

Mott-Anderson crossover without metallization in pressurized $\text{Sr}_3\text{Ir}_2\text{O}_7$

Xia Yin ^{1,*}, Weicheng Guo,^{1,*} Miao Li,² Chunlei Yang,³ Chengyu Li ¹, Dong Wang ¹, Jianbo Zhang ¹, Yongjin Chen ¹, Takeshi Nakagawa ¹, Caoshun Zhang,¹ Chunsheng Xia,¹ Pengfei Shan,¹ Xiaoping Yang ², Sheng Meng,³ Jun Chang ⁴, Ho-kwang Mao ^{1,5} and Yang Ding ^{1,†}

¹*Center for High Pressure Science and Technology Advanced Research (HPSTAR), Beijing 100193, China*

²*High Magnetic Field Laboratory, Hefei Institutes of Physical Science (HFIPS), Chinese Academy of Sciences, Hefei 230031, China*

³*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics (IOP), Chinese Academy of Sciences, Beijing 100190, China*

⁴*School of Physics and Information Technology, Shaanxi Normal University, Xi'an 710119, China*

⁵*Shanghai Key Laboratory of Material Frontiers Research in Extreme Environments (MFree), Shanghai Advanced Research in Physical Sciences (SHARPS), Shanghai 201203, China*



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A Mott insulator can lose its gap yet refuse to metallize. In the spin-orbit-entangled iridate $\text{Sr}_3\text{Ir}_2\text{O}_7$, transport reveals an abrupt collapse of the effective activation gap (Δ_{eff}) from ~ 0.1 – 0.2 eV to $\lesssim 0.02$ eV within 12–17 GPa, yet $R(T)$ remains nonmetallic down to 2.6 K. Above this window, the low-temperature conduction follows three-dimensional Mott variable-range hopping with a strongly reduced hopping scale T_0 . A concurrent magnetoresistance sign reversal further characterizes the postcollapse localized regime, with possible contributions from spin-related scattering. Helium-medium x-ray diffraction shows no resolvable symmetry change, supporting a Mott-Anderson crossover picture where localization preempts metallization. This disorder-inclusive framework may apply broadly to bandwidth-tuned correlated systems where quenched disorder coexists with a pressure-suppressed correlation gap.

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I. INTRODUCTION

Does closing a Mott gap inevitably produce a metal? $\text{Sr}_3\text{Ir}_2\text{O}_7$, the bilayer Ruddlesden-Popper iridate, offers a revealing test case. As a spin-orbit-entangled Mott insulator lying close to the metal-insulator boundary [1–3], where spin-orbit coupling (SOC) and correlations are comparable and both respond sensitively to compression [4–8], it is a prototypical candidate for a pressure-driven Mott collapse: clean-limit calculations predict gap closure and metallization at pressures of order 10–20 GPa [9]. Experiments, however, report strikingly different outcomes in a system for which clean-limit theory predicts a single metallization pressure. Li *et al.* [10] observed a 6-order-of-magnitude resistivity drop near 13 GPa accompanied by a collapse of the thermal activation energy, yet $R(T)$ remained nonmetallic at the lowest measured temperatures—an observation that left the low-temperature transport regime unresolved. At much higher pressure, Ding *et al.* [11] identified a confined metallic state above ~ 60 GPa, leaving unexplained the wide pressure interval over which the system is neither gapped nor metallic. Zocco *et al.* [12] found persistent nonmetallic behavior beyond 100 GPa in a different sample environment. In related iridates, disorder (vacancies, substitution, and local distortions) is known to enhance localization and insulating behavior [13–15]. Near

a bandwidth-tuned Mott boundary, pressure can suppress the correlation-derived gap, while quenched disorder sustains carrier localization—a regime governed by Mott-Anderson competition [16–19], in which the transport activation gap reflects activation toward a disorder-controlled mobility edge [16,19,20]. Because the relevant carriers are spin-orbit-entangled $J_{\text{eff}} = 1/2$ states, disorder and magnetic dephasing act in a pseudospin space and, once long-range magnetic order is strongly suppressed, the associated localization problem is expected to fall into the symplectic universality class [21].

