The concept of Hund’s metal has been introduced [1] to rationalize the observation that, in theoretical models of iron-based superconductors, ruthenates, and other materials, many relevant observables turn out to be remarkably dependent on Hund’s coupling $J$ rather than on the Hubbard interaction $U$. In Ref. [2] it has been shown that for integer occupation different from global half-filling–Mott localization happens in two steps. As a function of Hubbard $U$, the effective mass enhancement first grows, then flattens and remains relatively large in a wide range of interactions [2], while the critical $U$ for Mott localization is pushed to very large values [3]. Therefore we have a wide region of parameters with a strongly correlated metal which is resilient with respect to Mott localization despite a large value of $U$ and a sizeable $J$. This two-faced role of Hund’s coupling has been called Janus effect. Several theoretical studies [4–8] enriched the picture showing: (i) anomalies of the spin response [9] and finite-temperature spin-freezing at the crossover between a standard and a Hund’s metal [10]; (ii) an effective orbital decoupling associated with the quenching of charge excitations between different orbitals, favoring a strong differentiation in the degree of correlation of different orbitals (orbital selectivity) [11–13]; (iii) an anomalous increase of charge fluctuations associated with the effective mass growth [12]; (iv) enhancements of the nematic orbital differentiation [14] and charge susceptibilities [15]. The latter have been proposed as boosters for superconductivity and can be naturally connected with charge-ordering instabilities, which have been proposed in iron-based superconductors [16] and chromium pervoskite compounds $\text{ACrO}_3$ [17, 18].

Despite the fast advances of our understanding of these phenomena, the very existence of a metallic solution when both $U$ and $J$ are large remains surprising since both interactions are intuitively expected to constrain electron mobility. In this work we address this and related questions in a three-orbital correlated model, and we link the survival of the Hund’s metal to the existence of two or more competing insulating solutions. One of these is a charge-disproportionated insulator which is compatible with the experimental observation of an unusual metal-insulator transition in $\text{PbCrO}_3$ [17, 18]. We consider the popular three-orbital Kanamori model,

$$H_{\text{int}} = U \sum_a n_{a\uparrow} n_{a\downarrow} + (U - 3J) \sum_{a < b, \sigma} n_{a\sigma} n_{b\sigma}$$

$$+ (U - 2J) \sum_{a \neq b} n_{a\uparrow} n_{a\downarrow} + J \sum_{a \neq b} n_{a\sigma} n_{b\sigma}$$

$$+ J \sum_{a \neq b} \sum_{\sigma} \hat{d}_{a\sigma}^\dagger \hat{d}_{b\sigma}^\dagger \hat{d}_{a\sigma} \hat{d}_{b\sigma}$$

$$= (U - 3J) \frac{n^2}{2} - J \left( 2S^2 + \frac{1}{2} L^2 \right),$$

where $\hat{n} = \sum_{a, \sigma} \hat{d}_{a\sigma}^\dagger \hat{d}_{a\sigma}$ is the local electron number operator, whose expectation value is the average density $\bar{n}$ ($a = 1, 2, 3$ labels the orbital and $\sigma = \uparrow, \downarrow$ the spin components), while $S = \frac{1}{2} \sum_{a, \sigma, \sigma'} \hat{d}_{a\sigma}^\dagger \hat{\sigma}_{\sigma\sigma'} \hat{d}_{a\sigma'}$ and $L = \sum_{a, \sigma} \sum_{\sigma'} \hat{\epsilon}_{a\sigma} \hat{\epsilon}_{a\sigma'}$ are the spin and orbital angular momentum operators, with $\hat{\sigma}_{\sigma\sigma'}$ the Pauli matrices and $\hat{\epsilon}_{a\sigma} = -i \epsilon_{abc} \hat{O}(3)$ group generators.

The expression (2) is particularly transparent: the first term describes the on-site repulsion with strength $U = U - 3J$ and the second represents the exchange mechanism responsible for Hund’s rules, which, for positive $J$, favor high spin and orbital angular momentum configurations. The form (1), instead, details all the microscopic interactions. In particular, $U$ controls the repulsion between two electrons in different orbitals with same spin (second term in the first line). We solve this model in a wider range of parameters with respect to most previous investigations, considering also (i) the regime $U < 0$, which is usually discarded because some Coulomb matrix elements are negative, and (ii) both positive and negative interactions in a three-orbital correlated model, and
Figure 1. (a) Phase diagram from a color plot of $Z$ for $J > 0$ and $\bar{n} = 2.$ The dark blue region on the right is a Mott insulator (sketch on the right) whereas in the upper left part there are two insulating states: a Hund insulator (sketch on the top) in the dark blue region and an extreme negative charge-transfer insulator in the greyed area. (b): $Z$ plotted along different cuts at fixed $J/U$.

