Theory of metallic double perovskites with spin orbit coupling and strong correlations; application to ferrimagnetic Ba$_2$FeReO$_6$

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We consider a model of the double perovskite Ba$_2$FeReO$_6$, a room temperature ferrimagnet with correlated and spin-orbit coupled Re $t_{2g}$ electrons moving in the background of Fe moments stabilized by Hund’s coupling. We show that for such 3d/5d double perovskites, strong correlations on the 5d-element (Re) are essential in driving a half-metallic ground state. Incorporating both strong spin-orbit coupling and the Hubbard repulsion on Re leads to a band structure consistent with ab initio calculations. Using our model, we find a large spin polarization at the Fermi level, and obtain a semi-quantitative understanding of the saturation magnetization of Ba$_2$FeReO$_6$, as well as X-ray magnetic circular dichroism data indicating a significant orbital magnetization. Based on the orbital populations obtained in our theory, we predict a specific doping dependence to the tetragonal distortion accompanying ferrimagnetic order. Finally, the combination of a net magnetization and spin-orbit interactions is shown to induce Weyl nodes in the band structure, and we predict a significant intrinsic anomalous Hall effect in hole-doped Ba$_2$FeReO$_6$. The uncovered interplay of strong correlations and spin-orbit coupling lends partial support to our previous work, which used a local moment description to capture the spin wave dispersion found in neutron scattering measurements. Our work is of interest in the broader context of understanding metallic double perovskites which are of fundamental importance and of possible relevance to spintronic applications.

I. INTRODUCTION

Double perovskite (DP) materials $A_2BB'O_6$, where the transition metal ions B and B' reside on the two sublattices of a cubic lattice, can realize many complex phases. Metallic variants, such as Sr$_2$FeMoO$_6$, provide us with the simplest multi-orbital examples of ferrimagnetic order, kinetically stabilized by the Pauli exclusion principle. Insulating variants where only the B'-site ion is magnetic, such as Ba$_2$YMoO$_6$ and La$_2$LiMoO$_6$, provide material examples of quantum mechanical moments living on the geometrically frustrated face-centered cubic lattice. Metallic DPs, such as Sr$_2$FeMoO$_6$, are also of significant technological importance, being room temperature ferrimagnets with half-metallic band structures and a large spin polarization which is useful for spintronic applications.

Metallic 3d/5d DPs are of particular interest in this regard since they appear to have strongly reduced B/B' site mixing; samples of Ba$_2$FeReO$_6$ studied in previous work have low < 1% anti-site disorder. Such anti-site disorder, which is common in other DPs and which is detrimental to spintronic applications, appears to be alleviated in 3d/5d DPs by the B/B' ionic size mismatch suggesting that they might be better suited for applications. However, such 3d/5d DPs require us to confront the twin aspects of strong correlations and strong spin-orbit coupling, topics at the forefront of fundamental research motivated by the possibility of stabilizing states such as fractionalized topological insulators (TIs) or Weyl semimetals.