Taken together, these results pose a specific question: what transport regime governs the intermediate-pressure window after the Mott-derived gap is suppressed but before coherent metallization sets in? This question—and its SOC-specific aspects—cannot be resolved by resistivity magnitude alone; it requires (i) quantitative identification of the low-temperature transport mechanism in the postcollapse regime, (ii) an independent structural probe under near-hydrostatic conditions, and (iii) systematic cross-checks on sample and pressure-medium dependence. None of the previous studies [10–12] provided all three simultaneously. Here we combine diamond-anvil-cell (DAC) transport on multiple crystals (with pressure-medium and reversibility checks) with helium-medium single-crystal x-ray diffraction (XRD) under near-hydrostatic conditions. We identify a reproducible crossover window of $P_c \approx 12$ – 17 GPa where the transport activation gap Δ_{eff} collapses without metallization. Above P_c , transport shows localized-regime fingerprints—three-dimensional (3D) Mott variable-range hopping (VRH)

*These authors contributed equally to this work.

†Contact author: yang.ding@hpstar.ac.cn

with a strongly reduced T_0 and a magnetoresistance sign reversal—while helium-medium diffraction reveals no resolvable symmetry change across P_c within our experimental resolution. These results demonstrate that localization can preempt metallization in a pressure-tuned spin-orbit Mott system and provide a disorder-inclusive framework to rationalize the disparate high-pressure reports.

II. PRESSURE-DRIVEN GAP COLLAPSE AND MAGNETIC CORRELATIONS

High-pressure transport was measured in symmetric DACs using a four-probe configuration, following common DAC-based electrical transport experimental standards [10–12,22]. Flux-grown $\text{Sr}_3\text{Ir}_2\text{O}_7$ single crystals were measured in silicone oil (S1–S3) and NaCl (S4) pressure media to assess medium dependence; pressure was determined by ruby fluorescence at room temperature [23]. Because sample geometry is uncertain under high pressure, we report the resistance R and focus on geometry-independent quantities, including the effective transport activation gap Δ_{eff} , the VRH exponent and characteristic hopping scale T_0 , and the magnetoresistance (MR). Further experimental details and analysis procedures are provided in the Supplemental Material [24].

Figure 1 summarizes quasihydrostatic transport in $\text{Sr}_3\text{Ir}_2\text{O}_7$ single crystals up to 31.4 GPa. $R(T)$ remains nonmetallic throughout ($dR/dT < 0$), although its magnitude is markedly suppressed by pressure. In the high-temperature range 220–300 K, $R(T)$ is well described by an activated form [10,12],

$$R(T) = R_0 \exp \left[\frac{\Delta_{\text{eff}}}{2k_B T} \right], \quad (1)$$

from which we extract an effective transport activation scale Δ_{eff} . In the localized regime, this transport-derived scale may track activation toward a disorder-controlled mobility edge rather than a single-particle spectroscopic gap. This extraction is robust to fitting-window choice and is corroborated by the local activation energy $\Delta_{\text{loc}}(T) = 2k_B d(\ln R)/d(1/T)$ (see Supplemental Material [24]). As shown in Fig. 1(b), Δ_{eff} decreases gradually at low pressures and then collapses abruptly within 12 to 17 GPa, thereby defining P_c for all three crystals. After the collapse, Δ_{eff} saturates at $\lesssim 0.02$ eV, with modest sample-to-sample variations, and remains only weakly pressure dependent up to 31.4 GPa, while $R(T)$ remains nonmetallic down to 2.6 K.

Over the measured range, $R(T)$ shows no low-temperature saturation or sign change in dR/dT that would indicate the onset of coherent metallic transport. Notably, Δ_{eff} largely recovers upon decompression below P_c , supporting that the collapse is an intrinsic, reversible response to bandwidth tuning (see Supplemental Material [24]).

