

Exploring d -wave magnetism in cuprates from oxygen momentsYing Li^{1,2,*}, Valentin Leeb^{1,3,*}, Krzysztof Wohlfeld⁴, Roser Valentí⁵, and Johannes Knolle^{1,3,6}¹Physics Department, *Technical University of Munich*, TUM School of Natural Sciences, 85748 Garching, Germany²MOE Key Laboratory for Nonequilibrium Synthesis and Modulation of Condensed Matter, Shaanxi Province Key Laboratory of Advanced Materials and Mesoscopic Physics, School of Physics, *Xi'an Jiaotong University*, Xi'an 710049, China³*Munich Center for Quantum Science and Technology (MCQST)*, Schellingstr. 4, 80799 München, Germany⁴Institute of Theoretical Physics, Faculty of Physics, *University of Warsaw*, Pasteura 5, PL-02093 Warsaw, Poland⁵Institut für Theoretische Physik, *Goethe-Universität Frankfurt*, Max-von-Laue-Strasse 1, 60438 Frankfurt am Main, Germany⁶Blackett Laboratory, *Imperial College London*, London SW7 2AZ, United Kingdom

(Received 17 December 2024; revised 7 July 2025; accepted 21 August 2025; published 17 September 2025)

The antiferromagnetic parent phase of high- T_c cuprates has been established as a Néel state of copper moments, but early work pointed out the important role of ligand oxygen orbitals. Using the three-orbital Emery model, we explore how, and under which conditions, doping-induced antiferromagnetic ordering of weak magnetic moments on the oxygen sites can lead to unconventional d -wave magnetism with spin-split electronic bands. The mechanism for forming such altermagnetic (AM) states in cuprates does not rely on a lowering of the crystal symmetry but rather on interaction-induced formation of magnetic moments on directional oxygen orbitals within the crystallographic unit cell. Therefore, we obtain two different types of AM, namely, a $(0, 0)$ -AM and a (π, π) -AM. We explore different regimes and challenges for realizing oxygen AM supported by Hartree-Fock calculations and complementary exact diagonalization of small clusters. While the region of interacting parameters needed to realize these states may be difficult to achieve in known high- T_c cuprates, we propose a scenario to realize AM induced by oxygen magnetic moments in a cuprate-based candidate compound using density functional theory and discuss experimental implications.

DOI: [10.1103/vx12-r2k1](https://doi.org/10.1103/vx12-r2k1)

I. INTRODUCTION

Understanding the parent phase of cuprates has been key for elucidating the emergence of high- T_c superconductivity [1,2]. The insulating parent Néel state is typically described as a commensurate antiferromagnet (AFM) of a single orbital model of magnetic moments on the copper sites [3–5]. However, early on Emery discussed the relevance of oxygen orbitals for understanding the magnetic fluctuations upon doping which are responsible for pairing [6]. Not only is the exchange between copper moments mediated via the directional p_x , p_y orbitals [6–10] but the position of oxygen on the bond centers leads to a unit cell with an internal structure which allowed for the prediction of intra-unit-cell order, for example, of spin-nematic [11,12] or loop current type [13,14]. Neutron scattering experiments established the importance of the hybridization between oxygen and copper orbitals for understanding the magnetic form factors [15]. Famously, in the pseudogap phase neutron scattering measurements also observed magnetism without additional lattice symmetry breaking pointing to intra-unit-cell order [16,17],

which was argued to be consistent with staggered magnetic moments of the oxygen sites. Motivated by these experiments, we aim to understand the mechanism for stabilizing magnetism on oxygens, as well as the resulting unique properties of the magnetic states, in the context of cuprates described by the basic Emery model.

Recently, a new type of collinear antiferromagnetism, so called altermagnetism (AM) based on a spin-symmetry classification, has been proposed [18–22] which is a unified way to describe previous reported anomalous behavior in magnetic systems [23–26]. AM combines spin-split electronic bands expected for ferromagnets (FM) with a vanishing net moment of AFMs. The reason is the absence of translational (or inversion) symmetry between the AFM sublattices, which can either originate from a low crystal symmetry or appear spontaneously from directional orbital ordering [27]. In the context of AMs, the cuprate La_2CuO_4 has been proposed as a candidate material [20,28] arising from a reduced crystal symmetry due to the canting of CuO_6 octahedra. However, the resulting spin splittings are small and disappear when the structure becomes tetragonal under doping. In three-dimensional analogs of the parent compound $\text{CuAg}(\text{SO}_4)_2$, AM appears, again, because of crystallographically different Cu-Ag sublattices [29].

Here, we show that AM of the d -wave type can already appear in highly symmetric tetragonal CuO_2 planes if magnetic moments can be stabilized on directional oxygen orbitals. Typically, cuprates are modeled to have AFM moments on the Cu atoms (see upper right panel of Fig. 1). Here, we explore the possibility of additional AFM moments on the oxygen

*These authors contributed equally to this work.

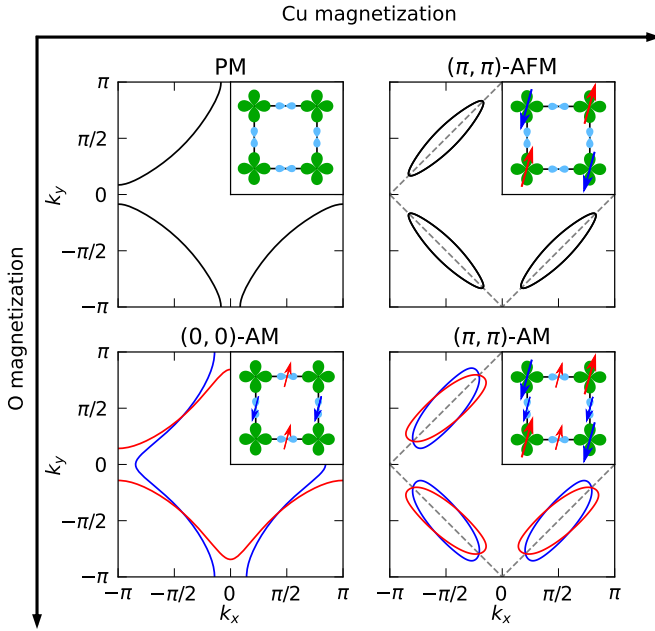


FIG. 1. Typically the AFM phase of cuprates features AFM moments on the Cu atoms (x axis). Here, we additionally explore the formation of AFM moments on the oxygen orbitals (y axis), which leads to two different types of AM states. The insets of the subpanels show the possible magnetic phases we obtain (Cu green, O light blue). The subpanels show the spin-resolved Fermi surface (FS) for the corresponding phase (black degenerate, red spin up, blue spin down). The magnetic Brillouin zone is shown as gray dashed square. Note that the lower left intra-unit-cell ordering does not break translational symmetry as suggested in previous neutron scattering experiments [16,17].

orbitals (lower panels), which can result in two distinct types of AM states: First, a $(0, 0)$ -AM which does not break any translational symmetry but leads to a spin splitting because of the intra-unit-cell AFM arrangement. Second, a (π, π) -AM can appear where both Cu and O have AFM moments and the unit cell is spontaneously enlarged leading to a backfolding of the electronic bands (see right panels). The interesting point is that for both scenarios intra-unit-cell oxygen AFMs are responsible for distinct spin-split electronic bands with a d -wave form factor as shown in the lower panels of Fig. 1.

