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Metal-insulator transition and topological properties of pyrochlore iridates

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Combining density functional theory (DFT) and embedded dynamical mean-field theory (DMFT) methods, we study the metal-insulator transition in $R_2\text{Ir}_2\text{O}_7$ ($R=\text{Y, Eu, Sm, Nd, Pr, and Bi}$) and the topological nature of the insulating compounds. Accurate free energies evaluated using the charge self-consistent DFT+DMFT method reveal that the metal-insulator transition occurs for an A-cation radius between that of Nd and Pr, in agreement with experiments. The all-in-all-out magnetic phase, which is stable in the Nd compound but not the Pr one, gives rise to a small Ir^{4+} magnetic moment of $\approx 0.5\mu_B$ and opens a sizable correlated gap. We demonstrate that within this state-of-the-art theoretical method, the insulating bulk pyrochlore iridates are topologically trivial.

Interest in $5d$ compounds has blossomed in recent years, as they provide a promising playground for interesting new physics¹⁻⁵ arising from the interplay of the atomic spin-orbit coupling (SOC), which scales as Z^4 with the atomic number Z , and the electronic correlations, which are reduced due to the spatially more extended wavefunctions of the $5d$ ions. Rare-earth pyrochlore iridates ($R_2\text{Ir}_2\text{O}_7$) have drawn intensive attention in recent years⁶ because of their geometrically frustrated lattice, which favors the spin-liquid phase,⁷ and the possibility to host various nontrivial topological phases in the bulk,^{8-11,13} thin films,^{14,15} and domain walls.¹⁶ Depending on the radius of the A-site cation, rare-earth pyrochlore iridates undergo a metal-insulator transition (MIT),¹⁷ concomitant with a transition to an all-in-all-out (AIAO) magnetic state.¹⁸⁻²⁰ Nevertheless, when the A-site cation is Pr or the more covalent Bi, the corresponding pyrochlore iridates are metallic down to very low temperature with no long-range magnetic ordering.^{7,21}

Recently, inspired by pioneering work based on a tight-binding model,⁸ there have been many studies focusing on nontrivial topological phases in bulk pyrochlore iridates.^{9-11,13} Based on calculations using the local density approximation (LDA) with Hubbard U (LDA+ U) including SOC, Wan *et al.* predicted that $\text{Y}_2\text{Ir}_2\text{O}_7$ can host nontrivial Weyl semimetal and axion insulator phases.¹¹ Using a single-band Hubbard model on the pyrochlore lattice, Witczak-Krempa *et al.* developed a topological phase diagram where various topological phases can be identified at the mean-field level,¹² and Go *et al.* confirmed the presence of certain nontrivial topological phases based on cluster-DMFT calculations.¹³

Iridates are known to be beyond the range of applicability of LDA+ U calculations, since dynamical local correlations are crucial to explain their complicated electronic structure. Instead, the LDA+DMFT method²³ correctly gives the boundary of the MIT in the Ruddlesden-Popper (RP) series, and the fine details of the ARPES measurements of Sr_2IrO_4 ²⁴ can only be understood by comparing with the LDA+DMFT spectral functions, which include essentially exact correlation effects local to a given Ir atom.²³ Moreover, the LDA+ U method overestimates the stability of the insulating mag-

netically ordered states, and the value of U , which is to some extent an adjustable parameter, often needs to be adjusted for each member of the series. Thus, a consistent treatment of the electronic correlations together with spin-orbit coupling at the LDA+DMFT level is necessary to shed light on the MIT in the pyrochlore iridates.

In this work, we have carried out all-electron charge self-consistent LDA+DMFT²⁵ calculations. We have studied a series of pyrochlore iridates in both the paramagnetic (PM) and AIAO magnetic states, focusing on the metal-insulator transition with respect to the A-cation radius and the topological nature of the insulating states. We demonstrate that the MIT occurs in those compounds which can host an AIAO magnetic ground state. Because of the large degree of geometric frustration in the pyrochlore lattice, a significant quasiparticle mass in the PM state is needed to destabilize the Fermi liquid formation at the expense of magnetic long-range order. The tuning across the boundary between the Kondo-screened Fermi-liquid solution and the magnetic AIAO solution is achieved with A-cation substitution. The most important consequence of the latter is the change of oxygen coordinates, which results in a slightly different Ir hybridization and effective Ir- t_{2g} bandwidth. Based on calculations of the wavefunction parities in a many-body context, we conclude that the insulating pyrochlore iridates are likely to remain topologically trivial.

