

Nature of the high-pressure insulating state in Sr_2IrO_4 : Mott pictureGuoren Zhang^{1,2,3}, Hanif Hadipour⁴, and Eva Pavarini³¹*School of Physics and Technology, Nantong University, Nantong 226019, People's Republic of China*²*Key Laboratory of Materials Physics, Institute of Solid State Physics, HFIPS,**Chinese Academy of Sciences, Hefei 230031, People's Republic of China*³*Peter Grünberg Institute, Forschungszentrum Jülich, D-52425 Jülich, Germany*⁴*Department of Physics, University of Guilan, Rasht 41335-1914, Iran*

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At ambient pressure, the t_{2g}^5 layered perovskite Sr_2IrO_4 is a correlated small-gap insulator. In the Mott picture, applying uniform pressure should therefore quickly close the gap; experimentally, however, the insulating state persists even under extreme pressures, suggesting that a mechanism other than Mott is at work. Yet, given the complexity of the system, it is unclear to what extent the Mott picture can be really excluded. Here, we thus reexamine the problem. We show that, surprisingly, the pressure-induced enhancement of the screened Coulomb interaction—combined with lattice distortions and spin-orbit driven $j_{\text{eff}} = 1/2$ orbital ordering—can hold the system close to the metal-insulator transition up to very high pressure.

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Introduction. The tetragonal layered t_{2g}^5 perovskite Sr_2IrO_4 [1] is a strongly correlated small-gap insulator [2]. The nature of the insulating state remains a matter of hot debate [3–8]. Nonetheless, there is a general agreement that the formation of the gap is assisted by the large spin-orbit (SO) coupling, enhanced by Coulomb repulsion. The picture is the following. Due to the layered structure, the t_{2g} electrons form, at low energy, a two-dimensional xy band and quasi-one-dimensional xz/yz bands. For Ir $5d$ electrons, the t_{2g} bandwidth W is large with respect to the local screened Coulomb repulsion U , and one would expect a weakly correlated metallic behavior. Because of the large SO interaction, however, t_{2g} states regroup into Kramers states: a $j_{\text{eff}} = 1/2$ doublet and a lower-energy $j_{\text{eff}} = 3/2$ quartet. Switching on the Coulomb repulsion, the $j_{\text{eff}} = 3/2$ bands become full and inert; the resulting electronic configuration is $(j_{\text{eff}} = \frac{1}{2})^1(j_{\text{eff}} = \frac{3}{2})^4$. Because of its narrow bandwidth, a gap opens in the half-filled $j_{\text{eff}} = 1/2$ band, even if U is modest—perhaps with a boost from magnetic superexchange. This yields the so-called $j_{\text{eff}} = 1/2$ single-band picture [9–13], which in turn suggests that Sr_2IrO_4 could be electronically similar to cuprates. Recently, however, we have shown that a consistent picture of the electronic properties can only be obtained including explicitly all t_{2g} bands in the Hamiltonian [14,15]. Thus it appears more appropriate to describe the $(j_{\text{eff}} = \frac{1}{2})^1(j_{\text{eff}} = \frac{3}{2})^4$ ground state as $(j_{\text{eff}} = \frac{1}{2})$ -orbital ordering, an inherently multiorbital phenomenon. The presence of the $j_{\text{eff}} = 3/2$ orbitals close to the Fermi level is confirmed in experiments [16]. This conclusion changes the perspective, but also hints at an intrinsic fragility

of the insulating state, making the persistence of the latter under extreme uniform pressure ($P \sim 195$ GPa) [17] even more surprising.