In this paper, we focus on metallic ordered DPs with mixed 3d/5d transition metal ions on the B/B' sites, specifically the Ba$_2$FeReO$_6$ material with the structure as shown in Fig. 1. We obtain the following main results. (i) We consider a model of the ordered double perovskite Ba$_2$FeReO$_6$ (see Fig. 1) retaining the relevant electronic states in the vicinity of the Fermi level. This model, after taking spin-orbit coupling as well as correlations effects into account within a self-consistent mean field theory, is shown to reproduce previous ab initio electronic structure results in the ferrimagnetic ground state. Our model accounts for the dominant energy scales in this material: (a) the strong Hund’s coupling on Fe, the Hubbard repulsion on Re, and the Fe-Re charge transfer energy (all on the scale of ∼ 1eV), (b) the strong spin-orbit coupling on Re (∼0.5eV), and (c) the nearest neighbor Re-Fe hopping terms which leads to...
electron itinerancy ($\sim 0.3$eV). In addition, we include weaker terms such as inter-orbital mixing and second neighbor hopping which are required to reproduce the band degeneracies at high symmetry points in the Brillouin zone found in earlier ab initio studies. (ii) Our theory accounts semi-quantitatively for the measured saturation magnetization, as well as X-ray magnetic circular dichroism (XMCD) experiments which find a significant orbital contribution to the Fe magnetization in the ordered state. (iii) Based on the orbital occupations in the magnetically ordered state, we predict a tetragonal distortion, with c-axis compression accompanying magnetic order, in agreement with experimental data. We also predict a specific doping dependence to this orbital order and distortion which could be tested in future experiments. (iv) The strong correlations on Re, inferred from our study, lends partial support to earlier work which showed that a local moment description of the ferrimagnetic state provides a reasonably good description of the magnetic dynamic structure factor obtained using inelastic neutron scattering experiments. This importance of strong correlation effects and local moment physics on the 5d element is in agreement with previous ab initio studies that discussed the emergence of local moments of closely related Cr-based 3d/5d DPs upon progressing through the series with B'=W,Re,Os. (v) From our computed band dispersion, we show the appearance of Weyl nodes in such metallic phases. (vi) Using the Kubo formula for the spin-orbit coupled bands, we find that Ba$_2$FeReO$_6$ itself appears to have only a small intrinsic anomalous Hall effect (AHE) in the ordered ferrimagnetic state at low temperature, but the AHE is significant in hole doped systems, and we speculate that it might also be significant at intermediate temperatures below the ferrimagnetic $T_c$ in Ba$_2$FeReO$_6$. Taking a broader viewpoint, Re-based layered quasi-two-dimensional oxides or heterostructures may be more strongly correlated than the three-dimensional DPs, and may lead to interesting Mott physics beyond the itinerates due to the local competition between interactions and spin-orbit coupling due to the d$^2$ configuration of Re$^{5+}$. Furthermore, one can carry out detailed inelastic neutron scattering studies in Re-based oxides, thus allowing for the possibility to explore the magnetism in more detail than in the itinerates. This may prove to be useful in future studies of exotic variants of Re-based oxides.

II. MODEL

The simple charge counting for Ba$_2$FeReO$_6$ suggests Re$^{5+}$ and Fe$^{3+}$ valence states on the transition metal ions. In this state, the five 3d-electrons on Fe are expected to be locked into a spin-5/2 moment due to strong Hund’s coupling in the half-filled d-shell. Here, we will treat this magnetic moment as a classical vector. The two 5d-electrons in the Re $t_{2g}$ orbital are mobile, able to hop on and off the Fe sites subject to a charge transfer energy $\Delta = \Delta_F - \Delta_R > 0$, and Pauli exclusion which constrains electrons arriving on Fe to be antiparallel to the direction of the local Fe moment. For a general direction of the Fe moment, $\vec{F} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ at a given site, we must project the added electrons onto the allowed direction to satisfy Pauli exclusion, locally setting $f_\uparrow = \sin \frac{\theta}{2} e^{-i\phi/2} f$ and $f_\downarrow = -\cos \frac{\theta}{2} e^{i\phi/2} f$, effectively “stripping” the electron of its spin degree of freedom. Such models have been proposed for other DP materials, and shown to capture the phenomenology of Sr$_2$FeMoO$_6$ including thermal phase transitions and disorder effects. However, most of these previous studies, with the notable exception of Ref. 11 have ignored spin-orbit coupling effects, which are expected to be extremely important for 5d transition metal oxides.