Our LDA+DMFT calculations were done using the projection/embedding implementation,²⁵ which avoids downfolding or model building, and adds dynamic local correlations to a set of localized quasi-atomic orbitals. The DFT part is implemented using the WIEN2k package.²⁶ All parameters of the calculation are identical to those used in our earlier work on RP iridates.²³ We also performed LDA+ U calculations using the VASP code²⁷ for comparison. Details are provided in the Supplemental Materials.²⁸

For the series of $R_2\text{Ir}_2\text{O}_7$ ($R=\text{Y, Eu, Sm, Nd, Pr, and Bi}$) compounds, we considered the PM and AIAO magnetic solutions. We found that the magnetic AIAO solution could be stabilized (that is, we could find a stable or metastable solution) only in the Y, Eu, Sm, and Nd compounds, and was always insulating (see spectral functions in the Supplement), while the PM solution could be

stabilized for all materials and was always metallic, consistent with experiments.^{17,21} Naively, the MIT in the six $R_2\text{Ir}_2\text{O}_7$ compounds can be understood in terms of the bandwidth of the Ir- t_{2g} bands, as shown as inset of Fig. 1(a). For instance, at the LDA level there is a critical value of about 2.6 eV for the Ir- t_{2g} bandwidth separating the compounds with insulating and metallic ground states. A detailed analysis reveals that the bandwidth is closely correlated with the hybridization function, and its variation can be attributed to the change of the oxygen coordinates (see the Supplement).

To determine the relative stability of the two phases, we computed the free energy differences between PM and AIAO states following Ref. 39, yielding the results displayed in Fig. 1(a). For comparison, the difference of LDA+U total energies is also shown, but reduced by a factor of 20 to fit in the plot. For each method, we used the same U as determined in our previous study of RP iridates.²³ The LDA+U energy difference is about 400 meV on average, much too large to be compatible with the known ordering temperatures. This is hardly surprising since the local moments are zero in the PM state within LDA+U, which cannot describe fluctuating local moments. As a result, the energy of the PM state is severely overestimated.

Experimentally, $\text{Pr}_2\text{Ir}_2\text{O}_7$ has a non-magnetic metallic ground state,¹⁷ correctly described by our LDA+DMFT calculations where the AIAO magnetic state can not be stabilized. In LDA+U by contrast, only $\text{Bi}_2\text{Ir}_2\text{O}_7$ is metallic, while the ground state of $\text{Pr}_2\text{Ir}_2\text{O}_7$ is an AIAO magnetic insulator. We also studied other possible magnetic phases within LDA+DMFT. We found that the three-in-one-out configuration can also be stabilized for some compounds, but is higher in energy than the AIAO state (*e.g.*, by 23 meV for $\text{Y}_2\text{Ir}_2\text{O}_7$). The two-in-two-out state, on the other hand, is not stable in this method. Therefore, the AIAO magnetic state is indeed the ground state of insulating pyrochlore iridates within LDA+DMFT.

Our self-consistent LDA+DMFT calculations also give reasonable estimates of the free-energy differences between PM and AIAO states as shown in Fig. 1(a). The average free-energy difference is about 20 meV, which is about twice the typical magnetic ordering temperature (about 100 K) observed experimentally.¹⁷ Moreover, the trend of the LDA+DMFT free-energy difference with respect to the A-cation radius follows loosely the experimental magnetic ordering temperature.^{17,21} One particular system is $\text{Nd}_2\text{Ir}_2\text{O}_7$ located close to the MIT phase boundary with an experimental critical temperature of about 30K. In our LDA+DMFT calculations, both AIAO insulating and PM metallic states can be obtained, and the PM state is slightly lower (~ 1.5 meV) in energy than the AIAO state. We note that this energy difference is rather small, comparable to the uncertainty of our quantum Monte-Carlo statistics. Given also the fact all our calculations are done at 50 K, we still believe that the AIAO insulating state is the preferred