In strongly correlated systems pressure typically induces an insulator-to-metal transition (IMT), since it reduces the U/W ratio. If the gap is already small at ambient pressure, correspondingly, the transition is expected at moderate P . This is even more so in multiorbital materials, since orbital degeneracy hampers the formation of an insulating state. In Sr_2IrO_4 , however, a metalliclike behavior is not observed up to 195 GPa. In fact, the resistance initially decreases with increasing P , abruptly rising again around 40 GPa, when the system becomes orthorhombic [2,17–20]. Within the Mott scenario, a possible explanation of the 40 GPa upturn is the reduction of W due to octahedral tilting in the orthorhombic phase [17,21,22]; however, this requires that a correlated insulating phase persists all the way up to 40 GPa, where the gap is still ~ 20 –30 meV [18,19]. Thus, alternative pictures have been suggested, e.g., charge-density wave instability, structural disorder, and/or Anderson localization [23]. In the presence of nontrivial multiorbital effects, however, first it has to be shown to what extent the Mott picture can be really ruled out.

In this Letter we thus reexamine the Mott picture. We show that, against expectations, the system remains close to the metal-insulator transition up to very high pressure (Fig. 1). The reason is the enhancement of the Coulomb interaction under pressure, and its subtle interplay with multiband effects. To show this, we focus on the most problematic conditions for a Mott state: (i) the tetragonal structure below 40 GPa and (ii) the paramagnetic phase; in fact, while it has been shown that antiferromagnetism helps the insulating phase [25], long-range magnetic order disappears experimentally around 20 GPa [18,20]. We obtain the following results. First, $(j_{\text{eff}} = \frac{1}{2})$ -orbital ordering persists up to high pressure. The shape of the $j_{\text{eff}} = 1/2$ state, however, is modified in a way

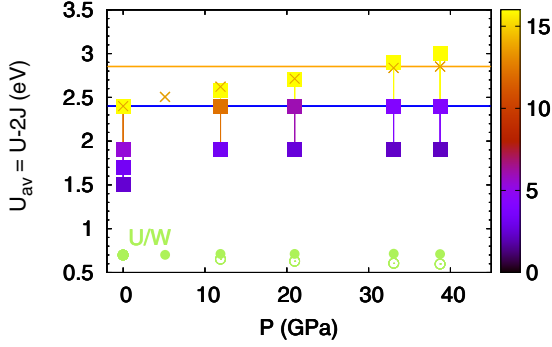


FIG. 1. Effective $j_{\text{eff}} = 1/2$ masses, $m_{j_{\text{eff}}=1/2}^*$, as a function of the pressure P and of the average Coulomb repulsion, U_{av} . Blue horizontal line: $U_{\text{av}} = U_0$, yielding the experimental gap at $P = 0$. Crosses: $U'_0(P) = U_0 + \Delta U(P)$, where $\Delta U(P)$ is the increase due to pressure. Orange horizontal line: U'_0 at $P = 40$ GPa. Open circles: U_0/W . Solid circles: U'_0/W . In the insulating phase $m_{j_{\text{eff}}=1/2}^*$ diverges [24].

that reduces the correlation strength [14], and shortens the distance to the topological point [15]. For what concerns the t_{2g} bandwidth W , pressure leads to two competing effects, volume reduction and an increase in IrO_6 octahedral rotation; while volume reduction dominates, the increase in rotation angle slows down its effect on W [26]. Combining all these elements together leads to a metallic state at low pressure, with moderate masses around 40 GPa (Fig. 1). We find that, however, surprisingly, the pressure-induced Coulomb enhancement is sufficient to keep the ratio U/W almost constant. This, in turn, can hold the system very close to the IMT up to 40 GPa.

