Quantum anomalous Hall phase in (001) double-perovskite monolayers via intersite spin-orbit coupling

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Using tight-binding models and first-principles calculations, we demonstrate the possibility to achieve a quantum anomalous Hall (QAH) phase on a two-dimensional square lattice, which can be realized in monolayers of double perovskites. We show that effective intersite spin-orbit coupling between $e_g$ orbitals can be induced perturbatively, giving rise to a QAH state. Moreover, the effective spin-orbit coupling can be enhanced by octahedral rotations. Based on first-principles calculations, we propose that this type of QAH state could be realized in La$_2$MnIrO$_4$ monolayers, with the size of the gap as large as 26 meV in the ideal case. We observe that the electronic structure is sensitive to structural distortions, and that an enhanced Hubbard $U$ tends to stabilize the nontrivial gap.

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I. INTRODUCTION

The quantum anomalous Hall (QAH) effect has drawn intensive attention recently, in part due to the dissipationless transport that can take place in the spin-polarized edge states, which are topologically protected against perturbative disorder. A generic model to achieve the QAH phase was first proposed by Haldane on the honeycomb lattice [1], where complex hoppings between next-nearest neighbors (NNNs) play a crucial role. Several systems have been proposed to host such nontrivial topological phases, such as magnetically doped topological insulators [2,3] and honeycomb lattices formed by transition-metal or heavy-metal ions [4–8]. For most of these systems, the occurrence of the QAH phase relies on the honeycomb lattice, and the topological properties are usually carried by the $sp$ bands. Meanwhile, spontaneous time-reversal symmetry breaking is usually induced by doping with magnetic ions or via a magnetic proximity effect. These two limitations greatly reduce the range of available candidate systems to search for the occurrence of a QAH state. In Cr-doped ($Bi_{1-x}Sb_x$)$_2$Te$_3$, for example, where the QAH phase has first been observed experimentally [9], the QAH effect is only observable below about 30 mK, due to the small exchange splittings induced by Cr doping.

In their seminal work, Xiao et al. proposed that in (111) superlattices of perovskite transition-metal oxides (TMOs), various topological phases can be obtained [10]. For TMOs with partially occupied $d$ shells, magnetism is relatively easy to obtain because the $d$ electrons are more localized than the $sp$ electrons. Furthermore, electronic correlations are usually significant in TMOs with localized $d$ electrons, and there is the possibility that nontrivial topological phases can develop by spontaneous symmetry breaking [11–13] with a dynamically generated spin-orbit coupling (SOC) [14]. It has even been theoretically argued that nontrivial topological phases can be realized in (111) TMO heterostructures without considering atomic SOC [15–17]. In all these proposals, the underlying honeycomb lattice facilitates the appearance of a topological phase. Unfortunately, it is difficult to synthesize (111) TMO superlattices experimentally with good atomic precision, although there has been some recent experimental progress in this direction [18].

The presence of a honeycomb lattice is not, however, a necessary condition for the occurrence of the QAH effect. For instance, topologically nontrivial phases can be obtained on square lattices with well-designed nearest-neighbor (NN) and NNN hoppings [19–22]. Recently, three proposals have been put forward to achieve the QAH effect in more realistic systems based on square-lattice symmetry, i.e., superlattices of CdO/EuO [23] and GdN/EuO [24] with the rocksalt structure and CrO$_2$/TiO$_2$ with the rutile structure [25]. For the latter case, the relevant bands are the $t_{2g}$ states of Cr; while these states show large exchange splittings, the topological gap is only about 4 meV due to the small strength of the onsite atomic SOC of Cr atoms.

In this work, we demonstrate the possibility of achieving a nontrivial QAH phase in (001)-oriented double-perovskite monolayers. Using a two-band model for $e_g$ orbitals on a square lattice, we show that complex effective intersite hoppings between two $e_g$ orbitals can be induced perturbatively by the atomic SOC, giving rise to a QAH state. Based on first-principles calculations, we further show that such a model can be realized in checkerboard La$_2$MnIrO$_6$ (LMIO) monolayers (ML) embedded in a nonmagnetic insulating host such as LaAlO$_3$ (LAO). The magnitude of the topological gap in the ideal case can be as large as 26 meV. The advantage of such a system is that (001) superlattices of perovskite compounds are well studied and can be synthesized with good atomic precision, resulting in controlled structural properties. Moreover, given the abundance of physical properties in perovskite TMO superlattices, including high-$T_c$ superconductivity [26], the QAH phase realized in (001) perovskite superlattices can also be integrated more easily with other functional oxides to achieve new physical properties.
The paper is organized as follows. In Sec. II, we present the tight-binding model for half-filled $e_g$ states on a square lattice. We demonstrate how the effective SOC can be induced following standard perturbation theory, focusing on the role of octahedral rotations. Detailed symmetry analysis is given to understand how the nontrivial topological phase develops. Our first-principles results are shown in Sec. III, where the model arguments in Sec. II are verified by considering a hypothetical structure. The effects of structural relaxations are then studied in detail and it is shown that epitaxial strain can be used to tune the LMIO monolayers close to the critical region where a nontrivial QAH state exists.