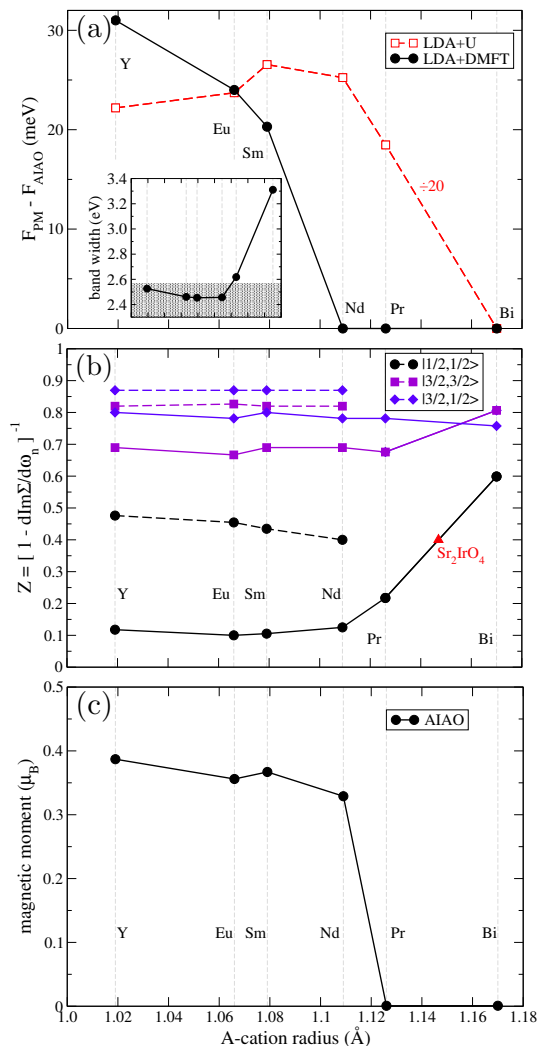


FIG. 1: (Color online) Variation with respect to the A-cation radius of computed properties of $R_2\text{Ir}_2\text{O}_7$ ($R = \text{Y, Eu, Sm, Nd, Pr, Bi}$). (a) Difference of free energies F between PM and AIAO magnetic states for $R_2\text{Ir}_2\text{O}_7$. Solid circles (hollow squares) denote the values obtained using the LDA+DMFT (LDA+U) method, plotted as zero when the AIAO state cannot be stabilized. Inset displays the LDA band-width of the t_{2g} bands; shaded region highlights the insulating compounds. (b) Quasiparticle renormalization amplitude Z_α (see text for the definition) for the $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ states. Solid (dashed) lines denote the values in the PM (AIAO) states, with Z for the $J_{\text{eff}} = 1/2$ orbital in the PM state of Sr_2IrO_4 (red triangle) shown for comparison. (c) Ordered moment of Ir atoms. All LDA+DMFT calculations are done at 50K.

ground state for $\text{Nd}_2\text{Ir}_2\text{O}_7$. More accurate calculations at lower temperature will be required to resolve this issue. We note in passing that the difference in electronic entropy S between the two phases is very small. For example, in $\text{Nd}_2\text{Ir}_2\text{O}_7$ we obtained $\Delta S = 0.033 k_B$ per Ir atom, which is similar to the experimentally determined $\Delta S = 0.028 k_B$ ¹⁷ (see the Supplement).