Our model does not explicitly account for additional superexchange interactions between the Fe local moment and the emerging local moments on the Re sites which is explicitly taken into account as a separate term in some previous studies (for example, Ref. 11); however, we think such terms should emerge more naturally from an effective tight-binding model when strong correlations are incorporated, as might be relevant to Mott insulating oxides like Sr$_2$CrOsO$_6$. Fe-Fe superexchange terms which we omit, since they are not necessary to drive the ferrimagnetic state observed in Ba$_2$FeReO$_6$, may prove to be important in understanding the complete magnetic phase diagram as a function of doping which is not addressed in this paper. However, they are likely to be small given the Fe-Fe separation in the DP structure. Further differences between the results of Ref. 11 and our work stem from the fact that their model is for d$^3$ configuration of Cr, as opposed to our d$^5$ state on Fe; while both spin components of the itinerant electrons are permitted on Cr (since the $e_g$ orbital is available), only one spin projection is allowed for itinerant electrons on Fe due to the Pauli exclusion.

A. Non-interacting tight binding model

The model describing Re electrons moving in the presence of Fe moments then takes the form $H_0 = H_{hop} + H_{so} + H_{ct}$. Here, the Hamiltonian $H_{hop}$ describes intra-orbital hopping of electrons on the lattice, from Re to Fe (nearest-neighbor) and from Re to Re (next-neighbor), as well as inter-orbital hopping of electrons between next-neighbor Re sites: $H_{so}$ is the atomic spin-orbit coupling on Re, projected to the $t_{2g}$ manifold, of strength $\lambda$; finally, $H_{ct}$ describes the charge transfer energy offset $\Delta$ between Re and Fe sites. For simplicity, we only focus on the case of a uniform magnetization on the Fe site, assuming $(\theta, \phi)$ which describe the Fe moment to be site-independent; it is straightforward to generalize
our work to a nonuniform spatially varying magnetization. We use the simple triclinic unit cell, with one Re and one Fe atom, as shown in Fig. 1 to study the model Hamiltonian; however in order to facilitate a comparison with published *ab initio* electronic structure calculations, we will later assume a body-centered tetragonal unit cell containing two Re and two Fe atoms, with lattice constants $d_\alpha = d_\beta = d_\gamma / \sqrt{2}$ as shown in Fig. 1, and use orthorhombic notation to plot the band dispersion of the eighteen bands in the Brillouin zone.

We label the electrons on the Fe and Re sites by $\sigma$ and $\tau$, respectively, with $\alpha = \uparrow, \downarrow$ being the spin. The Hamiltonian takes the following form in momentum space, where we assume implicit summation over repeated spin and orbital indices,

$$
H_{\text{hop}} = \sum_\mathbf{k} (\eta_\ell (\mathbf{k}) g_\sigma (\theta, \phi) d_{\ell \sigma} (\mathbf{k}) f_\ell (\mathbf{k}) + \text{h.c.})
+ \sum_\mathbf{k} \epsilon_\ell (\mathbf{k}) (d_{\ell \sigma} (\mathbf{k}) d_{\ell \sigma} (\mathbf{k}) + \alpha_f f_\ell (\mathbf{k}) f_\ell (\mathbf{k}))
+ \sum_{\mathbf{k}(\ell \neq \ell')} \epsilon_{\ell \ell'} (\mathbf{k}) (d_{\ell \sigma} (\mathbf{k}) d_{\ell' \sigma} (\mathbf{k}) + \alpha_f f_\ell (\mathbf{k}) f_{\ell'} (\mathbf{k}))
\tag{1}
$$

$$
H_{\text{so}} = \frac{1}{2} \sum_\mathbf{k} \epsilon_{\ell \ell'} (\mathbf{k}) \tau_{\ell \ell'}^{\sigma \sigma'} d_{\ell \sigma} (\mathbf{k}) d_{\ell' \sigma'} (\mathbf{k})
\tag{2}
$$

$$
H_{\text{ct}} = \Delta \sum_\mathbf{k} f_\ell (\mathbf{k}) f_\ell (\mathbf{k})
\tag{3}
$$

