We study the correlated electronic structure of single-layer iridates based on structurally-undistorted Ba$_2$IrO$_4$. Starting from the first-principles band structure, the interplay between local Coulomb interactions and spin-orbit coupling is investigated by means of rotational-invariant slave-boson mean-field theory. The evolution from a three-band description towards an anisotropic one-band ($J=\frac{1}{2}$) picture is traced. Single-site and cluster self-energies are used to shed light on competing Slater- and Mott-dominated correlation regimes. We reveal a clear asymmetry between electron and hole doping, notably in the nodal/anti-nodal Fermi-surface dichotomy at strong coupling. Electron-doped iridates appear comparable to hole-doped cuprates due to the different sign of the next-nearest-neighbor hopping $t'$. 

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Iridum oxides based on the Ruddlesden-Popper series pose a particular challenging electronic structure problem [1][3]. The cooperation of strong spin-orbit coupling (SOC) with 5d-shell Coulomb interactions stabilizes insulating phases at stoichiometry below room temperature. Since these compounds usually show also antiferromagnetic (AFM) ordering, it is debated if Mott- or Slater mechanisms rule the observed insulating states [4][6]. Despite formally assumed weaker electronic correlations, the question arises if iridates still display deeper analogies to layered ruthenates or high-$T_c$ cuprates in view of non-BCS superconducting properties [7].

While the Sr compound of single-layer ruthenates has ideal tetragonal symmetry, the sister compound Sr$_2$IrO$_4$ shows tilting of the IrO$_6$ octahedra. In contrast Ba$_2$IrO$_4$ (see Fig. 1) is again free from distortions [8] and thus serves as a canonical system with a single Ir ion in the paramagnetic (PM) unit cell [9][10]. The AFM insulating phase of Ba$_2$IrO$_4$ has an Ir local magnetic moment of 0.34$\mu_B$, with an easy axis perpendicular to the c-axis [11], and is stable up to $T_N=240K$. Only a small charge gap of about $\sim0.2$ eV is deduced from angle-resolved photoemission spectroscopy (ARPES) measurements [9].

Theoretical studies of (Ba,Sr)$_2$IrO$_4$ based on variational Monte-Carlo [12] as well as density functional theory (DFT) combined with dynamical mean-field theory [4][5][13] support the original heuristic picture of a correlation-mediated spin-orbit driven insulator. Therein the SOC discriminates the Ir 5$d$($t_{2g}$) into effective $J_{\text{eff}}=\frac{1}{2},\frac{3}{2}$ states [14]. While four electrons of Ir$^{4+}$ fill up $J_{\text{eff}}=\frac{3}{2}$ completely, one electron remains in $J_{\text{eff}}=\frac{1}{2}$ at low energy. The interacting half-filled band at the Fermi level is then either gapped mainly due to the Slater mechanism forming an AFM state or directly by electronic correlations with secondary magnetic ordering.

Doping of the iridates is achievable [15], and recent experimental works succeeded to reveal a subtle electronic structure for both electron- and hole doping [16][19]. By surface electron doping of Sr$_2$IrO$_4$ [16], the quasiparticle (QP) strength seems to vary along the Fermi surface, somehow reminiscent of the famous 'Fermi-arcs' known from hole-doped cuprates. Though effective hole-doping of Sr$_2$IrO$_4$ also shows $k$-selective features [17], the states within the Fermi area appear much more incoherent.

In this work we focus on single-layer tetragonal Ba$_2$IrO$_4$ as a test case for basic accounts on the intriguing spin-orbit assisted correlation physics. From the realistic band structure at stoichiometry effective low-energy three- and one-band Hubbard models are constructed to assess the possible correlation regimes. Local and non-local self-energy representations are employed to study metal-insulator transition and doping effects. Fermi-surface differentiations in qualitative agreement with recent experimental findings are revealed. An obvious dichotomy in the doped fermiology between electron and hole doping is found at strong coupling, pointing towards the electron-doped case as the candidate for a proper ana-
logue to the hole-doped cuprate regime.