II. TWO-BAND MODEL

Our tight-binding model simulates a double-perovskite ML with checkerboard ordering of the two sublattices, either isolated in vacuum or embedded in an inert (wide-gap nonmagnetic) perovskite host. To be specific, we consider a case in which one sublattice is populated with ions having large exchange splittings, typically high-spin 3$d$ transition-metal ions, while ions with filled (e.g., $t_2g$) orbitals, respectively, while

\[ H = \begin{pmatrix} t_{1a} f_1(k) + t_{2a} f_2(k) & c.c. \\ t_{1c} - i\lambda^{(1)} g_1(k) - (t_{2c} - i\lambda^{(2)}) g_2(k) & \Delta + t_{1b} f_1(k) + t_{2b} f_2(k) \end{pmatrix}, \]

where $f_1(k) = \cos k_x + \cos k_y$, $g_1(k) = \cos k_x - \cos k_y$, $f_2(k) = 2 \cos k_x \cos k_y$, and $g_2(k) = 2 \sin k_x \sin k_y$. The model is parametrized by the difference $\Delta$ between the onsite energies of $d_{x^2-y^2}$ and $d_{z^2}$ orbitals, the NN hoppings $t_{1i}$, the NNN hoppings $t_{2i}$, and the effective SOC parameters $\lambda^{(1)}$ and $\lambda^{(2)}$, which respectively denote the NN and NNN couplings between $d_{z^2}$ and $d_{x^2-y^2}$ orbitals induced perturbatively as explained in the following. For the hoppings $t_{1i}$ and $t_{2i}$, $i = a$ or $b$ refers to the like-orbital hopping between $d_{z^2}$ or $d_{x^2-y^2}$ orbitals, respectively, while $i = c$ denotes the unlike-orbital hopping between $d_{z^2}$ and $d_{x^2-y^2}$ orbitals. Note that these are all "effective hoppings" in the sense that the oxygen and iridium orbitals are regarded as having been integrated out.

The complex hopping terms $i\lambda^{(1)}$ and $i\lambda^{(2)}$ in Eq. (1) between $d_{z^2}$ and $d_{x^2-y^2}$ orbitals can be induced by considering perturbative processes involving SOC. When there is no rotation of the transition-metal-oxygen octahedra, the $i\lambda^{(1)}$ term arises following

\[ i\lambda^{(1)} = \frac{\langle \hat{d}_{z^2}^{Mn} | \hat{H} | \hat{d}_{z^2}^{Mn} \rangle \langle \hat{d}_{z^2}^{Mn} | \hat{S} | \hat{d}_{x^2-y^2}^{Mn} \rangle}{E_{z^2}^{Mn} - E_{z^2}^{Mn}}, \]

where $\hat{L}$ ($\hat{S}$) is the orbital (spin) angular-momentum operator, $\xi^{Mn}$ is the strength of the atomic SOC on Mn, and $\hat{H}$ denotes direct hybridization between $d_{z^2}$ and $d_{x^2-y^2}$ orbitals located on NN Mn sites. $E_{z^2}^{Mn}$ and $E_{z^2}^{Mn}$ denote the onsite energies of the $t_{2g}$ and $e_g$ subshells on the Mn ions in the cubic crystal field. In a general case when the in-plane rotation angle $\theta$ [cf. Fig. 1(b)] is nonzero, it can be shown that $i\lambda^{(1)} \propto i\xi^{Mn} \sin(2\theta^{Mn}) = i\xi^{Mn} \cos(2\theta)$, where $\theta^{Mn}$ denotes the rotation angle of the MnO$_6$ octahedra and $\theta^{Mn} = \theta + 45^\circ$. That is, $i\lambda^{(1)}$ is an even function of the rotation angle $\theta$. We note that second-order processes involving Ir $t_{2g}$ orbitals can also lead to an effective SOC between the $e_g$ orbitals located on NN Mn sites, but the two most obvious contributions, corresponding to hopping via the two Ir atoms adjacent to a given Mn-Mn bond, tend to cancel one another.

A finite rotation angle $\theta$ leads to a nonzero $i\lambda^{(2)}$ term between $e_g$ orbitals located on NNN Mn sites. It arises following

\[ i\lambda^{(2)} = \frac{\langle \hat{d}_{z^2}^{Mn} | \hat{H} | \hat{d}_{x^2}^{Ir} \rangle \langle \hat{d}_{x^2}^{Ir} | \hat{S} | \hat{d}_{x^2-y^2}^{Mn} \rangle}{E_{z^2}^{Ir} - E_{z^2}^{Mn}}, \]

where $\xi^{Ir}$ denotes the strength of the atomic SOC on Ir sites, $E_{z^2}^{Ir}$ and $E_{z^2}^{Mn}$ are the onsite energies of the Ir $t_{2g}$ and $e_g$ orbitals, and $\hat{H}$ denotes the direct hybridization between orbitals on Mn and Ir atoms. Similar virtual transitions involving coupling of the $d_{x^2-y^2}$ orbital of Mn to the $d_{xy}$ orbital of Ir also lead to nonzero contributions. The resulting total $i\lambda^{(2)} \propto i\xi^{Ir} \sin(2\theta)$, with $\theta$ the octahedral rotation angle. That is, $i\lambda^{(2)}$ is an odd function of $\theta$. Furthermore, the magnitude of $\lambda^{(2)}$ is determined