Here, in light of our previous discussion, we have only retained a single spin projection on the Fe site, with $g_\ell (\theta, \phi) = \sin \frac{\theta}{2} e^{-i \phi / 2}$ and $g_\ell (\theta, \phi) = -\cos \frac{\theta}{2} e^{i \phi / 2}$. The various hopping processes are schematically illustrated in Fig. 2. The first term in $H_{\text{hop}}$ describes nearest-neighbor intra-orbital hopping from Re to Fe, parameterized by $t_\pi, t_\delta$. The next two terms in $H_{\text{hop}}$ characterize next-nearest hopping processes, with the ratio of Fe-Fe hopings to Re-Re hoppings being $\alpha_f$; we will fix $\alpha_f = 0.5$. While the second term captures intra-orbital hopping between close pairs of Re atoms or Fe atoms (parameterized by $t', t''$), the third term captures inter-orbital hopping between close pairs of Re or Fe atoms (parameterized by $t_n$). Many of these hopping processes ($t_8, t_m, t_9'$) have a small energy scale; however they are important to reproduce the band degeneracies found in *ab initio* calculations at high symmetry points in the Brillouin zone. The explicit momentum dependence of the dispersion coefficients appearing in $H_{\text{hop}}$ is given in Appendix A.

### B. Interaction effects

Electron-electron interactions are partially accounted for by $H_0$ in the previous section — in part, by the charge transfer gap $\Delta$, and, in part, by the implicit Hund’s coupling which locks the Fe electrons into a high-spin state. However, electronic interactions on Re have been omitted in $H_0$. We now include these local Hubbard interactions on Re. The interaction Hamiltonian in the $\tau_2g$ orbitals of Re takes the form

$$
H_{\text{int}} = U \sum_{\ell \ell'} n_{\ell \uparrow} n_{\ell' \uparrow} + (U - 5J_H / 2) \sum_{\ell \neq \ell'} n_{\ell \uparrow} n_{\ell' \downarrow}
- 2J_H \sum_{\ell \neq \ell'} \vec{S}_{\ell \uparrow} \cdot \vec{S}_{\ell' \downarrow} + J_H \sum_{\ell \neq \ell'} (d^\dagger_{\ell \uparrow} d_{\ell' \downarrow} d_{\ell' \uparrow} d_{\ell \downarrow})
\tag{4}
$$

where $i$ labels the Re sites, and $\vec{S}_{i \ell} = \frac{1}{2} \sum_{\alpha \beta} \vec{\sigma}_{\alpha \beta} d_{i \ell \alpha} d_{i \ell \beta}$ is the spin at site $i$ in orbital $\ell$. We wish to then study the full Hamiltonian $H = H_0 + H_{\text{int}}$. For simplicity, we only retain only the dominant intra-orbital Coulomb repulsion, treating it at mean field (Hartree) level, as

$$
H_{\text{int}} \approx U \sum_{\ell \ell'} \left[ \rho_{\ell \ell'} (n_{\ell \uparrow} + n_{\ell \downarrow}) - 2m_\ell \cdot \vec{S}_{i \ell} - \rho_{\ell \ell'} \vec{S}_{i \ell} \cdot \vec{S}_{i \ell} / 4 + \vec{m}_\ell \cdot \vec{m}_\ell \right]
\tag{5}
$$

where $\rho_\ell = \langle n_{\ell \uparrow} + n_{\ell \downarrow} \rangle$, $\vec{m}_\ell = \langle \vec{S}_{i \ell} \rangle$, and we set $\vec{m}_\ell = -m_\ell (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$, with $m_\ell > 0$, so that $\vec{m}_\ell$ is anti-parallel to the Fe moment $\vec{F}$. For simplicity, we only focus on the case $\theta = \phi = 0$, so the Fe sites can only accommodate itinerant spin-$\downarrow$ electrons. We then numerically determine $m_\ell$ and $\rho_\ell$ in a self-consistent fashion, using the non-interacting ground state as the starting point for the iterative solution, while ensuring...
that the choice of the chemical potential lead to a total of two electrons per unit cell (i.e., per Re atom). Such a mean field treatment of electron-electron interactions does not capture all aspects of the strong correlation physics, e.g. bandwidth renormalization and mass enhancement. Nevertheless, recognizing this caveat, we use the self-consistent solution of the mean field equations to study the effects of interactions and spin-orbit coupling on the reorganization of the nine electronic bands, compare the physical properties with experimental results, and make qualitative predictions for future experiments.