We note that AM without translational symmetry breaking has been discussed previously [11,30,31] and, indeed, the spin-nematic mean-field state identified in Ref. [11] is our $(0, 0)$ -AM. Here, we concentrate on exploring the microscopic origin of oxygen-induced cuprate AM, the microscopic parameter regime, and its unique electronic properties. We identify three distinct microscopic mechanisms that could lead to the formation of magnetic moments on the oxygens: (i) a direct exchange between oxygen due to a large repulsive interaction U_p on oxygen sites; (ii) a strong charge transfer bringing the p -oxygen orbitals closer to the Fermi energy; and (iii) a large oxygen-oxygen hopping t_{pp} effectively shifting magnetic moments of copper to oxygens.

An important question concerns the parameter regime in which oxygen moments appear. We find that for oxygen-oxygen hoppings and Coulomb repulsion values usual for

cuprates the ligand sites remain nonmagnetic. However, using Hartree-Fock mean-field theory (MFT) and complementary exact diagonalization (ED) studies of small clusters, we find that slightly larger values, which are potentially relevant for certain families of Cu-O-based materials, d -wave magnetism could be relevant.

Our work is organized as follows. We explore the emergence and stability of distinct AM phases summarized in Fig. 1 within the three-band Emery model, which we introduce in detail. We identify distinct mechanisms on how AFM correlations between oxygen moments can be stabilized, which we support by MFT and ED of finite clusters. We then propose a potential cuprate material realizing AM using density functional theory (DFT). Finally, we discuss the unique experimental properties of the putative d -wave magnetism like anisotropic spin transport and splitting of spin-wave branches observable via inelastic neutron scattering experiments.

II. ELECTRONIC STRUCTURE AND MODEL HAMILTONIAN

All cuprates feature CuO_2 planes with Cu^{2+} ions in a $3d^9$ configuration with a localized hole in the Cu $3d_{x^2-y^2}$ orbital. The parent compounds are charge-transfer insulators [32], where the top of the band formed by the oxygen p_x, p_y orbitals is located above the lower Hubbard band. This is well captured within the three-band Emery model [6,9,10]. Under doping, holes transfer to the oxygen ligands and at large doping a metallic state emerges [33] which is also consistent with DFT calculations [34–38].

In this work we concentrate on the three-band Emery model

$$\mathcal{H} = \sum_{i\mu\sigma} \epsilon_\mu n_{i\mu\sigma} + \sum_{i\mu j\nu\sigma} t_{i\mu j\nu} \mathbf{c}_{i\mu\sigma}^\dagger \mathbf{c}_{j\nu\sigma} + \sum_{i\mu} U_\mu n_{i\mu\uparrow} n_{i\mu\downarrow}, \quad (1)$$

where i, j label the square lattice unit cell, consisting of the $d_{x^2-y^2}$ Cu orbital ($\mu, \nu = d$) which is surrounded by O p_x and p_y orbitals ($\mu, \nu = p_x, p_y$). $\mathbf{c}_{i\mu\sigma}^\dagger$ ($\mathbf{c}_{j\nu\sigma}$) create (annihilate) holes with orbital μ (ν) and spin σ . $n_{i\mu\sigma}$ is the corresponding number operator. U_μ is the Hubbard-type Coulomb repulsion for orbital $\mu = d, p$. ϵ_μ is the orbital onsite energy, and $t_{i\mu j\nu}$ is the tunneling matrix element between $i\mu$ and $j\nu$. Figure 2(a) shows the dominant hopping parameters in the CuO_2 plane, where t_{pd} and t_{pp} are the nearest-neighbor Cu-O and O-O hoppings, respectively. The parameters in the hole picture are obtained as $\epsilon_d = 0$ eV, $\epsilon_p = 2.2$ eV, $t_{pd} = 1.3$ eV, and $t_{pp} = 0.6$ eV for La_2CuO_4 from projective Wannier functions [39] applied to nonrelativistic full potential local orbital (FPLO) calculations [40].¹ Considering the electronic structure of copper oxides, at charge neutrality the three-orbital unit cell includes five electrons and one hole per unit cell such that the d -electron band is half-filled. Hole doping is quantified by δ , e.g., 4.5 (4) electrons or 1.5 (2) holes per unit cell means $\delta = 0.5$ (1).

¹We extracted the parameters employing the Perdew-Burke-Ernzerhof generalized gradient approximation (GGA) [41] as exchange-correlation functional and a mesh of $9 \times 9 \times 5$ $\mathbf{k}$ points in the first Brillouin zone (FBZ).

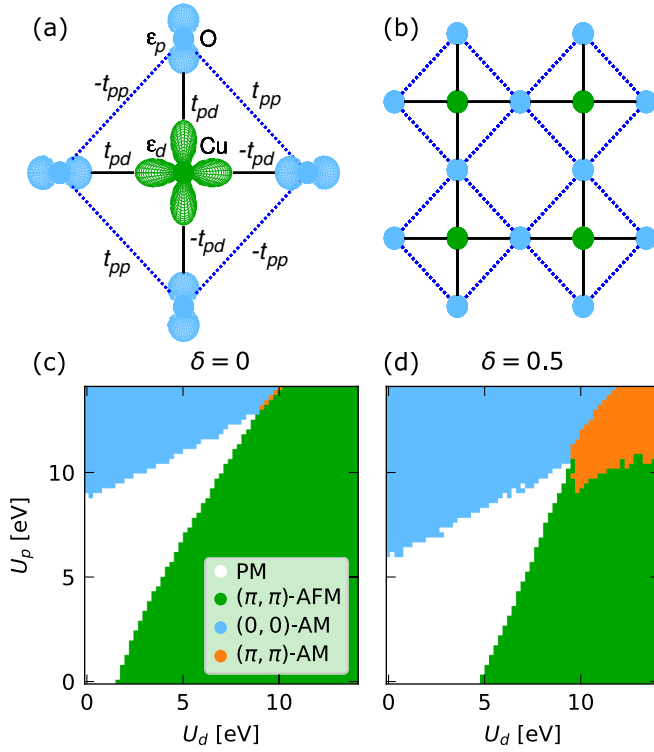


FIG. 2. Illustration of the hopping paths of the three-orbital Emery model (a), and the cluster considered in ED (b). MFT phase diagram for undoped (c) and hole-doped (d) case as a function of the Cu and O interaction strengths U_d and U_p . Hole doping is crucial to obtain a (π, π) -AM. It also reduces the critical interaction for the formation of moments on the O atoms, i.e., the $(0, 0)$ -AM. Parameters used, chosen to be realistic for cuprates [10,42,43], are $t_{pd} = 1.3$ eV, $t_{pp} = 0.6$ eV, and $\epsilon_p = 2.2$ eV; order parameter threshold 10^{-2} , tol 10^{-5} using a solver (see main text). Note that reasonable values of U_d in cuprates are from 7 to 10.5 eV while values from U_p are 4 to 6 eV [10].

III. AM DRIVEN BY REPULSION ON OXYGEN U_p

In general, the presence of AM in the cuprate planes requires a magnetic moment on the oxygen, thus, not full but only partial occupation, and a finite Hubbard repulsion U_p . It naturally appears in the negative charge transfer limit $\epsilon_p < 0$ at half-filling, when the oxygen orbitals are above the Cu d orbitals and their partial occupation then enables oxygen AFM once a small Hubbard repulsion U_p is present. Consequently $(0, 0)$ -AM sets in.

Interestingly, AM can also become stable in the far more realistic *positive* charge transfer regime $\epsilon_p > 0$. It has been shown that already for a low but still realistic charge transfer energy of $\epsilon_p = 2.2$ eV,² the magnetic moment calculated for the Emery model at half-filling has a 37% contribution from oxygen electrons [8]. This nonzero occupancy can be further enhanced by lowering ϵ_p or by increasing hole doping δ (see below). Naturally, a finite occupancy of oxygen orbitals only triggers AM once the Hubbard repulsion U_p is strong enough.