To follow the pressure evolution of antiferromagnetic (AFM) correlations, we track a pronounced kink/inflection in Arrhenius plots and define a transport anomaly scale T_N^{tr} [Fig. 1(c); see Supplemental Material [24] for details]. At ambient pressure, $T_N^{\text{tr}} \approx 280$ K matches the established Néel temperature of $\text{Sr}_3\text{Ir}_2\text{O}_7$. With increasing pressure, T_N^{tr} decreases and is sharply suppressed across the same P_c window, consistent with the reported softening of the two-magnon

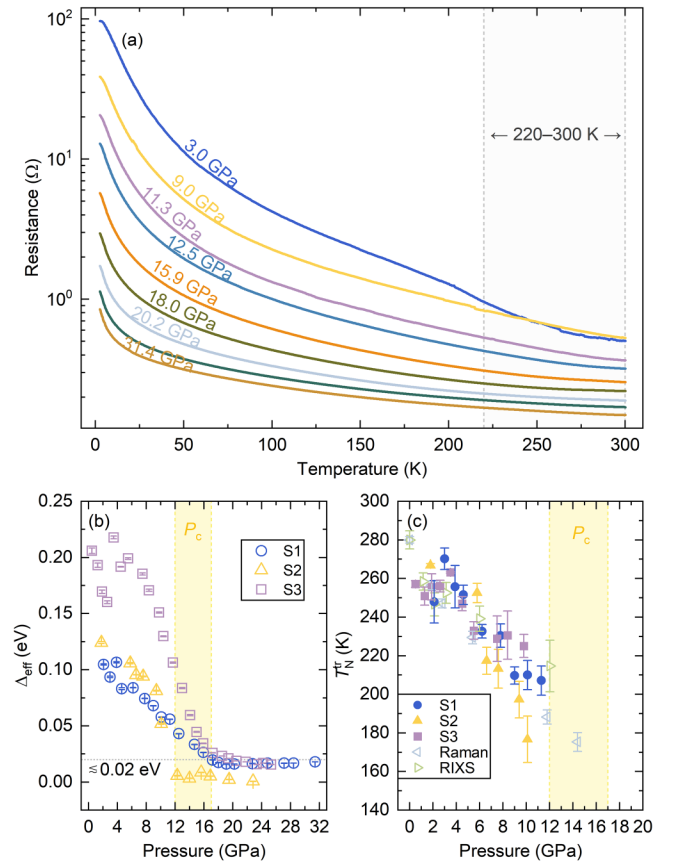


FIG. 1. Pressure-driven collapse of the transport activation gap in $\text{Sr}_3\text{Ir}_2\text{O}_7$. (a) Temperature-dependent resistance $R(T)$ of sample S1 at representative pressures up to 31.4 GPa (labels), plotted on a logarithmic scale. Pressure markedly suppresses $R(T)$ yet the transport remains nonmetallic ($dR/dT < 0$) down to 2.6 K. (b) Effective activation gap Δ_{eff} extracted from Arrhenius fits (220–300 K) as a function of pressure for three samples (S1, S2, S3); fitting uncertainties are smaller than symbol sizes. The shaded region marks the collapse window (12–17 GPa), defining a crossover pressure scale, P_c . (c) Transport-defined Néel temperature T_N^{tr} extracted from the kink/inflection in Arrhenius plots; literature Raman/RIXS two-magnon energy scales [25,26] are plotted as E_{2M}/k_B (i.e., in temperature-equivalent units) for comparison. See Supplemental Material [24] for robustness checks (Δ_{loc} and fitting-window sensitivity).

energy scale measured by Raman and resonant inelastic x-ray scattering (RIXS) [25,26], plotted as E_{2M}/k_B (i.e., in temperature-equivalent units) for comparison.