III. PHYSICAL PROPERTIES

We begin by discussing the effect of electronic correlations in the DPs in the absence of spin-orbit coupling. We show that such correlation effects appear to be crucial to stabilize a half-metallic state with complete polarization in the 5d perovskites, due to the large second-neighbor Re-Re hopping which otherwise prevents a half-metallic state. We then turn to the effect of spin-orbit coupling, and show that it reorganizes the band structure, yielding results which are in reasonable agreement with previous \textit{ab initio} electronic structure studies\textsuperscript{31} (As pointed out earlier, the band dispersions discussed below are plotted using the orthorhombic notation with an enlarged unit cell containing two Fe and two Re atoms, leading to eighteen electronic bands instead of nine.) Finally, we compare the mean field result for the saturation magnetization with experiments, and the spin and orbital magnetization on the Re site with previous XMCD data, and discuss other physical properties such as tetragonal lattice distortion and predictions for the AHE. Throughout this discussion, we will assume a ferromagnetic order of the Fe moments - a more complete study of the magnetic phase diagram as a function of doping and temperature will be the subject of future numerical investigations.

A. Correlations stabilize a half-metal

If we ignore Re correlations entirely, setting \( U = 0 \), and also ignore spin-orbit coupling by setting \( \lambda = 0 \), the band structure shown in Fig.\textsuperscript{3}(a) has decoupled spin-\( \uparrow \) and spin-\( \downarrow \) bands. The twelve spin-\( \downarrow \) bands correspond to electrons which can delocalize on Re and Fe. By contrast, the six spin-\( \uparrow \) bands correspond to electrons which cannot delocalize on Re and Fe. To make a reasonable comparison with the \textit{ab initio} calculations, we have to assume a significant \( t' = 0.3 \) (Re-Re hopping), but all other hoppings can be assumed to be small; for simplicity, we fix \( t_\| = t'' = t_m = 0.1 \). Finally, we have to assume a moderate charge transfer energy \( \Delta = 3 \) which splits the spin-\( \downarrow \) states into two groups: 6 lower energy Re-Fe hybridized spin-\( \downarrow \) states (dominant Re character) which form a \textit{broad} band, and 6 higher energy dominantly Re-Fe hybridized spin-\( \downarrow \) states (dominant Fe character) which form a \textit{narrow} band. Finally, the remaining 6 Re-\( \uparrow \) states form a \textit{narrow} dispersing band, crossing the chemical potential and overlapping in energy with the broad spin-\( \downarrow \) band. For \( U = 0 \), the system thus contains both spin states at the Fermi level. When we incorporate a Hubbard repulsion \( U = 8t_\pi \) at mean field level, we see from Fig.\textsuperscript{3}(b) that its main effect is to self-consistently shift the spin-\( \downarrow \) bands higher in energy, leaving only spin-\( \downarrow \) states at the Fermi level. The resulting band dispersion is in reasonably good agreement with LDA+U calculations. Although we have not attempted a detailed quantitative fitting to the LDA+U band structure, the features noted below are robust. (i) A rough comparison with the overall bandwidth in the \textit{ab initio} calculations without spin-orbit coupling\textsuperscript{31} suggests that \( t_\pi \approx 330\text{meV} \). This is somewhat larger than estimates for Sr\textsubscript{2}FeMoO\textsubscript{6} in the literature\textsuperscript{31,10} (\textasciitilde 270 meV). (ii) We estimate the interaction energy scale on Re to be \( U \approx 2.5\text{eV} \), smaller by a factor of two compared with typical values for 3d transition metals. (iii) There is a significant Re-Re hopping, \( t'/t_\pi \approx 0.3 \), we need to include in

