screening effects getting enhanced with increasing pressure. However, this ‘good metal’ Fermi-liquid description cannot reasonably be applied to a compound with such high resistivity and residual resistivity. Carriers are being heavily scattered, by some alternative (evolving) mechanism that is yet to be determined, but is most likely linked to the polar state, the low dimensionality, or indeed both.

At even lower temperatures, the resistivity deviates from the T^2 behavior and shows an upturn. Similar deviations have been previously reported in other $TMPSe_3$ materials and are still awaiting full explanations [4, 14]. In the case of $FePSe_3$, it should be noted that the upturn is quickly suppressed with increasing pressure, showing the same trend as the A -coefficient shrinking with pressure. It remains an open question whether the phase where this upturn dominates the conductivity (purple region in Fig. 6) is related to the antiferromagnetic critical transition pressure point, or the localized polar modes taking effect when the electrons have the smallest mobility values and are nearly free from thermal effects.

More generally, the strongly correlated electrons are likely to introduce quantum critical fluctuations close to any quantum critical point or endpoint associated with magnetic transitions [33, 37], leading to emergent phenomena including unconventional superconductivity or novel metallic states. The structural transition is strongly first order, so we do not have any clear smoking gun for any such physics, but hints of more subtle changes in order lying behind the phase diagram. Additionally, the broken inversion symmetry may yield a Rashba-type spin-orbit coupling [32] and thus exclude spin-triplet pairing in the HP phase of $FePSe_3$. Further investigations are needed to deepen the understanding of this non-centrosymmetric compound’s novel phases and underlying mechanism.

The transport properties contrast $FePSe_3$ from the rest of the $TMPX_3$ family and merit further investigations into the many questions they pose. Future work based on these discoveries should provide many insights into the fundamentals of van-der-Waals strongly correlated magnetic compounds and into polar metal states.

B. Interpretation of the Crystalline Transition

We note that our results showing the LP-HP transition model with essential interplanar lattice collapse contrasts with the intraplanar lattice collapse interpretation previously proposed by Wang *et al.* [13]. We would like to underline the fact that crystallography in layered materials has been challenging due to issues such as preferred orientation of lamellar powder grains and stacking faults along the interplanar axis. These issues turn more profound in the application of pressure, where the diffraction data collection angles are constrained. Similar cases have been previously reported in this compound family, such as in $NiPS_3$ [20]. We thus found it essential to optimize

the methodology and to draw on as wide a selection of experimental techniques as possible, so as to reach solid, unambiguous structural conclusions. This is reflected in the following aspects:

Using as-grown powder samples - Instead of the usual way to grind single crystals to powder, we used powder samples from growth to minimize preferred orientations. Wang *et al.* used pre-compressed $FePSe_3$ pellets and silicone oil as a PTM for data collection.

Optimized collection geometry - The key difference between our work and that of Wang *et al.* [13] is that we have a sufficiently wider q range to resolve the (003) peak. Tracking the 2θ angle where this peak is observed in powder diffraction data gives us direct and model-independent evidence of the interplanar d -spacing evolution. The most intense (113) peak, however, reflects changes in both inter- and intraplanar distances and thus cannot give solid conclusions.

Effect of pressure-transmitting medium - We additionally examined the presence of a PTM. We utilized helium as it has been documented to be the best available PTM, even in its solid state above ~ 12.1 GPa [38]. We found that, in the absence of a PTM, the onset of the LP-HP structural transition was observed at a higher pressure value (~ 9.6 GPa), and was followed by a co-existence region of both LP and HP until 16.3 GPa. A pure HP phase was not evident before this pressure value, as detailed in the SM [26].

Combined powder & single-crystal diffraction - We found that the powder measurements themselves are not enough to conclude the structural transition model. It was the single-crystal synchrotron diffraction that gave a solid solution. Our single-crystal measurements give model-independent proof through the $(h\bar{h}l)$ plane slices (Fig. 2(b)) that an interplanar collapse occurred during the LP-HP transition. After the transition, we discern a substantial breaking of the Friedel pairs’ structure factors, which can only be attributed to the broken inversion symmetry. It should be noted that such a symmetry change is not discernible by powder diffraction.