FIG. 1. (Color online) (a) Oblique view of the crystal structure of a La$_2$MnO$_6$ (LMIO) monolayer (ML) sandwiched between LaAlO$_3$ layers. (b) Sketch defining the parameters of the tight-binding model of Eq. (1) that describes the LMIO ML. Only the local $e_g$ orbitals on Mn atoms (purple, at origin) are shown; orbitals on Ir (brown, at the center) are suppressed. The intersite hoppings between Mn $e_g$ orbitals on nearest neighbors ($t_1$) and next-nearest neighbors ($t_2$) are shown by arrows (see main text for details). The octahedral rotations, denoted by angle $\theta$ in (b), are exaggerated for clarity of illustration.

The crystal structure of such an LMIO monolayer sandwiched between LAO layers is shown in Fig. 1(a). The corresponding tight-binding model for the two $e_g$ orbitals on each Mn site can be expressed in the local $(d_{z^2}, d_{x^2-y^2})$ basis as...
by the strength of the atomic SOC of the Ir atoms. We observe that for the LMIO monolayers considered in this work, the magnitude of $\lambda^{(2)}$ is about one order of magnitude larger than that of $\lambda^{(1)}$, due to the much stronger atomic SOC of Ir ($\sim 0.5$ eV) compared to that of Mn ($\sim 0.05$ eV).

The Hamiltonian in Eq. (1) is fully consistent with the symmetry requirements. Our analysis reveals that there are four kinds of symmetry operations that are important: twofold rotations $C_2$ centered at the midpoints of NN Mn-Mn bonds, twofold rotations $C_2$ centered at the Ir sites, mirrors $M_{x/y}$ with respect to a plane connecting the centers of the NN Mn-Mn bonds along either $x$ or $y$, and mirrors $M'$ connecting NNN Mn sites. Here, the rotational axes and mirror planes are all perpendicular to the MnIr plane, and $M'$ denotes a combination of a mirror and the time-reversal operations. A detailed analysis (cf. Appendix) confirms that both $\lambda^{(1)}$ and $t_{2g}$ are even functions of the rotational angle $\theta$, and both $\lambda^{(2)}$ and $t_{2g}$ are odd functions of $\theta$, leading to the Hamiltonian shown in Eq. (1).

The two-band model of Eq. (1) can be solved analytically by decomposing the Hamiltonian as $H = \sigma_x h_0 + \sum_{i=1}^{3} h_i \sigma_i$, where $\sigma_0$ is the unit matrix and $\sigma_i$ are the Pauli matrices. The Berry curvature for the lower-lying band can be obtained explicitly as

$$\Omega = -\frac{2}{h} \sum_{ij} \epsilon_{ijk} h_i \sigma_j h_k,$$

where $\epsilon_{ijk}$ is the Levi-Civita symbol, $h_0 = \sqrt{h_1^2 + h_2^2 + h_3^2}$, $h_{1,2,3} = \partial h_i / \partial k_{i'} (a = x,y)$, and $E_{\pm} = h_0 \pm h$ are the energy eigenvalues for the higher/lower bands. In our case,

$$h_0 = [\Delta + (t_{1a} + t_{1b}) f_1(k) + (t_{2a} + t_{2b}) f_2(k)]/2,$$

$$h_1 = t_{1c} g_1(k) - t_{2a} g_2(k),$$

$$h_2 = \lambda^{(1)} g_1(k) - \lambda^{(2)} g_2(k),$$

$$h_3 = [-\Delta + (t_{1a} - t_{1b}) f_1(k) + (t_{2a} - t_{2b}) f_2(k)]/2.$$

The band structure obtained from a model of this form is presented in Fig. 2(a), and the regions of strong Berry curvature, corresponding to small direct gaps, are shown as the (blue/red) shaded regions in Figs. 2(b)–2(d). The diagonal hopping parameters for the plots were obtained by fitting to the first-principles band structure of Fig. 3(b), yielding $t_{1a} = -0.27$ eV, $t_{1b} = 0.09$ eV, $t_{2a} = 0.05$ eV, $t_{2b} = -0.105$ eV, and $\Delta = 0.28$ eV. The off-diagonal terms were set to $t_{1c} = 0$ eV, $t_{2c} = 0.02$ eV, $\lambda^{(1)} = 0.02$ eV, and $\lambda^{(2)} = 0.08$ eV. The inset of Fig. 2(a) shows the computed edge states for an 80-unit-cell-wide ribbon from this model, providing the first evidence that the model exhibits a nontrivial topology.