The result of an attractive $U^* < 0$ and a large Hund’s coupling, favoring configurations with three electrons so as to achieve the highest possible spin. In order to have an average density of two electrons per site, one site must remain empty every two triply-occupied sites. We label this state as Hund’s insulator (HI) 22.

The two insulating solutions have purely atomic character and we can understand their competition in the atomic limit $W = 0$. The MI is the ground state for $0 < J/U < 1/3$ and the HI for $1/3 < J/U < 3/4$. The two insulators are degenerate for $J/U = 1/3$. Interestingly, on the same line a third insulator with either singly or (high-spin) triply occupied sites, with equal probabilities $p_{1,1/2} = p_{3,1} = 1/2$, is degenerate alongside the MI and HI. Although this state is never the lowest-energy insulator, its presence is relevant for the properties of the metallic regime. For $J/U > 3/4$ the (unphysically) large negative $U^*$ favors maximal charge imbalance, so that each site is occupied by either 0 or 6 electrons with weight $p_{0,0} = 2/3$ and $p_{6,0} = 1/3$ (grey region in the plot).

We now turn to the metallic region separating the MI and HI. Figure 1(b) shows cuts of $Z$ at fixed $J/U$. For $J/U$ close to $1/3$ we observe the two-step localization (‘Janus effect’), with an almost symmetrical behavior of $Z$ with respect to the $J/U = 1/3$ line. Along the latter,
$Z$ never vanishes. In Fig. 2(a) we follow the population of each multiplet as a function of $U$, keeping $J/U$ fixed to 1/3. For small $J$ and $U$ (bottom left part of Fig. 1 phase diagram) all atomic multiplets are populated and hopping processes take place between any pairs of multiplets that are compatible with Wigner-Eckart theorem. An increase of either $U$ or $J$ has the same net effect of reducing $Z$ by disfavoring some multiplets, thereby reducing the phase space for hopping: an increase of $U$ reduces the population of multiplets with particle number different from the average density (in this case, $\bar{n} = 2$), whereas an increase of $J$ tends to reduce the population of multiplets with low spin.

For weak interactions the reduction is relatively slow, but when we reach the crossover to the Hund’s metal region, the differentiation in the multiplet occupation suddenly becomes strong: most multiplets have basically zero population, while only a few states survive and have a large probability which remains almost constant by further increasing the interaction. These are the high-spin states for each charge sector, which are also the building blocks of the three degenerate insulators. However, this strong phase-space limitation does not lead to a suppression of $Z$ as one might naively expect. The reason is that among the surviving multiplets we have high-spin states whose local occupation differs by one unit, allowing for hopping processes. This is only possible owing to the disproportionated nature of the HI and the other degenerate insulator, which ensures that local states with $n \neq 2$ are populated. Exactly on the $U* = 0$ line we have indeed finite hopping even when $U/W$ and $J/W$ go to infinity. When we depart from this line, the atomic states are no longer degenerate. However, reasonably close to it, the energy splitting is still small. This implies that a relatively small value of hopping is sufficient to overcome the splitting and give rise to a metal. As the ratio $J/U$ moves away from 1/3, the critical $U$ is more and more reduced, as shown in Fig. 1(b). Figure 2(b) shows the evolution of multiplet occupations along a vertical cut connecting the MI and HI (white dashed line in Fig. 1 fixed $U = 3.5W$). The main message is that $Z$ is maximized when the populations of the various multiplets become comparable, thereby optimizing the kinetic energy. This observation leads us to identify the Hund’s metal as a mixed-valence state which exists because of the decrease of some multiplets, thereby reducing the phase space for hopping: an increase of $\Delta$ reduces the population of multiplets with particle number different from the average density (in this case, $\bar{n} = 2$), whereas an increase of $\Delta$ tends to reduce the population of multiplets with low spin.