To shed more light on the origin of the MIT, Fig. 1(b)

shows the DMFT-predicted quasiparticle renormalization amplitude $Z_\alpha = (1 - d\text{Im}\Sigma_{\alpha\alpha}/d\omega_n)^{-1}|_{\omega_n=0}$ in the paramagnetic metallic state, where α labels a correlated orbital and Σ indicates the self-energy on the Matsubara-frequency (ω_n) axis. The less active $J_{\text{eff}} = 3/2$ states are only weakly renormalized. Turning to the more active $J_{\text{eff}} = 1/2$ states, these are renormalized by a factor of almost ten in the PM state for compounds having an insulating ground state. By contrast, the $J_{\text{eff}} = 1/2$ quasiparticles in $\text{Bi}_2\text{Ir}_2\text{O}_7$ remain quite light, consistent with recent ARPES measurement.⁴⁰ Surprisingly, the $J_{\text{eff}} = 1/2$ orbital in $\text{Pr}_2\text{Ir}_2\text{O}_7$ is quite heavy (about five times the band mass), indicating that the correlations are considerably stronger than in the marginal Mott insulator Sr_2IrO_4 , whose Z_α is plotted as the red triangle in Fig. 1(b). In this sense, the frustration in the pyrochlore lattice plays an essential role in shifting the MIT boundary towards stronger correlations compared to unfrustrated RP iridates. The frustration thus promotes metallicity by penalizing competing long-range magnetic order, an effect which is well captured by DMFT but less adequately by LDA+U, explaining why LDA+U predicts the Pr compound to be insulating.

In short, the boundary of the MIT across the $R_2\text{Ir}_2\text{O}_7$ compounds is determined by a competition between the formation of quasiparticle bands via the Kondo effect in the PM state, and the tendency toward long-range magnetic order. In general, the occurrence of a MIT depends crucially on many factors, including dimensionality and frustration. For example, in Sr_2IrO_4 the two-dimensionality reduces the Néel ordering temperature, but the short-range order above the Néel temperature still preserves the charge gap in the excitation spectrum. Frustration also reduces the tendency to long-range order, but it promotes metallicity and allows very narrow quasiparticles to be observed in the PM state before the long-range order opens the charge gap. This is why $\text{Pr}_2\text{Ir}_2\text{O}_7$ remains in the paramagnetic bad-metal phase even though the correlation strength is much larger than in Sr_2IrO_4 .

For correlated Fermi liquids, the quantum and thermal fluctuations give rise to fluctuating moments in the PM state, which statically order when the magnetic energy gain is sufficient to overcome the gain available from band formation. According to Fig. 1(c), the magnitude of the ordered magnetic moment for $\text{Y}_2\text{Ir}_2\text{O}_7$ is about $0.38 \mu_B$, consistent with the experimental upper bound of $0.5 \mu_B$ obtained by neutron scattering.¹⁹ We note that spatial fluctuations, which are not fully accounted for in LDA+DMFT, are likely to further reduce the ordered magnetic moment. In the AIAO insulating state, there are no states at the Fermi energy, but the slope of the self-energy at zero frequency still gives some measure. As shown by the dashed lines in Fig. 1(b), the quantum renormalization effects are greatly reduced in the AIAO state as compared to the PM state.

We observed that, like the effective $J_{\text{eff}} = 1/2$ states in the RP iridates,²³ the $J_{\text{eff}} = 1/2$ wavefunction in the

pyrochlore iridates also strongly deviates from the rotationally invariant $SU(2)$ point. This is mostly due to the trigonal crystal field induced by compression of the IrO_6 octahedra. The trigonal crystal-field splitting in our calculations is derived from a very large energy window, and is on average about 0.39 eV (see the Supplement), in good agreement with resonant X-ray scattering measurements.⁴² Correspondingly, the resulting orbital-to-spin moment ratio is about 1.3 (see the Supplement), showing a significant deviation from the $SU(2)$ limit of two.²³ Nevertheless, the effective $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ bands are still well separated in energy, which facilitates our analysis of the topological character. Note that the values of local U and J parameters on Ir atoms in our LDA+DMFT calculations are the same for the pyrochlore and RP iridates,²³ confirming the more universal nature of local Coulomb repulsion across similar materials when the screening by hybridization effects at high energy are allowed in the calculation.

We now turn to the topological properties of the insulating pyrochlores. Fig. 2(a) shows a fictitious non-magnetic band structure of $\text{Nd}_2\text{Ir}_2\text{O}_7$, together with the irreducible representations for the $J_{\text{eff}} = 1/2$ states around E_F at the eight time-reversal-invariant momenta (TRIM). This band structure is obtained by neglecting the dynamic part of the LDA+DMFT self-energy, but using the self-consistent LDA+DMFT charge density, with time-reversal (TR) symmetry imposed. We note that these bands are quite similar to the LDA bands.