![FIG. 3: Band dispersion in the orthorhombic notation for the Re and Fe electronic states for different choices of Hubbard interaction \( U \) and spin-orbit coupling \( \lambda \), with energy on the \( y \)-axis in units of \( t_\pi \). The solid black line indicates the chemical potential. For no spin-orbit coupling, (a) \( U = 0 \) and \( \lambda = 0 \) and (b) \( U = 8t_\pi \) and \( \lambda = 0 \), we find decoupled spin-\( \downarrow \) (red, solid) and spin-\( \uparrow \) (blue, dashed) states. Comparing (a) and (b), we see that correlations on Re push the spin-\( \uparrow \) states to higher energy, leading to the stabilization of a half-metal ground state. A nonzero spin-orbit coupling, (c) \( U = 0 \) and \( \lambda = 2t_\pi \), and (d) \( U = 8t_\pi \) and \( \lambda = 2t_\pi \), leads to mixed-spin states and splits degeneracies, but for a physically reasonable value \( U = 8t_\pi \) preserves significant spin polarization \( \sim 90\% \) for states at the Fermi level.](image-url)
order to be able to capture the bandwidths of the spin-\(\uparrow\) and spin-\(\downarrow\) bands. All these observations are reasonable given the more extended nature of Re orbitals when compared with 3d or 4d transition metal ions. The presence of appreciable Re-Re hoppings has been pointed out in previous work\(^{[1-11]}\), although they did not take correlation effects on Re into account. More recent work has also arrived at similar conclusions regarding significant Re-Re hoppings\(^{[12]}\).

To summarize, we have obtained a tight-binding description including interactions of DPs with spin-orbit coupling. In contrast to 3d/4d DP materials like Sr\(_2\)FeMoO\(_6\), we find that 3d/5d DPs have a significant second neighbor hopping; strong correlations on the 5d element (Re) therefore play a crucial role in stabilizing a half-metallic ground state in the 3d/5d DPs.

**B. Spin-orbit coupling: Band reconstruction, spin/orbital magnetization, and comparison with magnetization and XMCD experiments**

We next turn to the effect of incorporating both spin-orbit coupling and Hubbard interactions on Re, solving the mean field equations in case of a nonzero \(U\). From Fig.3(c) and (d), where we have set \(\lambda = 2t_\pi\) (~660meV for our estimated \(t_\pi\)), we see that spin-orbit coupling clearly eliminates the degeneracies occurring at the \(\Gamma\)-point for \(\lambda = 0\). It also significantly reconstructs the dispersion of the eighteen bands, leading to reasonably good agreement with published \textit{ab initio} calculations which include spin-orbit coupling\(^{[11]}\). In the next section, we will discuss the resulting appearance of Weyl nodes in the band dispersion and the intrinsic anomalous Hall effect including interactions of DPs with spin-orbit coupling. In contrast to 3d/4d DP materials like Sr\(_2\)FeMoO\(_6\), we have derived at similar conclusions regarding significant Re-Re effects on Re into account. More recent work has also arrived at similar conclusions regarding significant Re-Re hoppings\(^{[12]}\).

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**C. Orbital order, tetragonal distortion in ferrimagnetic state, and doping dependence**

In the converged mean field state, with the magnetization along the \(z\)-axis, we find that the density on Re in the three orbitals are different, with \(\rho_{xy} \approx 0.60\) and \(\rho_{xz} = \rho_{yz} \approx 0.53\). This orbital imbalance is induced in the \(z\)-ferrimagnetic state due the spin-orbit coupling. The larger extent of the \(xy\)-orbital in the \(xy\)-plane, compared with its smaller extent along the \(z\)-direction, implies that this orbital charge imbalance would lead to a tetragonal distortion of the lattice, to occur coincident with ferrimagnetic ordering and with a shrinking of the \(c\)-axis, as has indeed been observed to occur experimentally. The precise extent of this distortion, which is observed\(^{[33]}\) to be \(~ 0.1\%\), depends on details such as the lattice stiffness, and is beyond the scope of our calculation.