Theoretical framework: First-principles DFT calculations in the local density approximation (LDA) are performed for Ba$_2$IrO$_4$ in the $I4/mmm$ space group according to crystal data by Okabe et al. Computations are performed using a mixed-basis pseudo-potential scheme with and without the inclusion of spin-orbit coupling. We construct maximally-localized Wannier functions (MLWFs) for the Ir 5$d$($t_{2g}$)-based low-energy bands close to the Fermi level from the LDA calculations without SOC. Therefrom an initial three-band Hubbard Hamiltonian in the original 5$d$($t_{2g}$) basis of orbitals $m, m' = yz, xz, xy$ and with local spin-orbit term on Ir sites $i$ is drawn, i.e.

$$H = \sum_{\mathbf{k} \sigma \mathbf{m} \mathbf{m}' \mathbf{\sigma} \mathbf{\sigma}'} \epsilon_{\mathbf{k} \mathbf{m} \mathbf{m}' \mathbf{\sigma} \mathbf{\sigma'}} c_{\mathbf{k} \mathbf{m} \mathbf{\sigma}}^\dagger c_{\mathbf{k} \mathbf{m}' \mathbf{\sigma}'},$$

where $c_{\mathbf{k} \mathbf{m} \mathbf{\sigma}}$, $c_{\mathbf{k} \mathbf{m} \mathbf{\sigma}}^\dagger$ are creation, annihilation operators for the MLWF states with spin projection $\sigma = \uparrow, \downarrow$. The $t_{2g}$ dispersion $\epsilon_{\mathbf{k} \mathbf{m} \mathbf{m}' \mathbf{\sigma} \mathbf{\sigma}'}$ excludes on-site parts, which enter the crystal-field term $H_{CF}$. A Slater-Kanamori parametrization with Hubbard $U^{t_{2g}}$ and Hund’s exchange $J^{t_{2g}} = 0.14$ eV is used for $H_{INT}$, including density-density and spin-flip and pair-hopping terms. The SO term reads $H_{SO} = \lambda \sum_{\mathbf{i}, \mathbf{j}} \sum_{\mathbf{\sigma}} \mathbf{s}_{\mathbf{i}} \cdot \mathbf{l}_{\mathbf{j}}$, where $\lambda$ is the coupling constant and $\mathbf{s}$, $\mathbf{l}$ are spin- and angular-momentum operators. Because of the shift of $5d(e_g)$ to higher energies, restricting the general spin-orbit interaction matrix to the 5$d$($t_{2g}$) manifold is justified. The full problem is solved by mean-field rotational-invariant slave-boson (RISB) theory, using a multi-orbital single-site self-energy (see Fig. 1). Neglecting $H_{INT}$ leads to spin-orbit QP bands in very good agreement with the LDA+SOC low-energy dispersion.

For larger $\lambda$, the three-band Hamiltonian may be reduced to a tailored one-band problem for the effective $J = 1/2$ state at low-energy. In this restricted orbital space we also allow for an enlarged real-space correlated subspace via clusters of two and four sites (see Fig. 1). Therewith nonlocal correlations up to next-nearest neighbor (NNN) are incorporated. The initial cluster embedding is of cellular type, $k$-dependent self-energies are obtained for the two-site ($\Sigma(2)$) cluster and for the four-site ($\Sigma(4)$) cluster via further periodization using \[\Sigma(2)(\mathbf{k}, \omega) = \Sigma_{11}(\omega) + \Sigma_{12}(\omega) (\cos k_x + \cos k_y), \]

\[\Sigma(4)(\mathbf{k}, \omega) = \Sigma_{11}(\omega) + \Sigma_{14}(\omega) (\cos k_x + \cos k_y) + \Sigma_{14}(\omega) \cos k_x \cos k_y.\]

Albeit approximative, the cluster-RISB method has been proven capable to shed light onto relevant features of non-local correlation physics.

From three-band to effective one-band: The LDA calculations for Ba$_2$IrO$_4$ reveal dominant $t_{2g}$-like bands at low-energy, and a minor $e_g$-like electron pocket around $\Gamma$.

Static DFT+U computations lead to an upward energy shift of the latter pocket into the unoccupied region. Thus that $e_g$-derived contribution plays no vital role in the key correlation physics and is neglected in the following. Figure 2 displays the MLWF-based $t_{2g}$-like low-energy bands adapted from LDA without SOC. Including spin-orbit coupling in the subsequent RISB treatment shifts the lower band manifold with effective $J^\prime = 3/2$ down in energy (see Fig. 2b). Inclusion of $H_{INT}$ shifts those bands even further away from the Fermi level $\epsilon_F$, eventually resulting in completely filled $J^\prime = 3/2$ and half-filled $J^\prime = 1/2$ states (cf. Fig. 2c). This limit may be understood from a constructive interplay between Hund’s third rule and the minimization of Coulomb interactions in the Ir(5$d^9$) shell. The orbital character of the remaining half-filled band at $\epsilon_F$ is indeed nearly exclusively of $J = 1/2$ kind. Due to its isolation, the low-energy physics of single-layer iridate can be further analyzed to a good approximation within a one-band picture. From the three-band calculation with $\lambda = 0.4$ eV and neglecting $H_{INT}$, we therefore Fourier transform the isolated $J = 1/2$ band to obtain a single-band tight-binding parametrization. In addition to a local Coulomb interaction scaling with a Hubbard $U$, a nearest-neighbor (NN) spin-spin interaction term is introduced to take care of the spin-orbit induced anisotropic spin ordering. The low-energy one-band iridate Hamiltonian is then given by