Regarding the irreducible representations, the space group of the pyrochlore iridates is nonsymmorphic. Thus, depending on the choice of inversion center, the four energetically equivalent L points are separated into one L' and three L points, with opposite parities for the L' and L points as shown in Fig. 2(a). Moreover, at each of the three X points all states are four-fold degenerate by symmetry, with each degenerate group comprising two even-parity and two odd-parity states. Finally, all eight $J_{\text{eff}} = 1/2$ bands at the Γ point have even parity. The situation is summarized in the PM column of Table I. It follows that if a global band gap could be opened so as to extend the shaded region of Fig. 2(a) throughout the Brillouin zone, corresponding to a half-filled $J_{\text{eff}} = 1/2$ manifold, seven out of 16 of the occupied Kramers pairs at the eight TRIM would be odd-parity ones. Since this number is odd, it would generate a strong-topological-insulator phase.⁴³ It has been suggested this might be achieved for $\text{Pr}_2\text{Ir}_2\text{O}_7$, for example, by applying a strain along the (111) direction.⁴⁴ If the TR symmetry would be weakly broken in this phase, the system would become an axion rather than a strong topological insulator, but we shall use the term “topological insulator” to cover both cases.

When the DMFT self-energy is taken into account, the TR symmetry is spontaneously broken and a global gap opens up. However, the topological character may be different from that described above, because of the drastic mixing of bands that results. Following the theory of in-

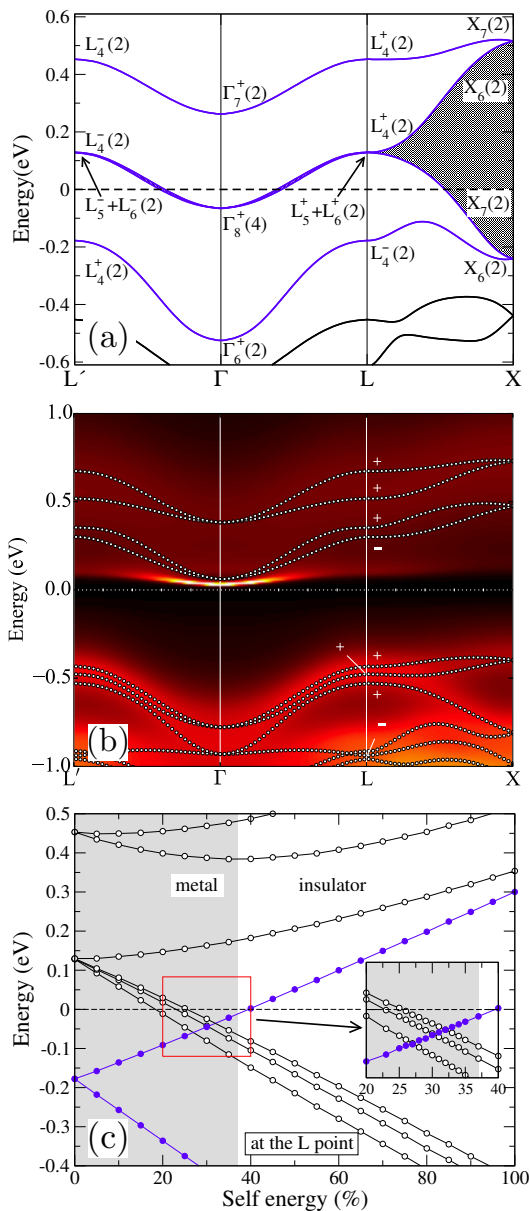


FIG. 2: (Color online) (a) The LDA-like band structure of $\text{Nd}_2\text{Ir}_2\text{O}_7$ with irreducible representations indicated at the time-reversal invariant momenta; numbers in parenthesis denote degeneracies. (b) Color map: spectral function of $\text{Nd}_2\text{Ir}_2\text{O}_7$ in the AIAO magnetic state. Dotted lines: band structure of the effective topological Hamiltonian (see text for details). The parities of eight $J_{\text{eff}}=1/2$ -derived bands are indicated by “+” or “-” at the L point. (c) Evolution of the eigenvalues of the effective topological Hamiltonian at L with mixing fraction of the DMFT self-energy. Inset zooms in on the critical region indicated by the red square.