To understand how these features of the band structure come about, it is useful to return to Eqs. (4) and (5). Note that $h_1$ and $h_2$ both have to be present in order to obtain a nonzero Berry curvature. Actually, we find that nonzero $t_{2g}$ and $\lambda^{(1)}$ or nonzero $t_{1c}$ and $\lambda^{(2)}$ can both lead to nontrivial topological phases, corresponding to the case without rotations and the case with only terms induced by rotations, respectively. Interestingly, the Chern numbers are of opposite sign for the two cases. Due to the much larger magnitude of $\lambda^{(2)}$, the Chern number of the system is determined in practice by the combination of $t_{1c}$ and $\lambda^{(2)}$.

Consider first the case that the octahedral rotation angle $\theta$ vanishes. Then, $t_{1c} = 0$ because symmetry prevents any direct hybridization of $d_{z^2}$ and $d_{x^2-y^2}$ orbitals on NN Mn sites. In this case, $\Delta^{(1)} = 0$ as well because the hybridization of $d_{z^2}$ and $d_{x^2-y^2}$ orbitals on NNN Mn sites via Ir atoms is forbidden. In fact, we observed that both $t_{2g}$ and $\lambda^{(2)}$ are proportional to $\sin 2\theta$, as explained above for $\lambda^{(2)}$. That is, without octahedral rotations only $t_{2g}$ and $\lambda^{(1)}$ in the off-diagonal terms of Eq. (1) are nonzero. Examining Eq. (1) reveals that without the off-diagonal terms proportional to $\lambda^{(1)}$ and $t_{2g}$, the eigenvalues of the Hamiltonian are degenerate wherever $h_3 = 0$, which turns out to be a loop centered at the $M$ point in the Brillouin zone (BZ) as shown by the green lines in Fig. 2(b). This reflects the fact that neither $f_1(k)$ nor $f_2(k)$ vanishes in the vicinity of $M$. By contrast, $g_1(k)$ vanishes along the $\Gamma-M$ lines, while $g_2(k)$ vanishes along the $X-M$ lines, as indicated by the (black) dashed and (blue) dotted lines in Fig. 2(b), respectively. Thus, the loop of degeneracy is reduced to four points (Dirac nodes)

![Fig. 2. (Color online) Electronic structure of the tight-binding model of Eq. (1) with parameters as given in the text. (a) Band structure along the high-symmetry $\Gamma-X-M-\Gamma'$ path. Inset shows the projected bulk band structure (shaded region) and edge states for an 80-unit-cell-wide ribbon in the reduced 1D BZ along $\Gamma-X-M$, color coded to distinguish the contributions from the two edges; dashed line denotes $E_F$. (b)–(d) Color map showing the distribution of negative (blue) and positive (red) Berry curvature in the BZ for three cases: (b) without octahedral rotations ($t_{2g}$ and $\lambda^{(1)}$ nonzero); (c) with octahedral rotations ($t_{2g}$ and $\lambda^{(2)}$ nonzero) but $t_{1c}$ and $\lambda^{(1)}$ artificially set to zero; and (d) with octahedral rotations, including all four terms. For guidance, nodes of $h_1$, $h_2$, and $h_0 + h_1$ in Eq. (5) are shown as dotted blue, dashed black, and solid green curves, respectively.](Image 322x513 to 337x526)

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located on the $X$-$M$ directions if $\lambda^{(1)}$ is turned on, or on the $\Gamma$-$M$ lines if $t_{2\epsilon}$ is turned on.

When both $\lambda^{(1)}$ and $t_{2\epsilon}$ are nonzero, the energy spectrum of Eq.~(1) is fully gapped, leaving concentrations of Berry curvature in the regions of the BZ where the gap is small, as shown by the (blue) shading in Fig.~2(b). Since the magnitude of $\lambda^{(1)}$ is comparable to that of $t_{2\epsilon}$, the distribution of the Berry curvature is quite smeared. The resulting total Chern number is shown by the (blue) shading in Fig.2(b). Since the magnitude of $\lambda^{(1)}$ is comparable to that of $t_{2\epsilon}$, the distribution of the Berry curvature is quite smeared. The resulting total Chern number is defined as given in Eq. (1), the nodes of $h_2$ shift slightly in the counterclockwise direction relative to the case of Fig.~2(c).

There is also a shift (larger and in the opposite direction) in the nodes of $h_1$ arising from the competition between $t_{1\epsilon}$ and $t_{2\epsilon}$. However, because of the large magnitude of $\lambda^{(2)}$, the avoided crossings follow the nodes of $h_2$. The result is that the four avoided-crossing points of the full Hamiltonian are rotated slightly in the counterclockwise direction around $M$ compared to Fig.~2(c). Moreover, the topological properties of our model are dominated by the combination of $\lambda^{(2)}$ and $t_{1\epsilon}$, so that the Chern number is $C = 2$. That is, the octahedral rotations induce a topological phase transition where the Chern number changes from $-2$ to $2$. The topological nontriviality is also confirmed by an explicit calculation of the edge states of a one-dimensional (1D) ribbon as shown in the inset of Fig. 2(a).