We thus confidently affirm our LP-HP structural transition model. Notably, this is an intuitive finding as the planes joined merely by weak van der Waals forces are much easier to compress than it is to compress the hexagonal network of covalent bonds within the layers. We have additionally developed a methodology to carry out the data collection in the most robust way to address the challenge in solving the crystallography in low-dimensional materials.

C. Suppression of Magnetic Ordering

Our powder neutron diffraction studies under pressure affirm that the LP phase experiences a paramagnetic to antiferromagnetic transition. The magnetic configuration of LP remains the same as that reported in the ambient-pressure model [12], with the magnetic prop-

agation vector $(\frac{1}{2}, 0, \pm\frac{1}{2})$ and the moments parallel to the c -axis. Zigzag ferromagnetic chains form within the vdW planes and couple with one another antiferromagnetically. The space group symmetry remains unchanged with varying temperature. Yet, the magnitude of the magnetic moment is suppressed with increased pressure, potentially due to emergent diffuse scattering. The magnetic transition temperature tends to increase slightly with elevated pressure. In the HP phase, we did not observe any presence of magnetic ordering from 300 K to the base temperature 1.6 K.

We find that the magnetic configuration of LP is consistent with the ambient-pressure model [12], while the size of the magnetic moments shrinks with increased pressure. Additionally, the value of T_N tends to increase with elevated pressure. We note that the interplanar distance shrinks more rapidly than the intraplanar atomic separation in the LP phase, which might affect the coordination around the magnetic fragments subtly. Further explorations, for instance, magnetic susceptibility measurements, neutron inelastic scattering under pressure or magnetic exchange interaction simulations, would be needed to gain full understanding into the magnetic phase transitions.

After the structural transition in the HP phase, however, no magnetic peaks or short-range-ordering features are visible in our neutron powder diffraction data. We thus conclude that FePSe₃ does not order magnetically in the HP phase, in interesting contrast to the sulfur compound FePS₃ [29]. The system may experience a reconfiguration of electronic states as FePSe₃ becomes more three-dimensional (3D) and carriers become itinerant after the LP-HP transition. Though we cannot distinguish whether the HP phase is paramagnetic or non-magnetic from our powder neutron diffraction data, we however deem it unlikely for magnetism to stay disordered down to 1.6 K. We thus strongly favor the latter interpretation that FePSe₃ turns from high-spin ($S = 2$) to non-magnetic ($S = 0$) state upon metallization, which also is in line with the X-ray emission spectroscopy observation by Wang *et al.* [13].

IV. CONCLUSIONS

To conclude, we tune the van-der-Waals compound FePSe₃, which is a Mott-insulator with antiferromagnetic ordering at ambient pressure, to a high-pressure polar phase showing semiconducting features at higher temperature, and metallicity at lower temperatures via the application of pressure.

We identify a crystalline transition from the centrosymmetric $R\bar{3}$ to the non-centrosymmetric $R3$ space group at approximately 7 GPa through rigorous synchrotron diffraction studies. This transition is accompanied by a substantial interplanar lattice collapse with no evidence of interplanar sliding. This is the first unambiguous structure transition model in FePSe₃ and

presents a unique transition towards a higher-order state under pressure. Most importantly, the local distortion of Se atoms and the buckling of the Fe honeycombs at the loss of inversion symmetry when the material enters the HP structural phase causes the formation of a polar axis and local dipoles. We also observe the suppression of magnetic ordering and an insulator-to-metal transition at the same pressure, the LP-HP structural transition.

Based on the symmetry analysis and change of conductivity, we report the emergence of a polar metal phase in FePSe₃ at accessible pressures. Polarization and metallicity have been assumed to be mutually exclusive, until the recent few experimental realizations. Our findings thus provide a new platform to explore polar metallicity, in a chemically simple and clean 2D system with strongly correlated electrons. The 2D nature of FePSe₃ also offers the possibility to control the polarization along the c -axis while maintaining the anisotropic metallicity in the ab plane. Further investigations, beyond the scope of this work, will be needed to explore the polarization and charge distributions in this polar metallic phase - an exciting opportunity for discovery in this new field.