²And for infinitely large copper repulsion U_d and realistic oxygen-oxygen hopping $t_{pp} = 0.6$ eV.

Therefore, in the following we explore U_p dependence of the phase diagram of the Emery model in the realistic positive charge transfer limit. To this end, we elucidate U_p -driven oxygen AFM within a standard MFT.

In the MFT we first split the Hamiltonian $\mathcal{H}$, Eq. (1), into a noninteracting part $\mathcal{H}_0$ and an interacting part $\mathcal{H}_U$, such that $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_U$. The noninteracting Hamiltonian $\mathcal{H}_0$ can be diagonalized by Fourier transformation, $c_{k\mu\sigma} = N^{1/2} \sum_i e^{-ikR_{i\mu}} c_{i\mu\sigma}$, and $R_{i\mu}$ is the position of orbital μ in the unit cell i . We obtain $\mathcal{H}_0 = \sum_{k,\sigma} (c_{kd\sigma}, c_{kx\sigma}, c_{ky\sigma})^\dagger h_0(k) (c_{kd\sigma}, c_{kx\sigma}, c_{ky\sigma})$ where

$$h_0(k) = \begin{pmatrix} \epsilon_d & 2it_{pd}s_x & -2it_{pd}s_y \\ -2it_{pd}s_x & \epsilon_p & 4t_{pp}s_xs_y \\ 2it_{pd}s_y & 4t_{pp}s_xs_y & \epsilon_p \end{pmatrix} \quad (2)$$

with $s_{x(y)} = \sin(k_{x(y)}/2)$. We focus on $(0, 0)$ and (π, π) instabilities and introduce two sublattices $\lambda = A, B$ labeling the two copper atoms in the two copper unit cells. We then combine the fermion operators in a 12-component vector $\Psi_{\mathbf{k}}$ (with indices explicitly written as $\Psi_{\mathbf{k}\mu\lambda\sigma}$).

We define the mean fields $m_d = \sum_i (-1)^i \langle S_{id}^z \rangle / N$, i.e., the staggered magnetization of the Cu atoms, and $m_p = \sum_i \langle (S_{ix}^z - S_{iy}^z) \rangle / N$, the intra-unit-cell staggered magnetization of the p orbitals. Note that by default there are two p orbitals with opposite magnetization in the Wigner-Seitz cell. To complete our *Ansatz*, we also need to account for an interaction-induced change of the relative occupation of the orbitals. By proper renormalization of the chemical potential, it can be incorporated by introducing the third mean field $n_d = \sum_{i,\sigma} \langle c_{id\sigma}^\dagger c_{id\sigma} \rangle / N$ as the filling of the d orbitals in analogy to the overall filling n . Decoupling the Hamiltonian in the charge channel, we find

$$\begin{aligned} \mathcal{H}_U = & \frac{U_d N}{8} (m_d^2 - n_d^2) + \frac{U_p N}{16} (m_d^2 - (n - n_d)^2) \\ & + \sum_{k,\lambda,\sigma} \frac{U_d}{4} (n_d - (-1)^{\lambda+\sigma} m_d) c_{kd\lambda\sigma}^\dagger c_{kd\lambda\sigma} \\ & - \frac{U_p}{8} (-1)^\sigma m_p (c_{kx\lambda\sigma}^\dagger c_{kx\lambda\sigma} - c_{ky\lambda\sigma}^\dagger c_{ky\lambda\sigma}). \end{aligned} \quad (3)$$

The resulting MFT Hamiltonian $\sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger h(\mathbf{k}) \Psi_{\mathbf{k}} + E_0$ [see Appendix A, Eq. (A4)] is a noninteracting Hamiltonian. The 12×12 Bloch Hamiltonian $h(\mathbf{k})$ can be efficiently diagonalized for each momentum to obtain the eigenenergies $\epsilon_m(\mathbf{k})$ and the Bloch eigenstates $|u_m(\mathbf{k})\rangle$ with band index m . Note that H is block diagonal in spin, but in contrast to the conventional AFM spin density mean-field solution [44] the two blocks are explicitly spin dependent. We have solved the mean-field equations self-consistently for fixed filling n , i.e., for $\mathcal{M} = (m_d, m_p, n_d, \mu)$ we calculated $\mathcal{M}_r$ from $H(\mathcal{M}_{r-1})$ until $r = 100$, and then applied a python in-built solver which solves the fix point equation self-consistently.

The resultant MFT results are displayed in Figs. 2 and 3 where we find wide regions with both types of AM states. The effect of different parameters is summarized in Table I. First, we observe that the key driving term is the interaction U_p on the oxygen orbital, which in conjunction with a direct hopping t_{pp} generates a strong AFM exchange between the oxygen moments. However, in order to avoid a full occupation

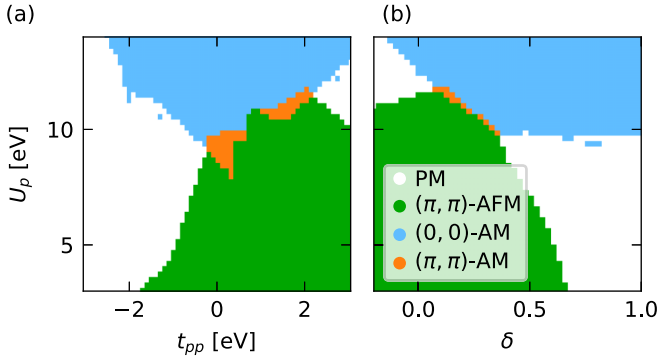


FIG. 3. MFT phase diagram as function of oxygen-oxygen hopping t_{pp} at doping $\delta = 0.5$ (a), and as function of hole doping δ at $t_{pp} = 0.6$ eV (b) [the chosen parameters, realistic for cuprates [10,42,43], are $t_{pd} = 1.3$, $\epsilon_p = 2.2$, and $U_d = 8$ eV; order parameter threshold 10^{-2} , tol 10^{-5} using a solver (see main text)].

of the p orbitals, nonzero doping is required or a raise of the onsite energies ϵ_p . At zero doping the single hole is always located entirely on the d orbital or entirely on the p orbital, no (π, π) -AM forms. Finite hole doping allows to distribute the additional hole charge between the otherwise competing orbitals which becomes visible by the formation of a (π, π) -AM state, at an increasing critical U_d and a decreasing critical U_p . Hole doping therefore strengthens the AM phases and weakens the AFM phase [see Fig. 3(b)].

The fact that oxygen and copper are competing for hole charge in order to form a magnetic moment is also evidenced by the observation that the critical U_d for AFM increases when U_p is increased and, vice versa, the critical U_p for $(0, 0)$ -AM increases when U_d is increased.

Beyond a basic direct exchange picture, the dependence on t_{pp} indicates the importance of a copper-mediated oxygen-oxygen superexchange. The MFT results [see Fig. 3(a)] show that oxygen moments form even for $t_{pp} = 0$ and the lowest critical U_p is found for small positive t_{pp} . Note that the phase diagram is not symmetric under sign changes of t_{pp} because for negative t_{pp} the FS has an increased contribution of the p orbitals and has not only d -orbital character.

IV. AM DRIVEN BY OXYGEN-OXYGEN HOPPING t_{pp}

While the MFT has shown that AM states can be stabilized, we next explore the phase diagram within ED calculations. We confirmed the U_p -driven AM observed in MFT, but interestingly find another route to intra-unit-cell oxygen AFM that does not depend on U_p . Instead, it can be triggered by oxygen-oxygen hopping t_{pp} . As this subtle effect depends

TABLE I. Summary of the parameter regimes supporting AFM correlations on the *oxygens* as obtained from ED compared to MFT.