It is evident that two edge states with the same group velocity are located on one edge, while another two edge states with the opposite group velocity are located on the other edge.

The fact that the absolute value of the Chern number is two, and not one, can be understood in several ways. For example, we note that in the limit of a very small coefficient of $g_2$, so that the four Dirac points are just barely opened, each should carry a Berry phase of $\pm \pi$ (as is standard for a simple avoided Dirac crossing); four of them add to $\pm 4\pi$, suggesting $C = \pm 2$. Another approach is to consider the limit that the four degeneracy points shrink to the $M$ point and merge. Specifically, we find that by tuning $\Delta$, the difference between onsite energies of valence $d_{\epsilon z}$ and conduction $d_{\epsilon z-\epsilon y}$...
orbits in the tight-binding model, the four avoided crossings shrink to a singular point with quadratic dispersion at $M$ when $\Delta_c = 1.034$ eV, and the gap reopens in a normal C phase for $\Delta > \Delta_c$. As pointed out in Ref. [27], the Chern-number transfer should always be $\Delta C = 2$ in the case of a critical quadratic band touching. This can also be understood based on the symmetry of the orbitals. At the $M$ point, $d_z^2$ and $d_{xy}$ states are both eigenstates of the $C_4$ symmetry operator, but with eigenvalues of $+1$ and $-1$, respectively. If these labels had been adjacent in the cycle of possible eigenvalues $(1, -1, -1, -1, ...)$, a Chern transfer $\Delta C = \pm 1$ would have been expected; but because they are not, we get $\Delta C = \pm 2$ [28].

Finally, we note that since the minimum avoided crossings are not in general located on the high-symmetry $X-M$ or $\Gamma-M$ $k$ path when octahedral rotations are present, the actual band gap is smaller than the one obtained from a band structure plotted along the $\Gamma-X-M-\Gamma$ high-symmetry lines. For example, a direct inspection of Fig. 2(a) suggests a gap of $30$ meV, compared to the true value of $25$ meV obtained from a more careful scan over the full 2D BZ.

III. FIRST-PRINCIPLES CALCULATIONS

In this section, we demonstrate how the tight-binding model discussed above can be realized in more realistic systems. The spin-polarized half-filled $e_g$ states could be realized by a $d^2$ or $d^3$ configuration, while non-spin-polarized sublattice could be populated by $d^0$, $d^6$, or $d^{10}$ ions. In this work we considered a specific system consisting of a monolayer of LMIO sandwiched into an LAO environment, as shown in Fig. 1(a), although the realization of the tight-binding model is not limited to this specific system. We have chosen LAO as the host environment because it has a large bulk gap of $5.6$ eV, so that the states around the Fermi energy ($E_F$) will be dominated by the orbitals in the LMIO layers.

Our first-principles calculations are done using the projector augmented wave method as implemented in VASP [29]. The exchange-correlation potentials are approximated using the Perdew-Burke-Ernzerhof functionals [30]. For all the structures considered, the in-plane lattice constants are fixed at $3.789$ Å, the cubic lattice constant of bulk LAO. For the self-consistent total-energy calculations, the plane-wave energy cutoff is taken to be $500$ eV. All our calculations are carried out using the $\sqrt{2} \times \sqrt{2} \times 3$ supercell shown in Fig. 1(a), which also accommodates octahedral rotations about the $z$ axis, and a $k$-point set corresponding to an $8 \times 8 \times 4$ mesh in the full BZ is used.

To treat Coulomb interactions for open shells, we applied the GGA+$U$ method [31] with double counting considered in the fully localized limit. Since the $t_{2g}$ shells of Ir are almost fully occupied, the GGA+$U$ corrections are only applied to Mn sites. Initially, our calculations are all carried out with $U = 5.0$ eV and $J = 1.0$ eV [32], corresponding to commonly accepted values for Mn$^{3+}$. Later, we study the effect of varying the $U$ value on the Mn sites, as discussed in the following. In all our calculations, we assume that the magnetic moments of Mn are ferromagnetically coupled. To shift the $4f$ states of La away from $E_F$, we impose $U_{4f} = 11$ eV and $J_{4f} = 0.68$ eV as used for other calculations on heterostructures [33]. The AHC is obtained by Wannier interpolation using an effective Hamiltonian constructed in a basis of 128 maximally localized Wannier functions [34] corresponding to all Mn-3$d$, Ir-5$d$, and O-2$p$ orbitals in the supercell.