When we solve the self-consistent equations at various dopings \(\delta\) (excess electrons per Re) assuming persistent ferrimagnetic order, the extent of this orbital imbalance, characterized by a tetragonal order parameter \(\eta_{\text{net}} = \frac{1}{\rho} (\rho_{xy} - \rho_{xz}/2 - \rho_{yz}/2)\), changes systematically as shown in Fig.3(a). Light electron doping leads to a slightly larger orbital population imbalance and should enhance the \(c\)-axis compression, while a larger electron doping leads to a gradual decrease of \(\eta_{\text{net}}\). Hole doping beyond \(\gtrsim 0.25\) holes/Re leads to \(\eta_{\text{net}} < 0\), which should cause elongation along the \(c\)-axis. The spin contribution to the magnetization on Re, arising from the different
orbitals, also shows a similar doping trend as seen from Fig. 4(b), while the orbital contribution to the magnetization on Re has the largest magnitude at zero doping. These results could be possibly be explored experimentally by partially substituting Ba by trivalent La (electron doping), or by Cs or other monovalent ions (hole doping).

Thus, in 3d/5d DP materials, spin orbit coupling and the ferrimagnetic order of itinerant electrons leads to orbital ordering. This, in turn, should lead to a compression along the c-axis, consistent with the experimentally observed tetragonal distortion, and we predict a specific doping dependence to this structural distortion.

D. Doping-dependent anomalous Hall effect

We next turn to the intrinsic AHE in the ferrimagnetic state of such 3d/5d DPs. As pointed out in recent work, for pyrochlore iridates with all-in-all-out order under uniaxial pressure, as well the ferrimagnetic infinite-layer ruthenate SrRuO$_3$, this intrinsic AHE contains two contributions: (i) a surface contribution arising from Fermi arc states associated with Weyl nodes in the dispersion, and (ii) a bulk contribution from carriers near the Fermi surface. A pair of such Weyl nodes for Ba$_2$FeReO$_6$ is shown in Fig. 5 obtained from the interacting band dispersion.

Both contributions to the intrinsic AHE are captured by the momentum-dependent Berry curvature of the spin-orbit coupled bands, which is, in turn, obtained from the Kubo formula

$$\sigma_{xy} = e^2\hbar \int \frac{d^3k}{(2\pi)^3} \sum_{m \neq n} \frac{f(\epsilon_{km}) - f(\epsilon_{kn})}{(\epsilon_{km} - \epsilon_{kn})^2} \text{Im}(v_{mn}^x v_{nm}^y). (6)$$

Here, $\epsilon_{km}$ is the single-particle energy at momentum $k$ and band $m$, $v^\alpha = \frac{1}{i}(\partial H_{mf}/\partial k_\alpha)$ are components of the velocity operator with $H_{mf}$ being the self-consistently determined mean-field Hamiltonian matrix, and $f(.)$ is the Fermi function. From the mean field solution corresponding to two electrons per Re, as appropriate for Ba$_2$FeReO$_6$, we find that $\sigma_{xy}$ at zero temperature is small, $\sigma_{xy} \sim 0.2 \times 10^{-3} \text{cm}^2/\text{A} \cdot \text{m}$, where $d_c \approx 8 \text{A}$ is the lattice constant in Fig. 1. This translates into $\sigma_{xy} \sim 8 \times 10^{-1} \text{cm}^{-1}$.

In order to explore $\sigma_{xy}$ over a larger space of parameters, we consider its variation with doping. Rather than simply shifting the chemical potential, we solve the Hartree mean field equations over a range of electron densities, and then compute $\sigma_{xy}$ in the resulting self-consistent band structure. We find that electron doping does not significantly enhance the AHE, but a hole doping of about 0.5-0.8 holes/Re leads to a larger AHE $\sigma_{xy} \sim 10^{-1} \text{cm}^{-1}$ to $\sim 25 \times 10^{-1} \text{cm}^{-1}$. Even this significant AHE is small in natural units ($\sim 0.1 \times 10^{-3} \text{cm}^2/\text{A} \cdot \text{m}$) at $T = 0$, which we attribute to the large spin polarization in the completely ordered ferromagnet. It is possible that the AHE is a non-monotonic function of temperature, peaked at some intermediate temperature below the magnetic $T_c$ even in the undoped compound.