In panels (c) and (d) of Fig. 2 we plot the total, interorbital, and intraorbital local charge correlations, $C_{ab} = \langle \hat{n}_a \hat{n}_b \rangle - \langle \hat{n}_a \rangle \langle \hat{n}_b \rangle$, along the same lines of panels (a) and (b), respectively. Panel (c) shows that, inside Hund’s metal, interorbital correlations are zero along the $U = 3J$ line, reflecting the physics of orbital decoupling [12], whereas intraorbital correlations are finite, so that the total charge fluctuations remain finite in the whole Hund’s metal region. More interestingly, panel (d) reveals that intraorbital fluctuations are actually frozen to a constant value throughout the large $U$ cut connecting the MI and HI. This is due to the complete quench of intraorbital double occupancies. Instead, interorbital correlations remain active and their variation drives the change in the total charge fluctuations. The latter are indeed zero in the MI and become finite when entering the Hund’s metal, increasing rapidly as we approach the charge-disproportionated HI (naming, for $J > U/3$, when interorbital correlations change sign). The increase of charge correlations prehears to a phase-separation instability. The above phenomenology is consistent with Ref. [12], where the behavior of charge fluctuations is understood via the energetic analysis of hopping processes between the allowed multiplets.

Our analysis can be used to interpret experiments on $ACrO_3$ compounds, where the 3d orbitals are populated by two electrons per site. BaCrO$_3$ has been characterized as a Hund’s system which becomes insulating because of spin-orbital ordering [28], while $A = Ca$ is a surprising antiferromagnetic metal with structural anomalies [24] and $A = Sr$ displays phase separation [25]. Most importantly for this manuscript, PbCrO$_3$ is experimentally characterized as a charge-disproportionated insulator. References [17, 18] propose the same pattern we find for $U < 3J$, whereas Ref. [19] invokes a different disproportionation pattern, although with the same net effect of maximizing the total spin. This scenario is compatible with a crossover where the Ba compound has a positive $U*$ which is progressively reduced when moving towards the Ca and Sr systems, becoming eventually negative with Pb. This evolution of the effective interaction can
be attributed to electron-phonon coupling, which might further strengthen the charge disproportionation by lowering the lattice symmetry. At global half-filling, \( \tilde{n} = 3 \), the Janus effect does not occur (see Fig. 3). The key point is that, in this case, the MI and HI coincide and consist in a uniform solution with three electrons per site and \( S = 3/2 \). In other words, \( U \) and \( J \) do not compete but rather cooperate to create the maximal spin state. Hence, the region of large \( U \) and \( J \) is simply occupied by this Mott/Hund insulator. All multiplets except the ground state are filtered out by interactions and the lack of degenerate states in the atomic regime completely destroys Hund’s metal physics, confirming the crucial role of degenerate atomic insulators in the genesis of the correlation-resilient metal.

Notice that the maximal degeneracy found on the \( U = 3J \) line is specific to the choice of using the Kanamori interaction, in particular to the ratio \( r = 4 \) between the coefficients of the \( S^2 \) and \( L^2 \) terms in Eq. (2). For \( r \neq 4 \) – yet still enforcing the spin and orbital rotation invariance – the degeneracy between the MI and HI is preserved (albeit at a different \( J/U \) ratio) and the Hund’s metal survives. Using more involved interactions, such as the full Coulomb interaction for the whole \( d \) manifold, is clearly expected to reduce the symmetry of the problem and split the degeneracy between the insulators. However, as long as the different insulating states are reasonably close in energy, we still expect a Hund’s metal phenomenology with a finite critical value of \( U/W \).

The link between the persistence of metallic solutions at strong coupling and the degeneracy between atomic insulators can be strengthened by analysing the model for negative \( J \). The Hamiltonian (2) with \( J < 0 \) is believed to describe alkali-doped fullerides \( A_2C_{60} \) \((A = K, Rb, Cs)\), where the valence band is a threefold degenerate \( t_{1u} \) level Jahn-Teller (JT) coupled to the \( H_g \) vibrational modes of the \( C_{60} \) molecule. This electron-phonon coupling induces an effective interaction which has the same functional form of Hund’s coupling and opposite sign. Since the amplitude of the JT contribution exceeds Hund’s term, the net exchange coupling is negative, favoring low spin and angular momentum configurations \( \tilde{n} = 3 \). Although in fullerenes realistic values of the interactions imply \( U \gg |J| \), leading to a superconducting ground state adjacent to the Mott insulator \( \tilde{n} = 3 \), we shall nonetheless study the \( J < 0 \) model in the whole parameter space. In particular, we focus on the underlying normal state (metallic or insulating) in order to unveil the attractive counterpart of Hund’s metal.