Consider first a hypothetical structure without octahedral rotations, specifically one in which the in-plane Mn-O and Ir-O distances are set to be equal, and the out-of-plane Mn-O distance is set to be $2.0$ Å. The first-principles band structure is shown in Fig. 3(a). Due to the strong atomic SOC of Ir atoms, their $t_{2g}$ bands are separated into a group of four lower-lying $J = \frac{3}{2}$ bands and two higher $J = \frac{5}{2}$ bands [35]. On the other hand, it is observed that the $3d$ bands of Mn are significantly spin polarized, with an average exchange splitting of about $2$ eV even without a local Hubbard $U$ applied. Therefore, the bands around $E_F$ are mostly a mixture of $e_g$ bands of the majority spin character from Mn and the $J = \frac{5}{2}$ bands from Ir. The $e_g$ states of Mn are half-filled, leading to an atomic magnetic moment of about $4\mu_B$. The hybridization of the Ir $t_{2g}$ states with the Mn $e_g$ states induces small (about $0.05\mu_B$) magnetic moments on the Ir sites.

Introducing octahedral rotations in the LMIO layers leads to significant changes of the band structure by inducing additional hybridizations. Figure 3(b) shows the band structure with a staggered rotation of the MnO$_6$ and IrO$_6$ octahedra of $15^\circ$ about the $z$ axis, while all the other degrees of freedom remain fixed. Now, only two bands, namely of Mn $d_{z^2}$ and $d_{xy}$ character, are left around $E_F$, although these orbitals hybridize strongly with the Ir 5$d$ orbitals. Recalling the arguments given above in connection with our tight-binding model, such hybridization is crucial for inducing the effective SOC $\lambda$ in Eq. (1), which in turn helps to give a nontrivial gap. We confirm that the gap is indeed nontrivial, with a quantized AHC of $2\hbar^2/\nu$ by direct calculation as shown in Fig. 3(c). The topological band inversion is also evident in the band structure of Fig. 3(b), where the band characters have clearly exchanged between the conduction and valence bands around the $M$ point. We note that the size of the gap is about $26$ meV as measured by the width of the quantized AHC plateau, which is smaller than the gap obtained from inspection along the high-symmetry $k$ path in Fig. 3(b); this is again due to the fact that the avoided-crossing points are not located on the high-symmetry lines [cf. Fig. 2(d)].

To be more realistic, we relaxed the structures by allowing the out-of-plane lattice constant and internal coordinates to vary, but keeping the in-plane lattice constants fixed at those of LAO. We find the relaxed octahedral rotation angle in the LMIO layers to be $15.6^\circ$, and the relaxed out-of-plane Mn-O distance is about $2.02$ Å. By these measures, the ideal structure discussed above is quite reasonable. However, the most drastic change occurs locally in the MnO$_6$ octahedra, where the local $c/a$ ratio (i.e., the ratio of apical to in-plane Mn-O bond lengths) increases to $1.06$, from $1.02$ in the ideal structure. This change results from a contraction of the in-plane Mn-O distances.

Figure 3(d) shows the band structure for the fully relaxed structure. Evidently, the $d_{z^2}$ and $d_{xy}$ bands are shifted to higher energies due to the variations of the onsite energies of the $d_{z^2}$ and $d_{xy}$ orbitals caused by the local distortions of the MnO$_6$ octahedra. The resulting $d_{z^2}$ and $d_{xy}$ bands no longer overlap anywhere in the BZ, and as a result the gap at $E_F$ is topologically trivial, as verified by our calculations of the AHC.
As emphasized in the inset of Fig. 3(e), and our calculation of the gap opened around is a consequence of the fact that the off-diagonal terms in the magnitude of the gap is quite small, only about 1 meV. This confirms that it is topologically nontrivial. However, the AHC confirms that it is topologically nontrivial. Moreover, the local conduction-band minimum at \( g \) below falls about 120 meV below the conduction-band minimum at \( g \) (not shown). Another consequence of the structural relaxations is that the local conduction-band minimum at \( y \) has shifted downward and now falls about 120 meV below the conduction-band minimum at \( M \). This is caused by a change in the sign of the hopping parameter between \( d_{x^2-y^2} \) orbitals located on NN sites, i.e., \( t_{1u} \) in Eq. (1). Thus, even if some means could be found to restore the band inversion at \( M \), this reversal in the energy ordering of the conduction-band minima could prevent the maintenance of a global gap, forcing the system metallic.

To overcome these negative effects of the structural relaxations, which disfavor the topological phase, tensile epitaxial strain can be applied to increase the in-plane lattice constants and decrease the out-of-plane one, thus reducing the local octahedral distortions. Figure 3(e) shows the band structure with a 2% tensile epitaxial strain applied to the LAO substrate. In this case, the \( d_{x^2-y^2} \) bands are shifted downward in energy relative to the \( d_{z^2} \) bands, once again overlapping with them. The gap opened around \( M \) shows a typical anticrossing behavior, as emphasized in the inset of Fig. 3(e), and our calculation of the AHC confirms that it is topologically nontrivial. However, the magnitude of the gap is quite small, only about 1 meV. This is a consequence of the fact that the off-diagonal terms in the Hamiltonian of Eq. (1) vanish as one approaches the \( M \) point because of the form of \( g_1(\mathbf{k}) \) and \( g_2(\mathbf{k}) \). To our satisfaction, we observe that the \( d_{x^2-y^2} \) conduction-band minimum at \( \Gamma \) remains above that at the \( M \) point, if only barely (by \( \sim 5 \) meV), so that the gap around \( M \) is a true global gap. We conclude that a tensile strain of at least 2% is needed to obtain the QAH state, and speculate that out-of-plane uniaxial pressure could help further.