	MFT	ED
t_{pp}	≈ 0 or small but positive	$ t_{pp} $ large
U_d	Small	Weak dependence
U_p	Large U_p crucial	Weak dependence
δ	$\delta > 0$ crucial	$\delta > 0$
ϵ_p	Small (as negative as possible)	

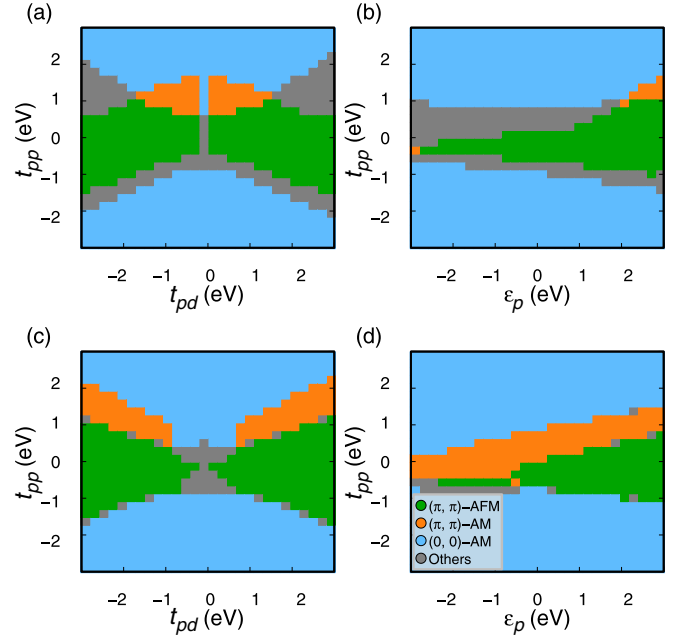


FIG. 4. Phase diagram from ED ($U_d = 8$ eV, $U_p = 4$ eV) for zero doping (4 holes) as a function of (a) t_{pd} and t_{pp} ($\epsilon_p = 2.2$ eV), as well as (b) ϵ_p and t_{pp} ($t_{pd} = 1.3$ eV). (c), (d) The corresponding cases for doping $\delta = 0.25$ (5 holes). All fixed parameters correspond to realistic values for cuprates [10,42,43].

on the nature of the correlated many-body wave function, it appears in ED but not simple MFT and can be accounted for within cell perturbation theory.

Using the three-orbital Emery model Hamiltonian [Eq. (1)] we performed ED for a cluster of four Cu and eight O with periodic boundary conditions at zero doping (4 holes) as well as $\delta = 0.25$ (5 holes), shown in Fig. 2(b). We used $U_d = 8$ eV and $U_p = 4$ eV. We note that for our small ED cluster the resulting ground states do not exhibit a finite magnetization since they cannot spontaneously break symmetries. Instead, we characterize the phases by measuring the spin-spin correlations $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ for the nearest-neighbor Cu-Cu and O-O sites (see Appendix B) to diagnose the (π, π) -AFM, $(0, 0)$ -AM, and (π, π) -AM states shown in Fig. 1. If spin correlations are incompatible with our *Ansätze*s, phases are labeled as *others*.

The obtained phase diagram for zero doping is displayed in Fig. 4(a) for t_{pd} and t_{pp} ($\epsilon_p = 2.2$ eV) as well as (b) ϵ_p and t_{pp} ($t_{pd} = 1.3$ eV). Figures 4(c) and 4(d) are corresponding results for $\delta = 0.25$. Similar to the MFT results, doping could enlarge the (π, π) -AM region. The results differ in two key aspects from the MFT results. First, we can identify t_{pp} as the main driving term of AM [see Fig. 4(a)]. Second, and in strong contrast to the MFT result, the formation of oxygen moments depends only weakly on U_p . It points towards a different mechanism driven by the hopping t_{pp} instead of the interaction U_p , which we discuss below.

The sign of t_{pd} does not affect the phase diagram while ϵ_p and t_{pp} are essential. When the magnitude of t_{pp} is small, the phase is a (π, π) -AFM, while increasing t_{pp} can enhance AFM correlations between oxygens and reduce them between coppers, leading to the transition to (π, π) -AM, and then to $(0, 0)$ -AM. When t_{pp} is fixed, smaller (as negative as possible)

ϵ_p increases the hole contribution of oxygen, inducing AM correlations.

The main result obtained by ED is that in the limit of realistic charge transfer energy ϵ_p , oxygen-copper hopping t_{pd} , and oxygen repulsion U_p , AFM correlations between oxygens already appear at half-filling, i.e., one hole per CuO_2 unit cell, for only slightly enhanced values of t_{pp} . Moreover, in this regime oxygen magnetism is independent of the value of the oxygen repulsion U_p ; instead it is triggered by the oxygen-oxygen hopping t_{pp} .

In Table I we summarize the parameter regimes supporting AFM correlations on the oxygens (and therefore AM correlations) obtained from our ED calculations compared to MFT.

The ED findings that AFM correlations on oxygens can strongly depend on t_{pp} but only weakly on U_p are rather counterintuitive. Interestingly, however, for the (0, 0)-AM case, this can be partially rationalized by noting that there exist spin-exchange processes between magnetic moments on oxygens that do *not* depend on U_p . In fact, in the limit of large $|\epsilon_p|$, U_p , U_d and no occupancy on copper sites, following [45], we deduce that the nearest-neighbor spin exchange between oxygens has three lowest-order contributions: (i) a dominant second-order term proportional to t_{pp}^2/U_p , (ii) third-order terms proportional to $t_{pd}^2 t_{pp}/\epsilon_p^2$ and $t_{pd}^2 t_{pp}/(|\epsilon_p|U_p)$, and (iii) fourth-order terms proportional to $t_{pd}^4/[\epsilon_p^2(U - 2\epsilon_p)]$ and $t_{pd}^4/[\epsilon_p^2(\epsilon_p + U_p)]$. Hence, in the strong-coupling perturbative limit, there are indeed spin-exchange processes whose strength does not depend on U_p . Naturally, what remains to be better understood is how such a strong-coupling result persists when ϵ_p is *not* large and negative, and the assumption of empty copper sites no longer holds. We leave this investigation for future work.

In contrast, the (π, π) -AM case presents a far more complex situation, as it requires understanding how the two seemingly competing AFM orders, on oxygen and copper, can coexist. In the following we provide a qualitative picture motivated by a cell perturbation theory [8]. Following Ref. [8] we can rewrite the charge-transfer model (1) in a different oxygen basis. We define the bonding and antibonding combinations of the four nearest-neighbor oxygen orbitals (labeled by L, R, T, B) surrounding the copper d orbital and obtain $|\alpha\rangle \equiv (|p_{xR}\rangle - |p_{yT}\rangle - |p_{xL}\rangle + |p_{yB}\rangle)/2$ and $|\beta\rangle \equiv (|p_{xR}\rangle + |p_{yT}\rangle - |p_{xL}\rangle - |p_{yB}\rangle)/2$. Interestingly, we can discard the oxygen repulsion U_p completely such that the Hamiltonian takes the form

$$\begin{aligned} \mathcal{H} = & \epsilon_p \sum_{i,\sigma} (\alpha_{i,\sigma}^\dagger \alpha_{i,\sigma} + \beta_{i,\sigma}^\dagger \beta_{i,\sigma}) + 2 \sum_{i,j(i),\sigma} [t_{pd}\mu(j)c_{id\sigma}^\dagger \alpha_{j\sigma} \\ & + t_{pp}\nu(j)(\alpha_{i\sigma}^\dagger \alpha_{j\sigma} - \beta_{i\sigma}^\dagger \beta_{j\sigma}) + t_{pp}\chi(j)\alpha_{i\sigma}^\dagger \beta_{j\sigma} + \text{H.c.}] \\ & + \sum_i U_{id} n_{id\uparrow} n_{id\downarrow}, \end{aligned} \quad (4)$$

where $\alpha_{i,\sigma}^\dagger$ and $\beta_{i,\sigma}^\dagger$ create holes in orthogonalized orbitals α and β centered around copper site i and $\mu(j)$, $\nu(j)$, $\chi(j)$ are the orthogonalization factors that strongly depend on the distance between the CuO_4 cells j (see [8] and Table II in Appendix D). The crucial point is that the hybridization strength $\chi(j)$ does not have an onsite component [i.e., $\chi(0) = 0$], though it couples nearest-neighbor orbitals α and β .