III. LOCALIZED TRANSPORT IN THE POSTCOLLAPSE REGIME

We now turn to low-temperature transport in the postcollapse regime, where activation across a clean gap no longer dominates. Conduction is well described by VRH between localized states [20,27–29],

$$R(T) = R_0 \exp \left[\left(\frac{T_0}{T} \right)^{1/(d+1)} \right], \quad (2)$$

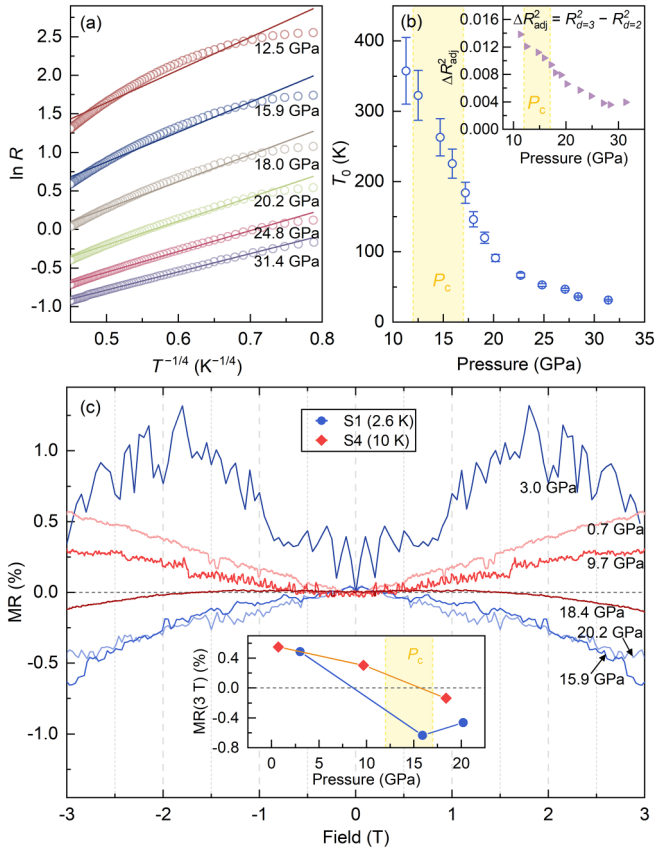


FIG. 2. Quantitative low-temperature signatures of localization in pressurized $\text{Sr}_3\text{Ir}_2\text{O}_7$. (a) $\ln R$ of sample S1 plotted versus $T^{-1/4}$ (2.6–25 K) at representative pressures approaching and above P_c ; solid lines are linear fits. (b) Hopping scale T_0 extracted from the slopes in panel (a); error bars represent the standard error from linear regression. Inset: Difference in fit quality $\Delta R^2_{\text{adj}} = R^2_{d=3} - R^2_{d=2}$ for sample S1, derived from linear regressions of $\ln R$ vs $T^{-1/4}$ ($d = 3$) and $T^{-1/3}$ ($d = 2$). The positive difference ($\Delta R^2_{\text{adj}} > 0$) favors $T^{-1/4}$ scaling over $T^{-1/3}$ within the tested low-temperature window. (c) Field-dependent magnetoresistance $MR(B)$ computed from the symmetrized longitudinal resistance $R_L(B) = [R(+B) + R(-B)]/2$ for sample S1 at 2.6 K and sample S4 at 10 K, showing a pressure-driven sign reversal. Inset: MR at 3 T as a function of pressure, highlighting the sign crossover across the crossover window. See Supplemental Material [24] for exponent discrimination and even/odd-in-field decomposition. Pressures quoted for low-temperature VRH and MR measurements are nominal room-temperature ruby pressures; the actual low-temperature pressure may be higher by several GPa due to thermal pressure shifts upon cooling.

where d is the effective dimensionality and T_0 is the characteristic hopping temperature. Upon approaching and above P_c , sample S1 exhibits an extended linear dependence of $\ln R$ on $T^{-1/4}$ over 2.6–25 K, consistent with three-dimensional Mott VRH ($d = 3$) [29]. Although $\text{Sr}_3\text{Ir}_2\text{O}_7$ is layered, the observed $1/4$ exponent implies effectively three-dimensional hopping under compression. The extracted hopping scale T_0 decreases from about 3.6×10^2 K near 12.5 GPa to about 3.1×10^1 K at 31.4 GPa for sample S1 [Fig. 2(b)]. Within the same temperature windows, fits to $\ln R$ versus $T^{-1/4}$ yield the smallest normalized residuals (highest adjusted R^2) compared