Additionally, we outline intermediate phases manifesting exotic metallic behaviors near the structural and magnetic transition pressure. A T^2 resistivity exponent suggests at first glance a simple Fermi-liquid metal (which in itself would be an interesting contrast to the behavior seen in FePS₃ [14]). However, both the resistivity magnitude and the residual resistivity exhibited place this weakly conducting state well outside the range where a Fermi liquid picture could be applied. The scattering mechanisms must be something different; more microscopic and electronics probes will be needed to ascertain exactly what their fundamentals are. Finally, the suppression of the coefficient of the T^2 term at low temperatures and the suppression of a resistivity upturn below the T^2 region, appearing only close to the magnetic critical transition pressure, strongly suggests some underlying quantum ordering (or remnant disorder), necessitating further exploration.

Overall, our multi-probe studies of the crystalline, magnetic structures and transport properties of FePSe₃ contribute significantly to the intrinsic understanding of the $TMPX_3$ compound family. This work presents the first fundamental understanding of the high pressures structures and the discovery of a new emergent 2D polar metallic phase. These findings offer a solid starting point for further physical properties investigations and *ab-initio* studies, providing insights into the fundamentals and tunability of van-der-Waals strongly correlated magnetic compounds and presenting a uniquely tunable low-dimensional model system for the newly-emerging field of polar metallicity. Further study, involving the control of individual layers and stacking patterns, will also be an area of interest for advancing the engineering of low-dimensional magnetism in van-der-Waals materials and applying them into atomically thin-layer device architectures.

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V. METHODS

A. Sample Synthesis and Characterization

FePSe_3 samples were synthesized via two synthesis routes. Both routes had starting materials sealed in quartz tubes that had been cleaned by an acid etching followed by rinsing with demineralized water. All starting materials were of 99.99% purity. Powder samples, used for x-ray and neutron diffraction, were made using a solid-state reaction. This route was used to minimize preferred orientation, known to occur when grinding single crystals due to the vdW nature of the compound. An equivalent of 2 g of starting materials were sealed in quartz tubes, with stoichiometric quantities of Fe and Se. The P content was 10% larger than stoichiometric, with the excess included to account for possible impurity phases. The tubes were sealed containing a pressure of 5 torr of argon. The tubes were then placed in a horizontal two-zone furnace with the starting materials in zone 1. The zones were independently heated to follow the temperature protocol listed in Table II. The protocol prevented vapor transport as zone 1 was always cooler

than zone 2.

The two-zone furnace was also used to synthesize single crystals of FePSe_3 . Equivalent of 1 g of starting material, consisting of stoichiometric quantities of Fe, P and Se, were placed in quartz tubes. A small amount of iodine (0.05 g) was included to facilitate vapor transport. The tubes were then sealed under vacuum and loaded in the furnace. The temperature protocol is also listed in Table II. Zone 2 was colder than zone 1 and the crystals grew in zone 2 via a vapor transport mechanism. The crystals were recovered at the end of the synthesis.

TABLE II. **Temperature protocols during sample synthesis.** Temperature protocols used to synthesize the powder and single crystal samples of FePSe_3 .

	Zone 1 ($^{\circ}\text{C}$)	Zone 2 ($^{\circ}\text{C}$)	Duration (h)
Powders	25 $\rightarrow$ 650	25 $\rightarrow$ 670	6
	650	670	168
	650 $\rightarrow$ 25	670 $\rightarrow$ 300	free cooling
Single Crystals	25 $\rightarrow$ 750	25 $\rightarrow$ 720	6
	750	720	168
	750 $\rightarrow$ 500	720 $\rightarrow$ 450	48
	500 $\rightarrow$ 150	450 $\rightarrow$ 120	6
	150 $\rightarrow$ 25	120 $\rightarrow$ 25	free cooling

B. Synchrotron Diffraction under Pressure

Powder diffraction data were collected at room temperature on the Extreme Conditions beamline (I15) at Diamond Light Source, UK. An incidence X-ray beam with focused spot size $30\text{ }\mu\text{m}$ and wavelength $0.4246\text{ }\text{\AA}$ (equivalently to $E = 29.2\text{ keV}$) was used to collect the diffraction patterns. A MAR345 area detector was used to record the diffraction pattern with an exposure time of 120 seconds for each scan at a distinctive pressure value. The instrument parameters were calibrated with LaB_6 . The data were initially processed using the data analysis software **DAWN – II** [39]. Further, Le Bail and Rietveld refinements were performed using **GSAS – II** [40].