interacting topological phases,^{45,46} the topological indices can be obtained by inspecting an effective single-particle Hamiltonian defined as $H_{\text{eff}}=H_0+\Sigma(\omega=0)$, where H_0 is the TR-invariant Bloch Hamiltonian and $\Sigma(\omega=0)$ is the DMFT self-energy at zero frequency (i.e., at the Fermi energy), which carries all the TR-breaking terms. The

TABLE I: Parity analysis of eight eigenstates around E_F in the PM and AIAO magnetic states, corresponding to Figs. 2(a-b) respectively. ‘MULT’ denotes the multiplicity in accounting for the eight time-reversal invariant momenta. The parities are given in order of increasing energy eigenvalues, with vertical bars indicating E_F .

	MULT	PM	AIAO
Γ	1	{+ + + +}	{+ + + + + + + +}
L	3	{- - + + + + + +}	{- + + + - + + +}
L'	1	{+ + - - - - - -}	{+ - - - + - - -}
X	3	{+ + - - + + - -}	{+ + - - + + - -}

resulting band structure is shown in Fig. 2(b), together with the full LDA+DMFT spectral function. Since TR symmetry is broken, the total parity of all occupied eigenstates at the TRIM should now be counted;⁴⁶ the system is a trivial or axion insulator if the total number is $4n$ or $4n+2$ respectively, with n an integer.

The parities of the $J_{\text{eff}}=1/2$ -derived eigenstates at the eight TRIM are also shown for the AIAO state in Table I. At the Γ point all states remain even-parity even with the addition of the self-energy. At the X points, each four-fold degenerate state splits into two doublets, and we find (see the Supplement) that including $\Sigma(\omega=0)$ does not induce any band inversion or otherwise change the ordering of parities, so that the number of occupied odd-parity states remains equal to two at each X point. The most significant change occurs at the L (L') points, where the TR-symmetry breaking splits each doubly-degenerate L_4^- (L_4^+) state into two singlets. Some of these cross the gap as the self-energy is turned on, as shown in Fig. 2(c), with the result that there is an exchange of parity between occupied and unoccupied states as shown in Table I. As a result, there are a total of twelve odd-parity occupied states at the TRIM. Similar behavior is also observed in the other insulating $R_2\text{Ir}_2\text{O}_7$ compounds studied. Thus, after properly including the DMFT self-energy, we find that the insulating $R_2\text{Ir}_2\text{O}_7$ compounds are topologically trivial insulators. This is in agreement with previous LDA+U results¹¹ in the large- U limit.

Nevertheless, in Refs. 11 and 13 it was argued that non-trivial axion insulator and Weyl semimetal phases occur with smaller values of on-site U . This is not supported by our DMFT calculations. For instance, for $\text{Y}_2\text{Ir}_2\text{O}_7$, which is the most insulating of the six compounds, the PM solution is more stable than the AIAO magnetic state when the on-site U value is reduced to 4.0 eV (see the Supplement). Moreover, as shown in Fig. 2(c), we observe that the parity exchange between occupied and unoccupied states at the L and L' points happens before a global band gap is opened. Therefore, we think it is very unlikely that a topological insulating phase can be found in the bulk pyrochlore iridates.

In summary, our calculations provide a clear picture of the origin of the MIT in pyrochlore iridates and its variation with respect to the A-cation radius. Moreover, our parity analysis of the many-body effective $J_{\text{eff}}=1/2$ wave

functions reveals that when insulating magnetic phases appear in these pyrochlore iridates, they are very likely to remain topologically trivial.

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Note: As we were finalizing this manuscript a recent paper appeared⁴⁷ which presents ARPES measurements for $\text{Nd}_2\text{Ir}_2\text{O}_7$, showing that there is no surface states of topological origin for the system in the all-in-all-out magnetic state. This is consistent with our conclusion.

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