with $T^{-1/3}$ and $T^{-1/2}$ (see Supplemental Material [24] and Refs. [27,30] therein for a quantitative comparison); no robust $T^{-1/2}$ scaling is found, indicating that an Efros-Shklovskii regime is not dominant in the measured range. Similar VRH scaling is observed in samples S2 and S3 over their accessible low-temperature ranges. Notably, both in-plane (S1, S3) and out-of-plane (S2) configurations favor the $p = 1/4$ scaling within fixed low-temperature windows (see Supplemental Material [24]), reinforcing effectively three-dimensional hopping under compression.

Magnetoresistance provides an additional probe of the localized regime. We define

$$MR(B) = \frac{[R_L(B) - R_L(0)]}{R_L(0)}, \quad (3)$$

where $R_L(B) = [R(+B) + R(-B)]/2$ is the symmetrized longitudinal component. Hall-voltage admixture is typically $\lesssim 0.1\%$ (at most $\sim 0.5\%$) and removed by symmetrization (see Supplemental Material [24]). Figure 2(c) shows a crossover from positive MR at low pressure to negative MR in the post-collapse regime. For sample S1 at 2.6 K, MR (3 T) changes from $+0.49\%$ (3.0 GPa) to -0.63% (15.9 GPa) and -0.46% (20.2 GPa). A second crystal (S4, 10 K) shows a similar evolution, with MR (3 T) becoming negative near 18.4 GPa. Notably, sample S4 was measured using NaCl as the pressure medium, demonstrating that the negative- MR signature is not a medium-specific artifact. Below P_c , the positive MR is consistent with the conventional wave-function-shrinkage contribution expected for activated or hopping-like transport in an insulating regime [31]. Above P_c , where the low-temperature transport follows a VRH-like form, negative MR can arise from quantum interference between different hopping paths and from intrinsic spin-dependent hopping processes [32,33]. In addition, because T_N^{tr} is suppressed in the same pressure window, field suppression of residual spin fluctuations or spin-disorder scattering provides an additional negative- MR channel specific to the pressure-induced magnetic instability in $\text{Sr}_3\text{Ir}_2\text{O}_7$. This possibility is particularly relevant given the strong spin-orbit-coupled magnetic interactions in this compound, including Dzyaloshinskii-Moriya and higher-order exchange terms [34]. The strong reduction of T_0 above P_c implies increased overlap between localized $J_{\text{eff}} = 1/2$ states, providing a natural setting in which interference, spin-dependent hopping, and residual spin-disorder effects can become visible in magnetotransport. We therefore interpret the MR sign reversal as a complementary magnetotransport signature of the postcollapse localized regime, rather than as an independent diagnostic of the Mott-Anderson crossover.

IV. STRUCTURAL CONSTRAINTS ON THE ELECTRONIC CROSSOVER

To determine whether the transport crossover has a structural origin, we performed single-crystal x-ray diffraction under a highly hydrostatic helium pressure medium. $\text{Sr}_3\text{Ir}_2\text{O}_7$ retains the ambient $C2/c$ monoclinic symmetry up to 19.9 GPa [35], and the lattice parameters, unit-cell volume, and Ir-O-Ir bond angles evolve smoothly with pressure, with no discernible anomaly across $P_c \approx 12$ –17 GPa (Fig. 3).