Single-crystal diffraction data were collected at room temperature on the Small Molecule Single Crystal Diffraction beamline (I19-2) [41] at Diamond Light Source, UK. Samples were manually reduced to a thickness of about $10\text{ }\mu\text{m}$ and squares of side length $80\text{ }\mu\text{m}$. The plate-like sample thus had the $c \parallel c^*$ axis, pointing perpendicular to the crystal planes. The sample was laid flat on an anvil. Thus, c^* was parallel to the anvil axis and close to the direction of the incident synchrotron beam. The experimental geometry is comprehensively described in the Supplemental Material (SM) [26]. An incident beam of wavelength $0.4859\text{ }\text{\AA}$ was used and diffraction data were collected while the crystal rotated in a standard configuration with a Dectric Pilatus 300 K detector. The data were then analyzed using **CrysAlis Pro** [42].

Diamond anvil cells (DAC) with rhenium gaskets were used for both powder and single-crystal samples. The culet size of a DAC was approximately $400\text{ }\mu\text{m}$. A hole was drilled through a rhenium gasket of a diameter less than half that of the culets. The gasket hole contained the FePSe_3 sample, and some small ruby crystals used as a pressure gauge. Two DACs were prepared for powder and single crystal diffraction experiments, both being loaded with helium gas serving as the pressure-transmitting medium (PTM).

Pressure was applied using a gas-loading membrane system. The generated pressure inside the sample space was determined by optical measurement of the characteristic R_1 fluorescence peak of ruby [43, 44]. The peak position was measured before and after data collection at each pressure point and the average value was taken as the estimated pressure during the measurement. The uncertainties in the pressure values were estimated to be approximately $\pm 0.1\text{ GPa}$.

Diffraction peaks from the DAC were identified by fitting the known diamond unit cell and were removed for subsequent treatment. Powder-like diffraction rings from the rhenium gasket were also identified and excluded from refinements. Details of pressure determination, sample data reduction and analysis are in the SM [26].

C. Powder Neutron Diffraction under Pressure

Powder neutron diffraction patterns were collected on the D20 instrument [45] at the Institut Laue-Langevin, France [46], in a comparable manner to our previous study on FePS_3 [29]. The neutron wavelength was $2.42\text{ }\text{\AA}$, set via a graphite monochromator. Data were collected using a Paris-Edinburgh type pressure cell press [47] with double-toroidal sintered diamond anvils [48]. The applied load on the cell was dynamically controlled to be constant as a function of temperature.

Powder samples from the same batch used in the synchrotron powder measurements were used for this experiment. The powder was packed into the two halves of a Ti/Zr null matrix spherical gasket [49] and then placed between the anvils. A 4:1 deuterated methanol/ethanol mixture served as a hydrostatic pressure medium. No pressure gauge was employed, as the pressure dependence of the sample's lattice parameters (at room temperature) was taken from the synchrotron diffraction results. The pressure was determined by refining the neutron diffraction patterns and extracting lattice parameters, which were then matched to the synchrotron data. While the synchrotron data were measured at ambient temperature, no great change in the positions of the nuclear Bragg peaks were observed as a function of temperature in the neutron data. Consequently, the uncertainty in sample pressure was estimated to be $\pm 0.3\text{ GPa}$. All pressure changes were made at temperatures where the pressure medium was known to be in its liquid phase [50].

Table III summarizes the scans for the different pressures taken at high- (HT) and low-temperature (LT). At HT, data were collected with 60-minute count times at 300, 300, 245 and 300 K for pressure values of 0, 1.2, 4.1 and 8.3 GPa, respectively. Subsequently, the cell was rapidly cooled to 80 K under constant load by being immersed in liquid nitrogen. At the highest pressure, the cell was subsequently cooled from 80 to 5 K using a closed cycle cryocooler and further cooled to 1.6 K by immersing the cell into liquid ^4He and pumping on it.