Interestingly, increasing the strength of the local Hubbard \( U \) on the Mn sites also tends to stabilize the topological phase. Figure 3(f) shows the resulting band structure obtained by increasing the Hubbard parameter to \( U = 7 \) eV on the Mn sites, with the conditions otherwise the same as in Fig. 3(e) (i.e., relaxed with 2% tensile epitaxial strain). Larger \( U \) not only shifts the conduction-band minimum at \( \Gamma \) upwards away from \( E_F \), but also enhances the magnitude of the nontrivial topological gap opened around the \( M \) point. The magnitude of the global band gap is calculated to be about 4 meV. This is much smaller than the band gap derived from states along the high-symmetry \( k \) path, which is about 25 meV, again because the avoided crossings are not located on the high-symmetry lines as explained in Sec. II. Moreover, significant changes occur in the hybridizations between the valence states, caused by the enhanced local atomic \( U \) values on the Mn sites. For instance, the \( d_{z^2} \) bands are shifted to lower energies [Fig. 3(e) versus Fig. 3(f)], and the first valence band below \( E_F \) acquires more \( d_{x^2-y^2} \) character with increasing \( U \) because of more significant hybridization with the \( d \) states of Ir atoms.

**IV. CONCLUSIONS**

In conclusion, we have demonstrated the possibility of achieving a quantum anomalous Hall phase on a square lattice via an appropriate pattern of intersite spin-orbit couplings between \( d \) orbitals, which can be realized in double-perovskite monolayers. We have shown that for a half-filled manifold of \( e_g \) orbitals, an effective SOC can be induced by hybridizing with other \( d \) orbitals located on the neighboring sites, even though no direct onsite SOC is present. We have found, in particular, that octahedral rotations can induce an effective SOC between \( e_g \) orbitals located on NNN sites. We have demonstrated that a simple tight-binding Hamiltonian encoding the most important features of the interactions gives rise quite generically to a quantum anomalous Hall phase. Then, using first-principles calculations, we have also shown that such a model can be realized in La$_2$MnIrO$_6$ monolayers. The gap can be as large as 26 meV in the ideal case. However, there are several open issues that need further investigation for this system. First, we have assumed ferromagnetic order, even though there is some tendency of the magnetic moments of the Mn ions to be coupled antiferromagnetically. For instance, we observed that the antiferromagnetic configuration is about 31 meV lower in energy than the ferromagnetic case for the relaxed structure without strain. Nevertheless, for the hypothetical structure, the ferromagnetic state is more stable, about 28 meV lower in energy than the antiferromagnetic state. Hence, the exchange coupling between neighboring Mn moments is quite sensitive to local distortions, which can hopefully be tuned by epitaxial strain. This problem may also be remedied by choosing a substrate with a magnetic order that can enforce the desired ferromagnetic state. Second, there is the issue of the assumed checkerboard compositional order. Even though both Mn and Ir are 3+ ions, which by itself would give no strong tendency toward ordering within the La$_2$MnIrO$_6$ monolayer, we argue that such a tendency may come instead from the substantial difference in the ionic radii. Third, it would be useful to understand the role of correlations in more detail. We have found that the \( e_g \) states of Mn are quite localized, leading to flat bands, and enhancing the local Hubbard \( U \) favors larger band gaps while maintaining the topological nontriviality. We note that our discussion is currently limited to the GGA+U scheme where the onsite electron interactions are treated in a Hartree-Fock manner, while the interplay of electronic correlations and SOC on the square lattices deserves further investigation from beyond-DFT methods.

Finally, as discussed above, the global gap may not remain open after the contraction of the in-plane Mn-O distances with relaxation. We have shown that this problem may be overcome by engineering structures utilizing tensile epitaxial strain of about 2%, making use of the sensitivity of the relevant \( d_{x^2-y^2} \) and \( d_{z^2} \) bands to local distortions. Further stabilization of the QAH phase might be achieved by varying the choice of the inert perovskite surrounding material, by applying vertical uniaxial pressure in addition to the tensile epitaxial strain, or by chemical doping within the double-perovskite layer or in the surrounding material. Other combinations of transition-metal ions, in which one has a half-filled \( e_g \) shell, should also be explored. Lastly, we suspect that the idea of intersite SOC on the square lattices should be applicable to ions with partially filled \( t_{2g} \) shells as well. These interesting questions are left for future investigations.