$$H_{1B} = \sum_{ij} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{\Gamma}{\langle ij \rangle} S_i^{(e)} S_j^{(e)},$$

where $t_{ij}$ marks the hoppings of the underlying
$J=1/2$ dispersion with bandwidth $W=1.55$ eV and $\Gamma>0$ as the AFM coupling in $x$-direction. The first near-neighbor in-plane hoppings amount to $(t, t', t'', t''') = (-205, -16, 35, 13)$ meV, and the inter-layer coupling is given by $t_{i,z} = -11$ meV. Based on the work of Katukuri et al. [24], a value $\Gamma = 12$ meV is computed for the anisotropic interaction. In the following we discuss the effective one-band physics obtained from mean-field RISB. Half filling is generally marked by the orbital occupation $n=1$.

One-band physics in single-site RISB: Let us first focus on the pure on-site self-energy treatments, neglecting inter-site terms. Disregarding the spin-spin interaction, the PM Mott transition with vanishing QP weight $Z=\left[1 - \frac{\partial}{\partial \Sigma} \Sigma_{\omega=0} \right]^{-1}$ occurs at $U_{c,PM} = 2.85$ eV, i.e. $U_{c,PM}/W \sim 1.84$. To account for AFM order we use a $\sqrt{2} \times \sqrt{2}$ unit-cell architecture, treating two NN Ir ions with their respective on-site $\Sigma$ (cf. Fig. 3). The explicit spin-spin interaction is now handled in mean-field decoupling, i.e. $S^{(x)}_{i,y} S^{(x)}_{i',y} \rightarrow S^{(x)}_{i,y} \langle S^{(x)}_{i',y} \rangle$. At stoichiometry antiferromagnetism with staggered moments aligned along the $x$-axis marks the ground state for any $U>0$. For $U_{c,AFM} = 0.8$ eV the system becomes insulating at a first-order transition (see Fig. 3a). Thus the critical $U$ for the metal-insulator transition (MIT) is strongly lowered when allowing for magnetic order. Figure 3 shows that the spin moment pointing along $x$ becomes highly susceptible to small interaction changes around $U_{c,AFM}$, but saturates only at much larger interaction strength. Therefore in this case the MIT is not of strong Mott type, i.e. does not result in complete electron localization. It has magnetic-driven signature, where the charge-gap opening results in the formation of increased-dispersive Slater-like bands [32]. Away from stoichiometry, the AFM order remains stable up to rather large doping, as long as $\langle S^{(x)} \rangle$ is finite. Symmetric 30% electron/hole doping is necessary to render $\langle S^{(x)} \rangle \rightarrow 0$ for $U=1$ eV.

One-band physics in cluster RISB: To evaluate the relevance of inter-site self-energy contributions especially in the doped regime, we extend the one-band investigations towards computations within a cluster framework. Therein the spin-spin interaction term in eq. 4 may be treated in complete many-body form on the local clusters. Already the minimal in-plane two-site cluster involving NN Ir sites allows for an insight on the key effects of an inter-site self-energy $\Sigma_{12}$. At half filling, the PM Mott transition occurs at $U_{c,PM}^{(2)} = 1.5$ eV, accompanied by a jump of the already negative NN spin-correlation $\langle S_1 S_2 \rangle$ towards even lower values (cf. Fig. 3a). This marks the dominance of the inter-site singlet cluster state in the Mott-insulating regime. When allowing for the AFM phase, Fig. 3b displays that the MIT occurs as in the two-single-site study at $U_{c,AFM}^{(2)} = 0.8$ eV.