Model and methods. We adopt the density-functional-theory+dynamical mean-field theory (DFT+DMFT) approach [27]. We calculate the electronic structure in the local-density approximation via the full-potential linearized augmented plane-wave method implemented in the WIEN2K code [28]. We construct Ir-centered localized t_{2g} Wannier functions [29] spanning the t_{2g} bands and build the associated t_{2g} Hubbard Hamiltonian,

$$\hat{H} = - \sum_{ii'} \sum_{mm'} \sum_{\sigma\sigma'} t_{m\sigma, m'\sigma'}^{i,i'} \hat{c}_{im\sigma}^\dagger \hat{c}_{i'm'\sigma'} + \frac{1}{2} \sum_i \sum_{mm'pp'} \sum_{\sigma\sigma'} U_{mm'pp'} \hat{c}_{im\sigma}^\dagger \hat{c}_{im'\sigma'}^\dagger \hat{c}_{ip'\sigma'} \hat{c}_{ip\sigma}, \quad (1)$$

where $\hat{c}_{im\sigma}^\dagger$ ($\hat{c}_{im\sigma}$) creates (annihilates) an electron at lattice site i with spin $\sigma \in \{\uparrow, \downarrow\}$ and orbital $m \in \{xy, yz, xz\}$. The terms $t_{m\sigma, m'\sigma'}^{i,i'}$ with $i \neq i'$ give the hopping integrals. The matrix $\varepsilon_{m\sigma, m'\sigma'} = -t_{m\sigma, m'\sigma'}^{i,i}$ gives the crystal-field (CF) and on-site SO matrix. Ordering the basis as $|xy\rangle_\uparrow, |yz\rangle_\uparrow, |xz\rangle_\uparrow, |xy\rangle_\downarrow, |yz\rangle_\downarrow, |xz\rangle_\downarrow$, this matrix can be expressed as

$$\varepsilon = \begin{pmatrix} \varepsilon_{xy} & 0 & 0 & 0 & \frac{\lambda_y}{2} & -\frac{i\lambda_x}{2} \\ 0 & \varepsilon_{yz} & \frac{i\lambda_z}{2} & -\frac{\lambda_y}{2} & 0 & 0 \\ 0 & -\frac{i\lambda_z}{2} & \varepsilon_{xz} & \frac{i\lambda_x}{2} & 0 & 0 \\ 0 & -\frac{\lambda_y}{2} & -\frac{i\lambda_x}{2} & \varepsilon_{xy} & 0 & 0 \\ \frac{\lambda_y}{2} & 0 & 0 & 0 & \varepsilon_{yz} & -\frac{i\lambda_z}{2} \\ \frac{i\lambda_x}{2} & 0 & 0 & 0 & \frac{i\lambda_z}{2} & \varepsilon_{xz} \end{pmatrix}. \quad (2)$$

Here, $\varepsilon_{xy}, \varepsilon_{yz}, \varepsilon_{xz}$ are the on-site energies, and the CF splitting is $\varepsilon_{\text{CF}} = (\varepsilon_{yz} + \varepsilon_{xz})/2 - \varepsilon_{xy}$; λ_x, λ_y , and λ_z are SO couplings, with average $\lambda = (\lambda_x + \lambda_y + \lambda_z)/3$. At ambient pressure, we obtain $\lambda_{x/y} \sim 346$ meV, $\lambda_z \sim 354$ meV, and $\varepsilon_{\text{CF}} = 213$ meV. The parameters $U_{mm'pp'}$ are elements of the screened Coulomb interaction tensor. Here, the essential terms are [30,31] the direct and exchange Coulomb interaction, $U_{mm'mm'} = U_{m,m'} = U - 2J(1 - \delta_{m,m'})$ and $U_{mm'm'm} = J$, as well as the pair-hopping and the spin-flip terms, $U_{mmmm'm'} = J$ and $U_{mm'm'm} = J$. The average Coulomb interaction is $U_{\text{av}} = U - 2J$. We solve (1) with dynamical mean-field theory using the interaction-expansion continuous-time [32] quantum Monte Carlo method, in the implementation of Refs. [33–37]. At ambient pressure, unless differently specified, we adopt the value $U_{\text{av}} = 2.4$ eV, established to well describe [14,16,38] the small experimental gap [9,39] of 0.1–0.6 eV.