Thus, in 3d/5d DP materials, spin orbit coupling and the ferrimagnetic order is expected to lead to an intrins-ic AHE. The AHE appears likely to be larger for hole doped systems compared to an expected small value for Ba$_2$FeReO$_6$ and is likely, in Ba$_2$FeReO$_6$, to be peaked at intermediate temperatures below $T_c$.

IV. CONCLUSION

We have obtained a tight-binding description of the metallic DPs, including spin-orbit coupling and strong correlation effects. Although we have here only applied it to Ba$_2$FeReO$_6$, finding good agreement with a broad variety of experiments and with electronic structure calculations, our work should be broadly applicable to other 3d/4d and 3d/5d DP materials as well. Our finding that strong correlation effects are needed to explain many of
the experimental observations also lends partial justification to our previous theoretical work which modelled the measured spin wave spectrum using a local moment model. Further theoretical work is needed to study the thermal fluctuation effects of the Fe moments, clarify what factors control the doping dependence of $\sigma_{xy}$, and to separate the bulk and surface contributions to the AHE. Furthermore, it would be useful to investigate if ferrimagnetic order in fact survives over a wide range of doping using an unbiased numerical approach. In future experiments, it would be useful to test our predictions for the doping dependence of the structural distortion and the AHE. Given that most DP materials are in the form of powder samples, measuring the AHE and separating the intrinsic contribution from extrinsic contributions would be experimentally challenging; Nevertheless systematic doping studies of the various properties of such 3d/5d DPs would be valuable. Finally, it appears extremely important to find ways to synthesize bulk single crystals or high quality thin films of such DP materials which would greatly open up the exploration of their physical properties and applications.

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\[ \epsilon_{xy} = -2t' \cos k_x a + \cos k_y a + 8t'' \cos \left( \frac{k_x a}{2} \right) \cos \left( \frac{k_y a}{2} \right) \cos \left( \frac{k_z c}{2} \right) \]
\[ \epsilon_{xz} = 2t'' \cos k_x a + \cos k_y a + 4t' \cos \left( \frac{k_x a - k_y a}{2} \right) \cos \left( \frac{k_z c}{2} \right) - 4t' \cos \left( \frac{k_x a + k_y a}{2} \right) \cos \left( \frac{k_z c}{2} \right) \]
\[ \epsilon_{yz} = 2t'' \cos k_x a + \cos k_y a + 4t' \cos \left( \frac{k_x a + k_y a}{2} \right) \cos \left( \frac{k_z c}{2} \right) - 4t' \cos \left( \frac{k_x a - k_y a}{2} \right) \cos \left( \frac{k_z c}{2} \right) \]

\[ \eta_{xy} = 4t_\sigma \cos \left( \frac{k_x a + k_y a}{2} \right) - 2t_\delta \cos \frac{k_z c}{2} \]
\[ \eta_{xx} = 2t_\sigma \cos \left( \frac{k_x a + k_y a}{2} \right) + 2t_\sigma \cos \frac{k_z c}{2} - 2t_\delta \cos \left( \frac{k_x a - k_y a}{2} \right) \]
\[ \eta_{yz} = 2t_\sigma \cos \left( \frac{k_x a - k_y a}{2} \right) + 2t_\sigma \cos \frac{k_z c}{2} - 2t_\delta \cos \left( \frac{k_x a + k_y a}{2} \right) \]

The intra-orbital terms take the form
\[ \gamma_{x,y,z} = -2t_m (\cos k_x a - \cos k_y a) \]
\[ \gamma_{x,y,z} = -4t_m \sin \left( \frac{k_x a + k_y a}{2} \right) \sin \frac{k_z c}{2} \]
\[ \gamma_{x,y,z} = 4t_m \sin \left( \frac{k_x a - k_y a}{2} \right) \sin \frac{k_z c}{2} \]