In the doped cases, we focus on cluster effects in the PM phase. Figures 3b-d show key information on the significance of non-local self-energy terms. For sizable $U$ the two-particle singlet on the two-site cluster dominates the multiplet states at $n=1$. With doping, increasing weight is transferred to the triplet as well as one/(three)-particle states when adding holes/electrons. Interestingly, there is a small electron-hole asymmetry: the singlet/triplet is more/less pronounced with hole- than electron doping, pointing towards a more robust AFM tendency in the hole-doped regime. Inter-site QP weights become relevant for $U>1$ eV. Their magnitude is sizable at small doping and negligible about 20% away from half filling. In the four-site cluster description the correlation strength is generally enhanced, documented by the smaller on-site QP weight $Z_0$. Moreover the relation $|Z_{NNN}| > |Z_{NN}|$ holds with electron doping, pointing...
towards anisotropic electronic correlations.

To elucidate this further, we finally discuss k-dependent signatures at finite doping. In principle two scenarios may hold: either doping right within the Slater-Hubbard bands takes place ($U=W$), or it results in the build-up of a renormalized FS readily from the original itinerant dispersion ($U\gg W$). In the first case, k-space differentiation occurs because of the energy dependence of the gap-forming bands (compare Fig. 3b). Then here, hole(electron) doping would lead to FS pocket-formation around $X'(M')$, as indeed verified by plotting the doped FS within our two-single-site approach in Fig. 5a,b. Such a scenario has been detected in ARPES measurements for effective hole doping of Sr$_2$IrO$_4$ [17].

In the stronger correlated scenario, k-selectivity in a one-band picture is usually due to finite inter-site terms $\Sigma_{\alpha\beta} \neq 0$ for $\alpha \neq \beta$. We may encounter such effects via our periodized in-plane cluster self-energies. For instance, the QP weight $Z=Z(k)$ for the model [4] can vary based on NN and NNN self-energies of the four-site cluster (see Fig. 5h) [25]. Let’s focus on the interacting fermiology, i.e., $Z=Z(k_F)$, where $k_F$ is the Fermi wave vector. For both dopings, hole- and electron-like, Figs. 5e,f show an obvious in-plane nodal/anti-nodal dichotomy. The QP weight on the FS along the node $(0,0)-(\pi/2,\pi/2)$ is larger than along the anti-node $(0,0)-(0,\pi)$. Though the absolute differences are small within cluster-RISB, it serves as a proof of principles for k-space differentiation by electronic correlations, in agreement with recent ARPES experiments on surface electron-doped Sr$_2$IrO$_4$ [10].

Second, there is a substantial difference in the $k$-differentiation of $Z(k_F)$ between both doping directions. The electron-doped case exhibits stronger QP-weight variation along $\epsilon_F$ than the hole-doped case. In other words for same interaction strength, theory predicts that electron doping of single-layer iridates is more likely to cause a Fermi-arc structure than hole doping. This finding is reminiscent of the electron-hole dichotomy in cuprates [36, 37], yet with a twist: in cuprates, the hole-doped case is assumed more susceptible for $k$-selective correlations and the electron-doped case is stronger ruled by AFM order. Note that the former identification of a more stable two-particle singlet on the iridate hole-doped side completes this mirroring. Additionally, for all encountered doping distances from $n=1$, the intra-site $Z$ is always somewhat lower on the electron-doped side.

The qualitative difference may be explained by the relevance of hopping characteristics beyond NN [38]. Because of the different sign of the NNN $t'$ in both compound families, the enhanced correlation-susceptible van-Hove singularity at $M$ in reciprocal space is above(below) the Fermi level for iridates(cuprates) as shown in Fig. 5c. Thus from a phase-space argument, hitting stronger correlations at the anti-node takes place by electron(hole) doping of iridates(cuprates). Note that hoppings beyond NNN are effective in shifting the iridate van-Hove singularity further away from $\epsilon_F$.

Summary: An effective $J=1/2$ low-energy one-band modelling is derived for single-layer iridates from the initial spin-orbit interacting $t_{2g}$ manifold. For $U \lesssim 1.25$eV Slater-like behavior dominates, while for $U \gtrsim 1.25$eV Mott-Hubbard physics is more in control. In reality a subtle interplay between both limits is expected [6]. Our theoretical approach predicts an electron-hole doping asymmetry, taking place at weaker as well as at stronger coupling and has partly already been confirmed by recent experiments [16, 17]. FS pockets that occur for weaker electron-electron interaction are more likely for hole doping, whereas Fermi arcs may set in for stronger interaction with higher tendency on the electron-doped side. Therefore electron-doped iridates are candidates for a possible analogue to hole-doped cuprates.

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