For calculating the effect of pressure on the structure we perform internal coordinate optimization. Bond distances and octahedral rotation angles were obtained with the same procedure (using however pseudopotentials) in Ref. [23], where the measurement of lattice constants was reported; our coordinates are in line with these results [40]. We determine the change in U_{av} due to pressure via the constrained random-phase approximation (cRPA) [41,42] as implemented in the SPEX code [43].

Finally, we define the orbital occupations per spin as n_{xy}^σ and $n_{xz/yz}^\sigma$, and the orbital polarization as $p = n_{xy}^\sigma - (n_{xz}^\sigma + n_{yz}^\sigma)/2$. The value of these parameters can locate the system on the map of CF effects obtained in Ref. [15].

Bandwidth and hopping integrals. First, we analyze the effects of pressure on the bandwidth W . Here, two effects compete: (i) The volume reduction enhances W , and (ii) the increase in the IrO_6 octahedral rotation angle reduces it. We adopt as the control case Ba_2IrO_4 , a material with a larger ambient pressure volume and no octahedral rotation, and which, accordingly, becomes metallic already at 13.8 GPa [44]. We find that, in Sr_2IrO_4 , while the volume reduction dominates, octahedral rotation sizably slows down the increase of W , reducing the speed with which a metallic state emerges under pressure. The increase of W is mostly coming from the first-neighbor hopping integrals [26]; second-neighbor hopping integrals also somewhat increase but remain small in comparison.

Stability of $(j_{\text{eff}} = 1/2)$ -orbital ordering. Our results show that $(j_{\text{eff}} = 1/2)$ -orbital ordering persists up to 40 GPa. This is an emergent phase in a multiband system, however, and the half-filled $j_{\text{eff}} = 1/2$ orbital is not rigid: With increasing P the system moves towards the topological point [15], located at $p = p_0 \sim 0$ (Fig. 2). This is reflected in changes in the form of the effective $j_{\text{eff}} = 1/2$ ground multiplet. The latter is defined as

$$\left| \frac{1}{2}, \sigma \right\rangle = \alpha_2 i |xy, \sigma\rangle - \alpha_1 \frac{|xz, -\sigma\rangle - 2i\sigma |yz, -\sigma\rangle}{\sqrt{2}}, \quad (3)$$

with $\alpha_1^2 + \alpha_2^2 = 1$; here, $|m, \sigma\rangle$ is a t_{2g}^5 multiplet with a hole in the m, σ state [26]. The coefficients (α_1, α_2) can be expressed in terms of the orbital polarization p , i.e., $\alpha_1^2 = 2n_{xy}^\sigma - 1 = 2(1 + 2p)/3$. At ambient pressure ($P = 0$) we obtain $\alpha_1 \sim 0.901$, close to the atomic limit [14];

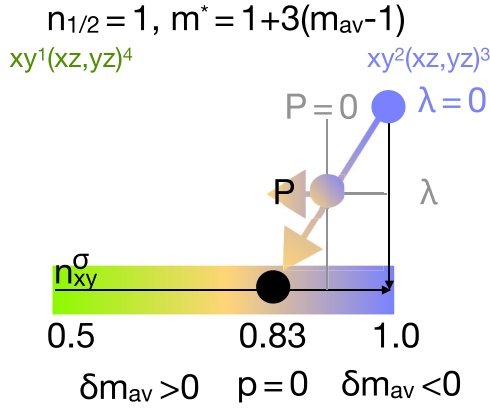


FIG. 2. Evolution of the ground state with increasing n_{xy} , P , and λ , yielding $(j_{\text{eff}} = \frac{1}{2})$ -orbital ordering at ambient pressure for realistic λ . Here, $m_{\text{av}} = (m_{xy}^* + m_{xz}^* + m_{yz}^*)/3$ is the average mass, and $\delta m_{\text{av}} = m_{xy}^* - m_{xz/yz}^*$.

correspondingly $n_{xy} = 0.89$ and $p > 0$. Increasing pressure we find that $p \rightarrow 0$, approached from the right-hand side, where the hierarchy of the effective masses [15] is such that $m_{xy}^* < m_{xz/yz}^*$ (Fig. 2). This is due to the fact that the SO couplings are basically P independent but ε_{CF} decreases (Fig. 3); such a result could be tested experimentally by, e.g., measuring the ratio of orbital and spin momenta, $\mathcal{R}_{L/S} = \langle L \rangle / \langle S \rangle$ (see Fig. 3, bottom panel).