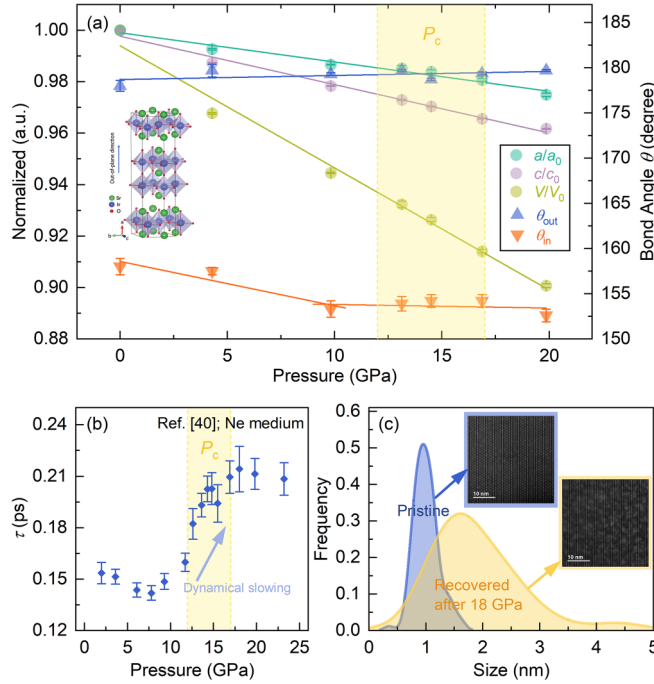


FIG. 3. Independent constraints on the pressure-driven crossover in $\text{Sr}_3\text{Ir}_2\text{O}_7$. (a) Helium-medium single-crystal x-ray diffraction shows smooth evolution of the unit-cell metrics and Ir-O-Ir bond angles with no discernible anomaly across $P_c \approx 12\text{--}17$ GPa (shaded). (b) Contact-free ultrafast pump-probe measurements under neon pressure [40] reveal a pronounced dynamical slowing (enhanced relaxation time τ) in the same P_c window. Data adapted from Ref. [40]. (c) *Ex situ* TEM on a crystal recovered after 18 GPa shows enhanced nanoscale inhomogeneity; distributions of characteristic length scales for pristine and pressure-exposed samples are compared (Insets: High-angle annular dark-field images; scale bars, 10 nm).

Our helium-medium single-crystal XRD constrains the room-temperature average structure and therefore does not rule out subtle low-temperature, local, or oxygen-sublattice distortions. Such caution is warranted because low-temperature pressure studies in the related spin-orbit-coupled iridate $\beta\text{-Li}_2\text{IrO}_3$ show that structural instabilities can be missed or mischaracterized by room-temperature measurements [36]. For $\text{Sr}_3\text{Ir}_2\text{O}_7$ itself, high-pressure structural studies have probed this pressure range, including low-temperature powder diffraction and high-pressure Raman/structural measurements. Donnerer *et al.* reported a first-order structural transition with volume collapse only at much higher pressure, well above the P_c window [9]. A subtle second-order structural anomaly near 14 GPa was observed in low-temperature 25 K powder XRD [37], and a combined Raman and structural study reported a magnetic transition coincident with a structural transition near 14.4 GPa, associated with IrO_6 octahedral tilting, but without a concurrent insulator-metal transition [26]. Because the reported ~ 14 GPa anomaly mainly involves IrO_6 octahedral tilting and subtle oxygen-sublattice distortions, our room-temperature helium-medium single-crystal dataset may have limited sensitivity to it. Accordingly, no discernible feature is resolved in our room-temperature data. This does not contradict previous reports, as our data

exclude discernible major structural reconstructions across P_c . Taken together, these results indicate that subtle lattice and magnetoelastic responses can accompany the suppression of magnetic order near P_c , but they do not support a major structural reconstruction as the primary driver of the transport crossover.

This structural continuity under helium, together with cross-crystal/media reproducibility and decompression reversibility (see Supplemental Material [24] and Refs. [38,39] therein), argues against a resolvable major structural reconstruction, irreversible damage, and medium-specific artifacts as the primary origin of the transport anomalies. Complementary support comes from a contact-free ultrafast pump-probe study under neon pressure, which reports a pronounced dynamical slowing (enhanced carrier relaxation time τ) in the same 12–17 GPa window [40] [Fig. 3(b)], consistent with an intrinsic electronic crossover. The central result is therefore electronic: Δ_{eff} collapses within P_c without metallization, and the postcollapse regime crosses into a disorder-dominated localized regime, characterized by 3D Mott VRH and a magnetoresistance sign reversal (Fig. 2).