In Fig. 4 we show the quasiparticle weight in the paramagnetic state at \( \tilde{n} = 2 \) and \( \tilde{n} = 3 \). The most notable feature is the similarity between the \( \tilde{n} = 3 \) JT system and the \( \tilde{n} = 2 \) Hund system (see Fig. 1). Analysing the atomic limit for \( \tilde{n} = 3 \), also here we find that the MI, characterized by three electrons on each site populating the lowest energy multiplet \( L = 1 \) and \( S = 1/2 \), becomes degenerate with a distinct and competing insulating state at a certain \( |J|/U \) ratio, in this case \( 1/2 \). Indeed, for \( |J|/U > 1/2 \) the energetically favorable insulator is an inhomogeneous state consisting of spin and orbital singlets, where every site is occupied by either two or four electrons, populating the \( L = S = 0 \) states with fractions \( p_{2,0,0} = p_{4,0,0} = 1/2 \). Charge fluctuations are gapped here by the singlet binding energy. This disproportionated insulator can be seen as a pairing insulator \( \tilde{n} = 2 \) formed by an incoherent collection of bosonic pairs which are not allowed to condense in a superfluid.

Just like in the \( \tilde{n} = 2 \) Hund system, a metallic solution intrudes between the two insulators. The evolution of the multiplet occupation mirrors the results of Fig. 2. For small interactions all multiplets are populated. Then, a sharp crossover leads to a highly correlated JT metal \( \tilde{n} = 3 \) (in analogy with Hund’s metal), where the only populated multiplets are those already present in the insulating states, namely the low-spin states with \( 2, 3, \) or 4 electrons per site. As in the Hund system, the emergent JT metal can be regarded as a mixed-valence state where, due to the degeneracy between insulating states, the systems gains kinetic energy via hopping processes connecting the local configurations of the two insulators. The maximum energy gain and largest \( Z \) are reached exactly on the line of degeneracy, where the ground state is precisely halfway between the two insulators, i.e., \( p_{3,1,1/2} = p_{2,0,0} + p_{4,0,0} = 1/2 \). The similarity between JT and Hund’s metals is further strengthened by the ‘Janus effect’, shown in Fig. 4(d). The lower degree of degeneracy of the JT system results
in a smaller asymptotic value of $Z$ with respect to the Hund system. For $n = 2$ we recover a situation similar to the $n = 3$ Hund case. Here the pairing insulator (lowest spin state) coincides with the MI (minimal charge imbalance). Just like in Hund’s case, $U$ and $J$ promote the same insulating state. Therefore the metallic solution survives only in the bottom left part of the phase diagram and no JT metal emerges. There is however an important difference between the JT and Hund systems. Despite the common origin of the mixed-valence metals, in the JT system the charge-transfer gap remains positive for any $J < 0$ and does not vanish on the line of degeneracy. Consequently, in the JT system the metallic solution at half-filling and large $U \sim 2|J|$ remains stable upon doping.

In this manuscript we have established a strong link between the existence of a Hund’s (or Jahn-Teller) metal (i.e., a metal which survives when the interactions are much larger than the bare kinetic energy) and the degeneracy between competing insulators. In the paradigmatic case of a Kanamori model with two electrons in three orbitals, the two insulators are a high-spin MI and a charge-disproportionated HI. The Hund’s metal appears as a mixed-valence state in which the local state fluctuates between the atomic configurations of the two insulators. For three electrons in three orbitals the two interactions favor the same insulating solution and the Hund’s metal state disappears. A negative Hund’s coupling gives rise to an analogous scenario, where for three electrons we find degenerate insulators and a correlation-resilient Jahn-Teller metal [29, 33], while two electrons experience a standard Mott transition as the two interactions cooperate to form a pairing insulator. We eventually connect the physics of Hund’s metals with that of charge disproportionation. Remarkably, the exact charge disproportionation characterizing the HI has been experimentally observed in PbCrO$_3$.

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