Crystal-field splitting under pressure. The reduction of ε_{CF} with increasing pressure in Fig. 3 can appear puzzling

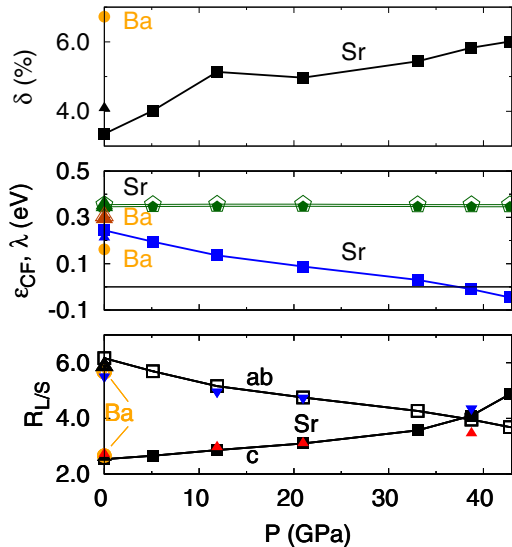
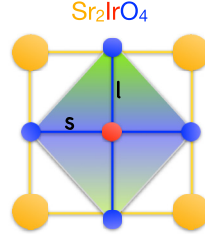


FIG. 3. Parameters as a function of pressure. Top: octahedral tetragonal distortion, $\delta = 2(l - s)/(l + s)$, where l and s are the long and short Ir-O bonds. Center: CF splitting (squares) and SO couplings (pentagons; open: λ_z ; solid: λ_{xy}). Bottom: $\mathcal{R}_{L/S}^\alpha = \langle L_\alpha \rangle / \langle S_\alpha \rangle$, with $\alpha = ab$ (open symbols) or $\alpha = c$ (solid symbols). Red and blue triangles: DMFT. Squares: atomic limit [26]. Calculations are for optimized coordinates. $P = 0$, solid triangles: experimental structure. $P = 0$, circles: Ba_2IrO_4 .

TABLE I. CF splitting (in meV) for $\lambda = 0$ and no rotations, with ($\varepsilon_{\text{CF}}^*$) and without ($\varepsilon_{\text{CF},0}^*$) Ir-O covalency effects; $\varepsilon_{\text{CF},0}^*$ is obtained in each case from the associated 15-orbital Ir t_{2g} +O $2p$ model. I_1 : $\delta = 0$. I_c : $\delta = 0$ and regular Sr cubes. I_c^∞ : I_c plus a large interlayer vacuum slab. I_2 : $\delta \sim 0.6\%$. For Ba_2IrO_4 , I_2 is the experimental case.

					
Sr_2IrO_4	$\varepsilon_{\text{CF}}^*$	$\varepsilon_{\text{CF},0}^*$	Ba_2IrO_4	$\varepsilon_{\text{CF}}^*$	$\varepsilon_{\text{CF},0}^*$
I_1	260	147	I_1	362	154
I_c	230	139	I_c	246	125
I_c^∞	380	147	I_c^∞	398	111
I_2	47	126	$I_2(E)$	158	146