V. MOTT-ANDERSON INTERPRETATION AND DISORDER EFFECTS

Within a Mott-Anderson framework [16–19], increasing bandwidth suppresses the correlation-derived activation scale, while intrinsic disorder can keep a mobility edge above the Fermi level and preempt coherent metallic transport. For spin-orbit-entangled $J_{\text{eff}} = 1/2$ carriers, disorder and magnetic dephasing act in a pseudospin space and, once magnetic order is strongly weakened, the corresponding localization problem is expected to fall into the symplectic class [21]. The small residual Δ_{eff} above P_c , together with 3D Mott VRH and the MR sign reversal, is thus compatible with activation toward a disorder-controlled mobility edge and with interference corrections in hopping transport whose sign and magnitude are sensitive to the spin-orbit texture of the localized states [31–33].

In a high-pressure experiment, the effective disorder governing the Anderson component of the crossover can have both intrinsic and extrinsic origins. Intrinsic quenched disorder, including local stoichiometric variations and structural distortions, is already present in the as-grown material, as evidenced by the nanoscale inhomogeneity resolved by transmission electron microscopy (TEM) in the pristine crystal. In addition, a nonhydrostatic pressure environment transmits deviatoric stress and imposes spatial pressure gradients. Because the $J_{\text{eff}} = 1/2$ bandwidth and correlation-derived activation scale of $\text{Sr}_3\text{Ir}_2\text{O}_7$ are strongly pressure-tunable, such gradients can generate spatial variations in local strain, Ir-O-Ir geometry, bandwidth, activation scale, and hopping barriers. Nonhydrostatic strain therefore does not merely mimic disorder; it constitutes a genuine extrinsic contribution to the effective random landscape that, near the collapse of the correlation-derived activation scale, broadens the crossover and promotes localized hopping. This perspective also explains why experiments performed with pressure media of different hydrostaticity can report different apparent critical pressures and residual nonmetallicity [10–12]. They sample

different effective disorder strengths. Crucially, the localized regime itself is not an artifact of nonhydrostaticity. The transport crossover is established from multiple crystals in the S1–S3 datasets, sample S4 provides an independent NaCl-medium magnetoresistance pressure-medium check, intrinsic nanoscale disorder is already present in the pristine crystal, and near-hydrostatic helium XRD and neon pump-probe measurements corroborate the same electronic crossover. Nonhydrostatic strain therefore modulates the strength of the effective disorder rather than creating the localized state.

Clean-limit generalized gradient approximation+SOC+ U calculations indicate gap closure between 12 and 15 GPa and a concurrent destabilization of the low-pressure G -type AFM insulating state (see Supplemental Material [24] and Refs. [6,35,41–47] therein for details). In the relaxed high-pressure structures, the magnetic solution carries only a very small ordered moment, corresponding to a magnetically near-disordered metallic state. Thus, the robust theoretical result is not a specific magnetic-sequence assignment, but the loss of a robust AFM insulating state and the emergence of a near-magnetically disordered metallic clean-limit solution at the bandwidth scale of P_c . This bandwidth-driven magnetic instability coincides with the experimental suppression of T_N^U , whereas the experimentally observed nonmetallic transport beyond P_c indicates that disorder-controlled localization preempts coherent metallization.

From the pressure dependence of T_0 for sample S1, we estimate an order-of-magnitude localization length $\xi \approx 4\text{--}10$ nm using a representative density of states $N(E_F) \sim 10^{22} \text{ eV}^{-1} \text{ cm}^{-3}$ (see Supplemental Material [24] and Refs. [9,31,48] therein for details). Allowing the density of states to vary over an order of magnitude gives a broader range of several to ~ 20 nm. This length scale exceeds the compressed interlayer spacing, naturally accounting for the observed three-dimensional hopping despite the layered crystal structure. Because the density of states near the pressure-induced activation-scale collapse is not independently known, this estimate should be regarded as order-of-magnitude only. Complementary *ex situ* TEM on pressure-recovered crystals provides real-space evidence for nanoscale structural and electronic inhomogeneity that can influence carrier localization in the postcollapse regime. Because the VRH-derived localization length is only an order-of-magnitude transport estimate, the TEM length scale should not be interpreted as a one-to-one measure of the localization length.