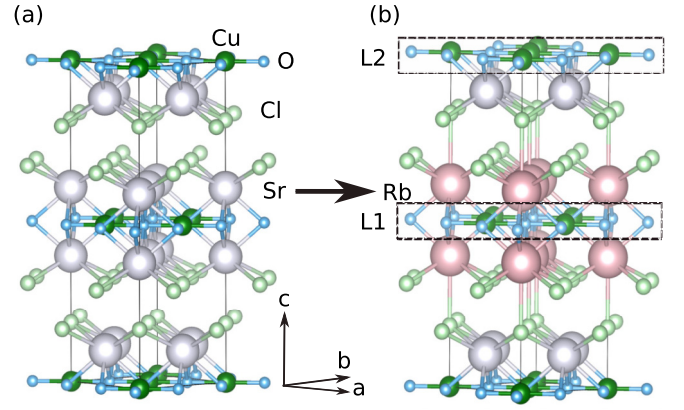


FIG. 5. Crystal structure of (a) $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ and (b) $\text{SrRbCuO}_2\text{Cl}_2$. L1 and L2 label two CuO_2 layers.

In most treatments of the charge-transfer model (4) the β orbitals are neglected leading to a two-band model. This, *inter alia*, gives rise to the well-studied AFM superexchange processes and the Zhang-Rice singlets upon doping the cuprates [7]. However, our key observation is that neglecting the β orbitals is no longer justified once $|t_{pp}|$ is larger than assumed in most cuprate works and once the β orbital is not fully occupied. It is the nearest-neighbor hybridization between oxygen orbitals that induces oxygen AFM.

Then a simple picture emerges best explained in a minimal toy model. From Eq. (4) we concentrate on two plaquettes (one of each A and B sublattices) and two holes. First, for infinite Coulomb repulsion on the copper d orbital strong superexchange leads to copper AFM [8], e.g., plaquette A spin up in the d orbital and plaquette B spin down. Next, the standard local hybridization between d and α orbitals leads to AFM correlations between plaquettes admixing oxygen states to the magnetic moment [8]. Going beyond this picture, we take into account the nearest-neighbor α, β hybridization. An up spin in α orbitals of site A induces an up spin in β orbitals at site B and vice versa. As a result, the spin of bonding and antibonding oxygen orbitals is antialigned in each plaquette (thus aligned between neighboring plaquettes). The hybridization between the nearest-neighbor α, β orbitals is directly proportional to t_{pp} and when calculating spin correlations in this state between the original oxygen ligands in the unit cell they are indeed AFM.

V. AM DRIVEN BY LARGE DOPING IN A MATERIAL CANDIDATE

Finding large values of U_p and t_{pp} in cuprate materials is not straightforward, therefore, we considered a few candidates for AM using *ab initio* DFT-based calculations. The first candidate is the widely studied cuprate La_2CuO_4 , where it is difficult to obtain in-plane oxygen moments since the extra holes from doping prefer to stay in apices O locations. We therefore turn our attention to the compound $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ [46], where the out-of-plane oxygen ions at the apices of the CuO_6 octahedra are replaced by Cl. In addition, a smaller ϵ_p has also been proposed [42,43].³

³See Table I of Ref. [43], i.e. the preprint version of Ref. [42].

In Fig. 5(a) we show the tetragonal structure of $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ with space group $I4/mmm$. Unlike La_2CuO_4 , no long-range tilting or rotation of the CuO_4Cl_2 octahedra is present. The structure leads to a very small magnetic anisotropy [47–50] and very weak interlayer coupling [50], making $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ a better two-dimensional AFM insulator [50–52]. Both inelastic neutron and light scattering measurements found a high-energy continuum of magnetic fluctuations [53] and multiple-magnon excitations [54]. Similar to La_2CuO_4 , the valence of the Cu atoms is +2, with one hole per copper atom. In order to have oxygen magnetic moments, we replaced half of the Sr atoms by Rb so that the system $\text{SrRbCuO}_2\text{Cl}_2$ [see Fig. 5(b)] is hole doped and becomes metallic. The CuO_4 layer neighbor to Rb (Sr) is labeled as L1 (L2). These two layers capture different dopings, as discussed in the next paragraph. The structure was fully relaxed with GGA using the Vienna *ab initio* simulation package (VASP) [55,56]. Total energies for various spin configurations and corresponding band structures were calculated with GGA + U . Band structure plots were prepared using VASPKIT [57]. We considered contributions of the Coulomb repulsion [58] $U_{\text{eff}} = U_d - J_H = 8$ eV ($U_d = 8$ eV, $J_H = 0$ eV) for Cu and $U_p = J_H = 0$ eV for O following [59] within GGA + U . Note that in the Dudarev's implementation of GGA + U , U and J_H are implemented as $U_{\text{eff}} = U_d - J_H$, therefore, the combinations of U_d and J_H with the same U_{eff} would not change the results. Here we observe that in GGA + U , a finite U stabilizes the magnetic moments of Cu.

By comparing the energy for various magnetic configurations [Cu AFM order, (π, π) -AM order, and $(0, 0)$ -AM order] we find that (π, π) -AM order is 241.5 meV lower than $(0, 0)$ -AM and 53.9 meV lower than (π, π) -AFM. The magnetic moments are $0.6\mu_B$ for Cu, similar to La_2CuO_4 [38]. However, in contrast to La_2CuO_4 , which has a small oxygen moment on the apical oxygen [38] but no doping-induced oxygen magnetism in the CuO_2 plane, in $\text{SrRbCuO}_2\text{Cl}_2$ the magnetic moments are $0.4\mu_B$ for oxygen. We integrated the partial density of states for Cu and O and find that the additional four holes from doping are mostly localized at the four O in the L1 layer shown in Fig. 6(b) with doping $\delta = 2$ (3 electrons, 3 holes per CuO_2).

The resulting band structure within GGA + U ($U_d = 8$ eV, $U_p = 0$ eV) obtained from DFT is displayed in Fig. 6 for (π, π) -AM order, $(0, 0)$ -AM order, and (π, π) -AFM order. The bands have opposite spin-splitting signs along Γ -X and Γ -Y. We stress that, different from the case of La_2CuO_4 , where AM originates from CuO_6 octahedra distortions, in $\text{SrRbCuO}_2\text{Cl}_2$ such distortions are absent and AM arises from oxygen moments.