TABLE III. **Key scans of magnetic scattering data.** Scans used in the main text for exploring the magnetic properties evolution in powder samples of FePSe_3 .

Pressure (GPa)	HT		LT	
	T (K)	Count Time	T (K)	Count Time
0	300	60 min	—	—
1.2	300	60 min	80	8×15 min
4.1	245	60 min	80	8×15 min
8.3	300	60 min	1.6	8×30 min

The raw diffraction data had a broad, diffuse background due to the pressure environment and liquid pressure medium. An independent background from an empty cell could not be accurately measured because of uncertainties positioning the cell and equating the same volume of pressure medium as well as changes in the background scattering due to temperature and pressure. Consequently, the background was estimated by manually specifying background points away from any diffraction peaks and fitting a smooth Chebychev polynomial through them to subtract from the raw data. The method allows for Bragg peaks to be indexed and their intensities to be determined, but could not discriminate against any diffuse scattering that may come from the sample. All intensities are normalized to a monitor placed in the incident beam. The resulting data were then refined using the Rietveld method as implemented in software **GSAS-II** [40] and **FullProf** [51]. More details on data processing can be found in the SM [26].

D. Resistivity Measurement with Cubic Anvil Cell

The resistivity of a FePSe_3 single crystal sample, from the same batch used for synchrotron diffraction, was measured under various pressures at the Cubic Anvil Cell (CAC) station of Synergetic Extreme Condition User Facility (SECUF), China [52]. The sample was prepared with dimensions 0.23 mm in length (l), 0.22 mm in width (w) and 0.01 mm in height (h). A standard four-probe method was used for resistivity measurements under high pressure. Silver paste (DuPont 4929N) was used to glue gold wires, 20 μm in diameter, to the sample. The resistance (R) of the sample was measured, and the intrinsic resistivity ρ was calculated using the formula, $\rho = R * (w * h) / l$.

The sample was placed inside a Teflon capsule filled with glycerol PTM before being loaded into the CAC. The three-axis compression geometry together with the adoption of a liquid PTM ensured excellent pressure homogeneity. The pressure values in the CAC were estimated from a pressure-loading force calibration curve pre-determined at room temperature. Temperature-dependent measurements from 300 to 1.6 K were performed using a liquid ^4He cryostat with a 9 T superconducting magnet. A helium exchange gas was used to equilibrate the temperature of the CAC. A thermometer was inserted inside the CAC to keep good thermal contact. Details about assembling samples and calibrating the pressure of a CAC can be found in Ref. [53].

At the lowest pressure point (6.0 GPa), where the sample was expected to be an insulator, two methods were used to measure R . From 300 to 215 K, R was measured using a Lake Shore Model 372 in alternating current (AC) mode with a current of 10^{-7} A at a frequency of 13.7 Hz. From 215 K down to the base temperature 1.6 K, data were collected with a Keithley 2400 and 2182 in direct current (DC) mode with an average current of 10^{-7} A. For higher pressure points, where the sample was expected to be a metal, the data were collected using a Keithley 2400 and 2182 in DC mode with an average current of 10^{-5} A for 7.0 and 8.0 GPa, and 10^{-4} A for 10.0, 12.0 and 13.5 GPa from 300 to 1.6 K.

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Author contributions Conceptualization: SD, MJC, CRHS, SSS; Facility support: DD, MRW, TCH, SK, AW, PY, BW, JG; Sample synthesis: CK, JGP; Data col-

lection: SD, MJC, CRSH, HH, DMJ, XZ, ARW; Data analysis and discussions: SD, MJC, CRSH, HH, GIL, DMJ, ARW, XZ, CL; Initial paper writing and editing: SD, MJC, CRSH, ARW, SSS. All authors have read the and contributed to the final paper. We also acknowledge meaningful discussions with E. Artacho, P. L. Alireza, C. Morice and J. van Wezel, and repeated resistivity measurements at CAC by Q. Dong.

Competing interests “There are no competing interests to declare.”

Supplementary Material Figs. S1 to S16; Tables S1 to S5

Supplementary Files

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