at first. We identify its origin in covalency effects. This is related to the so-called “sign inversion” of ε_{CF} at ambient pressure. Since the octahedra are elongated (their tetragonal distortion is $\delta \sim 0.4\% > 0$, Fig. 3), one would in fact at first expect $\varepsilon_{\text{CF}} < 0$ [18,45–47], but this is at odds with, e.g., the experimental $\mathcal{R}_{L/S}$ [18,45]. Several calculation methods have shown that indeed $\varepsilon_{\text{CF}} > 0$ [14,38,48–50], recovering the agreement with experiments [48]. The inversion has been ascribed to various effects: long-range surroundings [48], π -bonding enhancement for the yz/xz orbital by staggered octahedral rotations [51], and the anisotropy of charge-transfer energies [49,52]. While all these mechanisms play a role, the latter turns out to be key to explain the CF reduction under pressure.

In order to show this, for simplicity we first set $\lambda = 0$ and undo octahedral rotations; for $P = 0$ this yields $\varepsilon_{\text{CF}}^* \sim 140$ meV. Pressure, while decreasing the CF splitting, increases the tetragonal distortion δ (Fig. 3). Consistently, increasing δ up to the maximum in Fig. 3 yields a very large reduction of $\varepsilon_{\text{CF}}^*$ (Table I). The associated CF splitting with no Ir-O covalency effects, $\varepsilon_{\text{CF},0}^*$, is obtained from the corresponding model including explicitly all the O $2p$ orbitals. For $\delta = 0$, we perform these calculations also for regular Sr cubes, and with a vacuum slab between IrO_6 layers. In all these cases $\varepsilon_{\text{CF},0}^*$ changes little. The large variation in $\varepsilon_{\text{CF}}^*$ mostly arises from the anisotropy of Ir-O charge-transfer energies [26]. Table I shows that this conclusion also applies to Ba_2IrO_4 , for which we find $\varepsilon_{\text{CF}} > 0$, in agreement with the experimental in-plane pseudospin configurations [50,53], and is likely to apply to most t_{2g} layered perovskites.

Persistence of insulating state under high pressure. We now put everything together and address the core question: Do strong correlations persist up to very high pressure, or, as one would expect in a small-gap Mott system, pressure quickly suppresses them, leading to moderate masses and a

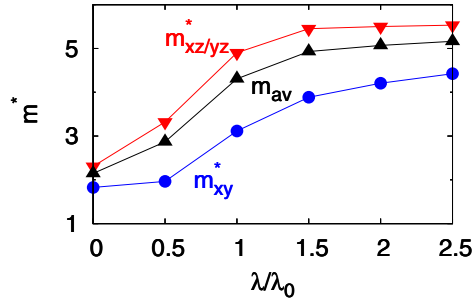


FIG. 4. Effective masses as a function of λ , for $U_{av} = 2$ eV, everything else remaining fixed. Calculations are for $\beta = 20$, sufficient to capture the trends; λ_0 is the actual value of λ .

weakly correlated metallic state? First, we find that persistent ($j_{\text{eff}} = \frac{1}{2}$)-orbital ordering imposes constraints on how a correlated insulating state can be formed and stay under pressure. This is because of the three-band-like nature of the system. Indeed, for $\lambda = 0$ the solution is metallic, masses are moderate, the configuration is close to $(xy)^2(xz, yz)^3$, and of course there is no ($j_{\text{eff}} = \frac{1}{2}$)-orbital ordering. Switching on λ , the configuration eventually changes to $(j_{\text{eff}} = 1/2)^1(j_{\text{eff}} = 3/2)^4$. Since the $j_{\text{eff}} = 1/2$ state is half filled, the associated effective mass is $m_{j_{\text{eff}}=1/2}^* = 1 + 3(m_{av} - 1)$ [15], where m_{av} is the average mass. The latter is relatively small for moderate λ and U_{av} , but, everything else staying the same, it increases with λ (see Fig. 4); instead, $m_{j_{\text{eff}}=3/2}^* \sim 1$ since the $j_{\text{eff}} = 3/2$ quartet is fully occupied.