VI. CONCLUSION

We have studied d -wave magnetism in cuprates induced by magnetic moments on the oxygen atoms. Focusing on the three-orbital Emery model we employed Hartree-Fock MFT, ED of finite clusters, and cell perturbation theory to uncover distinct mechanisms stabilizing AFM correlations between oxygen moments. Two different AM states can be stabilized. First, a $(0, 0)$ -AM with AFM ordered moments only on the

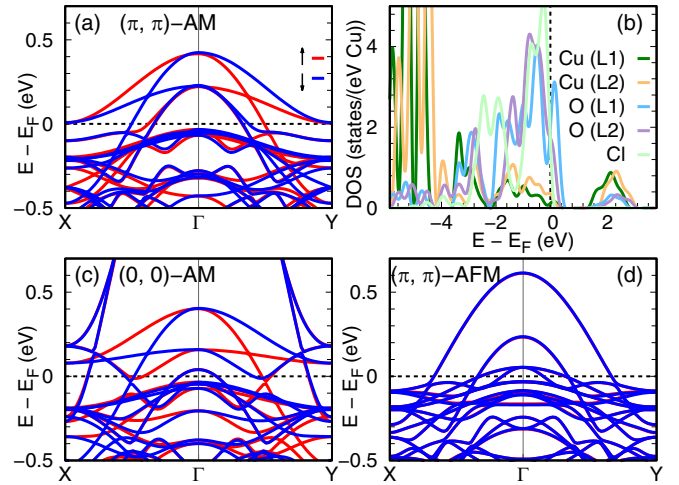


FIG. 6. (a) Band structure and (b) density of states for $\text{SrRbCuO}_2\text{Cl}_2$ in the (π, π) -AM magnetic order within GGA + U from DFT as shown in Fig. 1. Band structure in the (c) $(0, 0)$ -AM magnetic order and (d) (π, π) -AFM order are also shown. Colors red (spin $\uparrow$) and blue (spin $\downarrow$) indicate the up bands and down states, respectively. The AM ordering is clear from the spin splitting along Γ -X and Γ -Y.

oxygen sites, such that the magnetic unit cell is identical with the crystal unit cell. The Fermi surface is characterized by a large spin splitting of the formerly (π, π) centered pocket into open Fermi surfaces along the crystallographic axis (see Fig. 1). Interestingly, this intra-unit-cell magnetic order has been proposed previously to explain inelastic neutron scattering data [16,17]. Second, we find a (π, π) -AM with AFM order of the oxygens as above coexisting with the standard (π, π) magnetic order on the copper atoms. Here, we observe a Fermi surface with two closed pockets which are additionally weakly spin split by opposite tilting (see Fig. 1).

Our work shows that while AM states in cuprates can be stabilized, the required parameter regimes for t_{pp} and U_p are challenging to obtain for typical cuprates. The exact form of the phase boundaries is method dependent, e.g., MFT favoring large U_p while our ED of small clusters points to the relevance of t_{pp} . Despite these challenges, we identify parameters which are potentially relevant for certain cuprate families. Based on our observations, we then identify three distinct mechanisms potentially realizing AM in cuprates: (i) a direct exchange between the oxygens requiring a large oxygen interaction U_p , (ii) strong charge transfer, i.e., ϵ_p small, bringing the p -oxygen orbital closer to the Fermi energy, and (iii) a large oxygen-oxygen hopping t_{pp} effectively such that the magnetic order of the copper atoms induces magnetic order on oxygen sites. We note that *ab initio* calculations of Ref. [42], and in particular Table I available in [43], suggest that a number of cuprates have surprisingly low charge-transfer energy $\epsilon_p < 2$ eV and a large oxygen-oxygen hopping $t_{pp} \sim 0.6$ – 0.7 eV. Further indications of the substantial values of the oxygen-oxygen hopping in the cuprates are discussed in, e.g., Refs. [60–62].

Beyond our proof-of-principle calculations on the minimal Emery model, we also investigated the realization of AM in a novel candidate material $\text{SrRbCuO}_2\text{Cl}_2$ based on the well-known existing system $\text{Sr}_2\text{CuO}_2\text{Cl}_2$. The DFT results

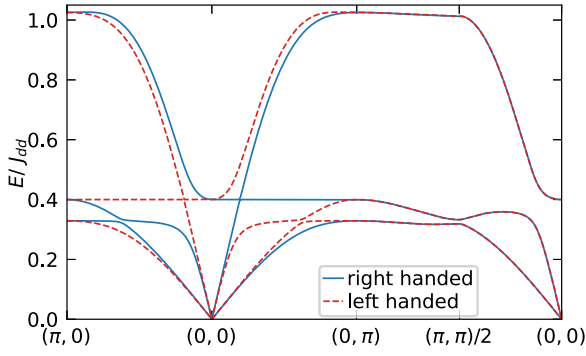


FIG. 7. Magnon spectrum along a high-symmetry path through the magnetic Brillouin zone, determined from linear spin-wave theory of an effective Heisenberg-type spin-only model with moments on the oxygens and copper atoms for the (π, π) -AM state [see Eq. (C1) in Appendix C]. The different chiralities (left and right precessing) are distinguished by color and line style. Note, we have used an artificially large splitting for presentation: $[J_{pd} = 0.5J_{dd}$ and $J_{pp} = 0.4J_{dd}]$.

suggest the (π, π) -AM order to be the most stable one emerging from AFM correlations between oxygen moments. Since the AM in our case is induced by magnetism on the p orbitals, detailed DFT calculations for each material are required. As this is computationally very involved, we leave it for a next study to do a database materials.

The prospect of AM in doped cuprates raises the question on how it would manifest in experiments. Apart from the standard spin-transport signatures predicted for AM [20,21,63], angle-resolved photoemission spectroscopy (ARPES) or spin ARPES [64] could directly probe the spin splitting of the Fermi surface. Alternatively, the split FS can be detected by quantum oscillation measurements which have a long history in underdoped cuprates [65,66]. Interestingly, since Zeeman coupling will lead to different areas for the spin-polarized FSs, the temperature dependence of QO can be unconventional [67,68] and new low-frequency oscillations could appear [69,70]. Most promising would be to revisit previous inelastic neutron scattering experiments [16,17]. Magnon spectra of AM display chirality split magnons away from high-symmetry points. The splitting is potentially small and requires high resolution. In Fig. 7 we show predictions for magnon spectra with chirality split magnon bands. Note, we treated a simple Heisenberg-type model of spins on copper and oxygen sites (see Appendix C for details), and the optical magnon modes from intra-unit-cell excitations are strongly split, but because of the weak oxygen moment might be hard to observe. Finally, AM in cuprates or other related compounds would have important ramifications for

superconductivity. AM have been shown to lead to unconventional phenomena [71–75] such as pair density states (PDW) with finite-momentum pairing [76–79]. Especially the $(0, 0)$ -AM offers, due to its large spin splitting and absence of magnetism for the copper sites, an interesting platform for realizing PDWs. Searching for signatures thereof in experiment and understanding the microscopic mechanism for its realization in realistic settings will be important directions for future research.

Note added. Recently, we became aware of two related independent works. Dürrnagel *et al.* [80] explored an itinerant AM phase transition emanating from an electronic model on a related Lieb lattice [81], where AM originates from sublattice interference of the enlarged unit cell. Kaushal *et al.* [45] studied itinerant AM in a Lieb lattice Hubbard model for a different parameter regime of an “anti- CuO_2 ” lattice.