These results also clarify the relationship to earlier high-pressure reports on $\text{Sr}_3\text{Ir}_2\text{O}_7$. The dramatic resistivity suppression near 13 GPa reported in Ref. [10] coincides with our crossover window; our work now identifies the postcollapse regime as localized, following 3D Mott VRH with a residual activation scale consistent with a disorder-controlled mobility edge. The confined metallic state reported at much higher pressure in Ref. [11] is compatible with a scenario in which disorder delays coherent metallization over a broad pressure interval, with additional high-pressure lattice effects potentially assisting the eventual onset of metallic transport. Conversely, persistent nonmetallicity up to megabar pressures [12] can be naturally accommodated

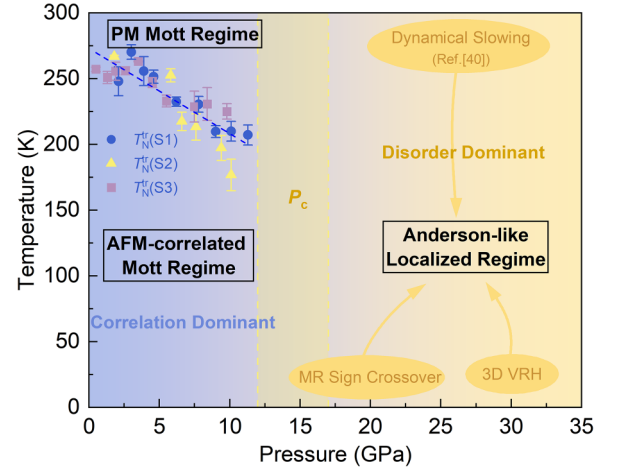


FIG. 4. Schematic pressure-temperature phase diagram of $\text{Sr}_3\text{Ir}_2\text{O}_7$ illustrating the Mott-Anderson crossover. The shaded region marks the crossover window $P_c \approx 12\text{--}17$ GPa, separating the correlated Mott regime from the disorder-dominated Anderson-like localized regime. Symbols denote the transport-defined Néel temperature from Fig. 1(c). The ellipses mark key signatures: 3D Mott VRH and MR sign crossover from Fig. 2, and the dynamical slowing from Ref. [40]. Dashed lines are guides to the eye.

if effective disorder and/or hydrostaticity differ, keeping the mobility edge above E_F . Variations in intrinsic disorder and pressure environment thus reconcile the spread of apparent “critical pressures” within a single Mott-Anderson picture. The resulting correlation-to-disorder crossover is summarized in Fig. 4.

VI. SUMMARY

In summary, the effective transport activation gap (Δ_{eff}) in $\text{Sr}_3\text{Ir}_2\text{O}_7$ collapses within $P_c \approx 12\text{--}17$ GPa without metallization down to 2.6 K, and the postcollapse regime is instead governed by disorder-dominated localization. Helium-medium single-crystal diffraction reveals no resolvable symmetry change across P_c within our experimental resolution. These results demonstrate that, near a pressure-tuned spin-orbit Mott boundary, disorder-driven localization can preempt metallization even after the correlation-derived transport activation scale collapses—providing a microscopic transport diagnosis for the resistivity collapse reported in Ref. [10] and establishing a localization-dominated intermediate regime that naturally precedes the confined metallic state observed at higher pressures [11]. In spin-orbit-entangled $J_{\text{eff}} = 1/2$ systems, disorder acts in a pseudospin space and can realize localization and hopping-MR phenomenology distinct from conventional weak-SOC Mott-Anderson materials [21,32,33], motivating a disorder- and symmetry-aware view of bandwidth-driven “Mott collapse” in correlated oxides.

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DATA AVAILABILITY

The data that support the findings of this article are publicly available [50].

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