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APPENDIX: SYMMETRY ANALYSIS OF THE TWO-BAND MODEL

For the two-band model we considered in the main text, it can be decomposed following

\[ H = h_0 \sigma_0 + h_1 \sigma_x + h_2 \sigma_y + h_3 \sigma_z, \]  

(A1)

where \( \sigma_0 \) denotes the unit matrix and \( \sigma_i \) (\( i = x, y, z \)) denote the Pauli matrices. We note that \( h_1 \) (\( i = 0, 1, 2, 3 \)) can be written as \( h_1 = h'_1 + h''_1 \), where \( h'_1 \) and \( h''_1 \) indicate the matrix elements for hopping to the nearest neighbors (NNs) and next-nearest neighbors (NNNs), respectively.

The symmetry operators we considered are \( C_4, C_2, M'_i, \) and \( M''_i, \) as shown in Figs. 4 and 5. The \( C_4 \) symmetry relates hoppings in directions \( \varphi \) and \( \varphi + 90^\circ \) in an obvious way, but does not impose constraints on the form of the hoppings themselves. Moreover, both \( C_2 \) operations are symmetries even when octahedral rotations are present, and act at mid-bond positions as shown in Figs. 4 and 5. Examining the transformation of the Hamiltonian matrix elements reveals that \( H_{\alpha\beta} = H'_{\alpha\beta} \). That is, \( H \) is Hermitian, so that \( h_i \) (\( i = 0, 1, 2, 3 \)) in Eq. (A1) are real functions of \( \mathbf{k} \).

Further constraints on the form of the Hamiltonian can be obtained for the following two situations.

1. When there are no octahedral rotations (cf. Fig. 4): In this case, both \( M'_i \) and \( M''_i \) are symmetry operators:
   - (i) Applying \( M'_i \) to the NN hoppings reveals that \( h'_i = 0 \).
   - (ii) Applying \( M''_i \) to the NNN hoppings reveals that \( h''_i = 0 \).

   That is, for the off-diagonal term, only \( h'_i \) and \( h''_i \) are allowed by symmetry, which correspond to the \( \lambda^{(1)} \) term and the \( t_{2g} \) term in our Hamiltonian in the main text, respectively.

2. When there are finite octahedral rotations: In this case, both \( M'_i \) and \( M''_i \) relate the cases with positive and negative octahedra-rotational angles. For instance,

   \[ \text{Config } (\theta) = M'_i \text{ Config } (-\theta), \]

   \[ \text{Config } (\theta) = M''_i \text{ Config } (-\theta), \]

where \( \text{Config } (\pm \theta) \) denotes the configuration with positive/negative rotational angle \( \theta \):

   - (i) Applying \( M'_i \) to the NN hoppings reveals that \( h'_i(-\theta) = -h'_i(\theta) \), and \( h''_i(-\theta) = h''_i(\theta) \). That is, \( h'_i \) (\( h''_i \)) is an even (odd) function of rotational angle \( \theta \).
   - (ii) Applying \( M''_i \) to the NNN hoppings reveals that \( h'_i(-\theta) = h'_i(\theta) \), and \( h''_i(-\theta) = -h''_i(\theta) \). That is, \( h'_i \) (\( h''_i \)) is an even (odd) function of rotational angle \( \theta \).

In this sense, when the octahedral rotations are switched on, \( h'_i \) and \( h''_i \) are allowed, which correspond to the \( t_{1u} \) term and \( \lambda^{(2)} \) term in our Hamiltonian in the main text, respectively.

In short, following the symmetry argument, both \( \lambda^{(1)} \) and \( t_{2g} \) are even functions of the rotational angle \( \theta \), and both \( \lambda^{(2)} \) and \( t_{1u} \) are odd functions of the rotational angle \( \theta \). We note that \( \lambda^{(1)}, \lambda^{(2)}, t_{2g}, \) and \( t_{1u} \) can be grouped into \{\( \lambda^{(1)}, t_{2g} \)\} and \{\( \lambda^{(2)}, t_{1u} \)\}, which act separately to induce nontrivial topological properties of the system. As discussed above, \{\( \lambda^{(1)}, t_{2g} \)\} are finite without octahedral rotations and can be shown to be even functions of the octahedral-rotation angle \( \theta \), while \{\( \lambda^{(2)}, t_{1u} \)\} are not allowed by symmetry when \( \theta = 0 \) and are odd functions of \( \theta \). Our tight-binding derivation using the Slater-Koster parameters confirms also this picture, and two important effective spin-orbit coupling parameters \( \lambda^{(1)} \) and \( \lambda^{(2)} \) are given explicitly in Eqs. (2) and (3) in the main text. We also note that either \{\( \lambda^{(1)}, t_{2g} \)\} or \{\( \lambda^{(2)}, t_{1u} \)\} can lead to nontrivial topological phases (as shown in Fig. 2 in the main text), with opposite resulting Chern numbers. The final state is determined by the competition between these two groups of parameters, and we observed that the \{\( \lambda^{(2)}, t_{1u} \)\} term would be dominant due to \( |\lambda^{(2)}| > |\lambda^{(1)}| \). This is confirmed by our first-principles calculations, and detailed discussion is given in the main text.