ACKNOWLEDGMENTS

We thank M. Fisher, K. Plumb, A. Mook, and I. I. Mazin for insightful discussions. Y.L. acknowledges support from the Alexander von Humboldt Foundation through a post-doctoral Humboldt fellowship and Fundamental Research Funds for the Central Universities (Grant No. xzy012023051). V.L. acknowledges support from the Studienstiftung des deutschen Volkes. J.K. acknowledges support from the Imperial-TUM flagship partnership. R.V. acknowledges support by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) for funding through Project No. TRR 288–422213477 (Project No. A05, B05) and Project No. VA 117/23-1–509751747. K.W. acknowledges the support of National Science Centre in Poland under Project No. 2024/55/B/ST3/03144. Collaboration between J.K., L.V., and K.W. was supported by the Tandems for Excellence: Visiting Researchers Programme of the University of Warsaw.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: MEAN-FIELD THEORY

The 12×12 Hamiltonian is block diagonal in spin. We consider the ordered basis

$$\psi_{\mathbf{k}} = (c_{kA\uparrow}, c_{kx\uparrow}, c_{ky\uparrow}, c_{kB\uparrow}, c_{kx\downarrow}, c_{ky\downarrow}, c_{kA\downarrow}, c_{kx\downarrow}, c_{ky\downarrow}, c_{kB\downarrow}, c_{kx\downarrow}, c_{ky\downarrow})^T. \quad (\text{A1})$$

The matrix elements are

$$h(\mathbf{k}) = \mathbb{1} \otimes \begin{pmatrix} h_{AA}(\mathbf{k}) & h_{AB}(\mathbf{k}) \\ h_{AB}(\mathbf{k})^\dagger & h_{AA}(\mathbf{k}) \end{pmatrix} - \frac{U_d m_d}{4} \tau^z \otimes \tau^z \otimes \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} + \frac{U_d n_d}{4} \mathbb{1} \otimes \mathbb{1} \otimes \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \\ - \frac{U_p m_p}{8} \tau^z \otimes \mathbb{1} \otimes \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix} + \frac{U_p (n - n_d)}{8} \mathbb{1} \otimes \mathbb{1} \otimes \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (\text{A2})$$

$$h_{AA}(\mathbf{k}) = \begin{pmatrix} 0 & -t_{pd}e^{-ik_x/2} & t_{pd}e^{-ik_y/2} \\ -t_{pd}e^{ik_x/2} & 0 & 2t_{pp}\cos\left(\frac{k_x-k_y}{2}\right) \\ t_{pd}e^{ik_y/2} & 2t_{pp}\cos\left(\frac{k_x-k_y}{2}\right) & 0 \end{pmatrix}, \quad (\text{A3})$$

$$h_{AB}(\mathbf{k}) = \begin{pmatrix} 0 & t_{pd}e^{-ik_x/2} & -t_{pd}e^{-ik_y/2} \\ t_{pd}e^{ik_x/2} & 0 & -2t_{pp}\cos\left(\frac{k_x+k_y}{2}\right) \\ -t_{pd}e^{ik_y/2} & -2t_{pp}\cos\left(\frac{k_x+k_y}{2}\right) & 0 \end{pmatrix}, \quad (\text{A4})$$

and τ^z is the Pauli- z matrix.

APPENDIX B: SPIN-SPIN CORRELATIONS FROM ED

Figure 8 displays the spin-spin correlations for ED of four-copper-sites cluster using $U_d = 8$ eV, $U_p = 4$ eV. Note that here we neglect the small positive values for Cu-Cu correlation.

APPENDIX C: MAGNON SPECTRUM

1. Model

We study the effective Heisenberg-type spin-only model

$$H_{\text{spin}} = \sum_{\langle ij \rangle} J_{\alpha\beta} \mathbf{S}_{i\alpha} \mathbf{S}_{j\beta} \\ = \sum_{\langle ij \rangle} J_{\alpha\beta} \left(S_{i\alpha}^z S_{j\beta}^z + \frac{1}{2} S_{i\alpha}^+ S_{j\beta}^- + \frac{1}{2} S_{i\alpha}^- S_{j\beta}^+ \right) \quad (\text{C1})$$

which emerges in the large- U limit from Eq. (1) of the main text. It includes AFM exchange $J_{dd} > 0$ between the spin $\frac{1}{2}$ located at the positions of the Cu atoms, AFM exchange $J_{pp} > 0$ between the spin $\frac{1}{2}$ located at the positions of the O atoms, and an interaction J_{pd} between O and Cu moments. $S_{i\alpha}^z$ are the spin- $\frac{1}{2}$ operators and $S_{i\alpha}^{\pm}$ the raising and lowering operators.

While we take the spin model as an effective phenomenological model we can estimate the spin-exchange energy scales from perturbation theory:

(1) A textbook result (e.g., see [82]) gives for the positive charge-transfer limit: $J_{pd} = 2t_{pd}(\frac{1}{U_p + \epsilon_p} + \frac{1}{U})$.

(2) Similarly, in the positive charge-transfer limit, $J_{pp} = \frac{4t_{pp}^2}{U_p}$ cf. [45]. Note that the higher-order terms $\propto t_{pp}t_{pd}^2$ (i.e., going “on a O-Cu-O” triangle) can in principle be considered but they will (i) be smaller for the positive charge-transfer limit, and (ii) will depend on the occupancy of the “middle” copper sites (i.e., probably will vanish once this site is occupied, see below).

(3) Estimating J_{dd} is more difficult. Once the oxygen is *not* occupied, we obtain the textbook result $J_{dd} = \frac{4t_{pd}^4}{\epsilon_p^2}(\frac{2}{U_p + 2\epsilon_p} + \frac{1}{U})$ (valid in the positive charge-transfer limit) [7,82]. However, once the oxygen is occupied *and* assuming that the middle spin *does not* actively participate in the spin exchange,⁴ then there is *no* spin-dependent interaction (there

merely is a lowering of the total energy due to the virtual processes, however, these are spin independent). In fact, this result stays in line with what is written in [82] [see Eq. (5) in that paper].

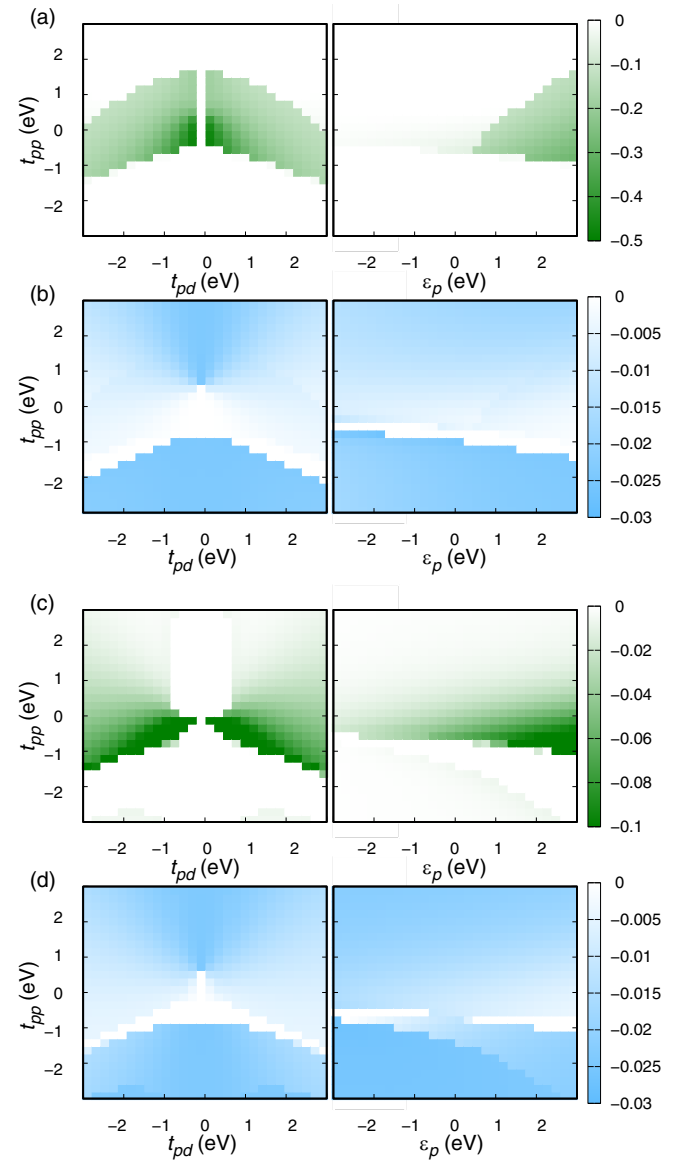


FIG. 8. Spin-spin correlations for zero doping with respect to t_{pd} and t_{pp} ($\epsilon_p = 2.2$ eV) as well as ϵ_p and t_{pp} ($t_{pd} = 1.3$ eV). (a) Nearest-neighbor Cu-Cu correlation ($\langle S_i^d S_j^d \rangle$). (b) Nearest neighbor O-O correlations ($\langle S_i^p S_j^p \rangle$). (c), (d) The corresponding cases for doping $\delta = 0.25$ (5 holes).