Starting from these conclusions, depending on the actual parameters, for realistic U_{av} , it can be seen that the mass $m_{j_{\text{eff}}=1/2}^*$ can diverge (yielding a Mott phase) in three qualitatively different ways [15]: (i) xz/yz -orbital order: $\varepsilon_{CF} < 0$ and large, the configuration is $(xy)^1(xz, yz)^4$, and $m_{av} \sim [2 + m_{xy}^*(n_{xy}^\sigma = \frac{1}{2})]/3$, with the mass tensor a diagonal matrix, and $\alpha_1 = 0$. In this case the system is on the left-hand side of the topological point (Fig. 2), and m_{av} is large because the xy channel is about half filled. (ii) xy -orbital order: $\varepsilon_{CF} > 0$ and large, $\alpha_2 = 0$; this is less likely to be insulating for moderate U_{av} , since the $n_{xz}^\sigma = n_{yz}^\sigma = 3/4$. Here, $m_{av} = [1 + 2m_{xz/yz}^*(n_{xz/yz}^\sigma = \frac{3}{4})]/3$, and the only nonzero off-diagonal masses are $|m_{xz, yz}^*| = m_{xz/yz}^* - 1$. (iii) $j = \frac{1}{2}$ -orbital ordering: The t_{2g} orbitals have fractional occupations, the configuration is close to $[(xy)^{5/3}(xz)^{5/3}(yz)^{5/3}]$, and the mass tensor has large (symmetry allowed) off-diagonal

elements

$$|m_{xz, yz}^*| = m_{xz/yz}^* - 1 = (m_{av} - 1) \left(1 - \frac{\delta m_{av}}{3(m_{av} - 1)} \right), \quad (4)$$

$$|m_{xy, xz/yz}^*| = \frac{m_{av} - 1}{\sqrt{1 - 2p - 8p^2}} \left(1 - p + \frac{(1 + 8p)\delta m_{av}}{6(m_{av} - 1)} \right), \quad (5)$$

where $\delta m_{av} = m_{xy}^* - m_{xz/yz}^*$, and $p = 0$. At ambient pressure, the system is closer to (iii) for realistic λ values (Fig. 2), but with $p \neq 0$. Increasing pressure, we find that the picture does not change; the system moves even closer to (iii). Everything else staying the same, m_{av} should correspondingly increase in this process, but moderately; accounting in addition for the increase in W due to pressure, m_{av} decreases. This leads to mass reduction (see Fig. 1, masses for fixed $U_{av} \sim 2.4$ eV), and a breakdown of the insulating state at low pressure.

However, so far, a key element was neglected: The screened Coulomb interaction also increases under pressure. In $3d$ systems [54–56], with the cRPA method, the increase was found to be too small to avoid an IMT under pressure [55]. Remarkably, for Sr_2IrO_4 ($5d$ electrons), with the same approach we find that it is however sufficient to keep U_{av}/W almost constant (Fig. 1 and Supplemental Material [26]). The $j_{\text{eff}} = 1/2$ effective masses obtained including such an effect are identified by crosses in Fig. 1. The figure shows that they sizably increase with respect to the constant U_{av} case, at 40 GPa from about 5 to 20, keeping the system close to the IMT. This corresponds to an increase in m_{av} , while $\delta m_{av} \rightarrow 0$ and $p \rightarrow 0$.

Conclusion. In this Letter we studied the effects of uniform pressure P on the electronic properties of the spin-orbit assisted correlated insulator Sr_2IrO_4 . We have shown that, surprisingly, the screened Coulomb repulsion increases sufficiently to keep the U_{av}/W ratio constant up to 40 GPa. This leads to very high masses, holding the system close to the insulator-to-metal transition. Without such an effect, metalization with moderate masses would already occur below 12 GPa. In this situation, weak interactions, e.g., leading to short-range correlations, charge-density wave instabilities, or disorder, can be enough to turn the system insulating. Similar effects are likely to be at play in other $5d$ systems.

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