⁴This would lead to the three-spin interaction.

2. Linear spin-wave theory

We evaluate the magnon spectrum assuming the ordered ground state of the (π, π) -AM, shown in Fig. 1 of the main text. We introduce two sublattices λ , employ a Holstein-Primakoff transformation, and take the large- S limit. If the spin at i, α, λ is $\uparrow$ the transformation reads as

$$\begin{aligned} S_{i\alpha\lambda}^z &= S_\alpha - a_{i\alpha\lambda}^\dagger a_{i\alpha\lambda}, \\ S_{i\alpha\lambda}^+ &= \sqrt{2S_\alpha} a_{i\alpha\lambda}, \\ S_{i\alpha\lambda}^- &= \sqrt{2S_\alpha} a_{i\alpha\lambda}^\dagger \end{aligned} \quad (\text{C2})$$

and

$$\begin{aligned} S_{i\alpha\lambda}^z &= -S_\alpha + a_{i\alpha\lambda}^\dagger a_{i\alpha\lambda}, \\ S_{i\alpha\lambda}^+ &= \sqrt{2S_\alpha} a_{i\alpha\lambda}^\dagger, \\ S_{i\alpha\lambda}^- &= \sqrt{2S_\alpha} a_{i\alpha\lambda} \end{aligned} \quad (\text{C3})$$

for a $\downarrow$ spin. The operator $a_{i\alpha\lambda}$ ($a_{i\alpha\lambda}^\dagger$) annihilates (creates) a magnon, i.e., a spin flip, at i, α, λ .

Keeping only terms linear or quadratic in S_α

$$\begin{aligned} H_{\text{spin}} &= -4J_{dd}S_d^2 - 8J_{pp}S_p^2 + \sum_{\mathbf{k}} \eta_{\mathbf{k}L}^\dagger h_L(\mathbf{k}) \eta_{\mathbf{k}L} \\ &\quad + \eta_{\mathbf{k}R}^\dagger h_R(\mathbf{k}) \eta_{\mathbf{k}R} + O(S_\alpha^0) \end{aligned} \quad (\text{C4})$$

which simplifies for $S_\alpha = \frac{1}{2}$ to

$$H_{\text{spin}} = -J_{dd} - 2J_{pp} + \sum_{\mathbf{k}} \eta_{\mathbf{k}L}^\dagger h_L(\mathbf{k}) \eta_{\mathbf{k}L} + \eta_{\mathbf{k}R}^\dagger h_R(\mathbf{k}) \eta_{\mathbf{k}R}. \quad (\text{C5})$$

We defined $a_{\mathbf{k}\alpha\lambda}^\dagger = N^{-1/2} \sum_j e^{i\mathbf{k}\mathbf{R}_{j\alpha\lambda}} a_{j\alpha\lambda}^\dagger$ where $\mathbf{R}_{j\alpha\lambda}$ is the position of a site j, α, λ and the fields

$$\eta_{\mathbf{k}R} = (a_{\mathbf{k}dA}, a_{\mathbf{k}xA}, a_{\mathbf{k}xB}, a_{-\mathbf{k}dB}^\dagger, a_{-\mathbf{k}yA}^\dagger, a_{-\mathbf{k}yB}^\dagger), \quad (\text{C6})$$

$$\eta_{\mathbf{k}L} = (a_{\mathbf{k}dB}, a_{\mathbf{k}yA}, a_{\mathbf{k}yB}, a_{-\mathbf{k}dA}^\dagger, a_{-\mathbf{k}xA}^\dagger, a_{-\mathbf{k}xB}^\dagger). \quad (\text{C7})$$

The matrix elements are given by

$$h_R(\mathbf{k}) = \begin{pmatrix} h_{pd}(k_x) & h_{pd}(k_y) + h_0(\mathbf{k}) \\ h_{pd}^\dagger(k_y) + h_0(\mathbf{k}) & h_{pd}(-k_y) \end{pmatrix} + (J_{dd}, J_{pp}, J_{pp}, J_{dd}, J_{pp}, J_{pp})\mathbb{1}, \quad (\text{C8})$$

$$h_0(\mathbf{k}) = \begin{pmatrix} \frac{J_{dd}}{2}(\cos k_x + \cos k_y) & 0 & 0 \\ 0 & \frac{J_{pp}}{2} \cos \frac{k_x - k_y}{2} & \frac{J_{pp}}{2} \cos \frac{k_x + k_y}{2} \\ 0 & \frac{J_{pp}}{2} \cos \frac{k_x + k_y}{2} & \frac{J_{pp}}{2} \cos \frac{k_x - k_y}{2} \end{pmatrix}, \quad (\text{C9})$$

$$h_{pd}(k) = \frac{J_{pd}}{4} \begin{pmatrix} 0 & e^{ik/2} & e^{-ik/2} \\ e^{-ik/2} & 0 & 0 \\ e^{ik/2} & 0 & 0 \end{pmatrix} \quad (\text{C10})$$

and $h_L[(k_x, k_y)] = h_R[(-k_y, -k_x)]$. Note, however, that $h_0[(k_x, k_y)] = h_0[(-k_y, -k_x)]$ such that the matrices differ only in terms which are proportional to J_{pd} .

We diagonalize (C5) by a numerical Bogoliubov transformation, i.e., we multiply $h_{R,L}$ with the matrix $\mathbb{1}(1, 1, 1, -1, -1, -1)$ and then diagonalize this non-Hermitian matrix. The positive eigenvalues yield the magnon spectrum of (C1).

3. Results

The resulting magnon spectrum (see Fig. 7) features two Goldstone modes, a flat mode which can be associated with the ordering of the oxygen moments and a steep mode from the copper moments, because $J_{dd} > J_{pp}$. Additionally, an optical mode with oxygen orbital character appears due to the fact that the AFM ordering of the oxygen moments is intra-unit cell. We observe large chirality splitting of the magnon modes because we expect $J_{pd} > J_{pp}$. We expect that most of the spectral weight is located on the magnon branches with Cu orbital character. However, quantitative studies of the dynamic

spin structure factor of more microscopic model remains a formidable task for future.

APPENDIX D: VALUES OF THE ORTHOGONALIZATION FACTORS

The orthogonalization factors μ, ν, χ are given in Table II. As the values of the orthogonalization factors are strongly diminished with increased distance between the unit cells, only values for CuO₄ cells being up to third neighbor apart from each other are given in the table.

TABLE II. Dependence of the orthogonalization factors μ, ν, χ with distance between the CuO₄ cells j . Table adopted from Ref. [8].

j	μ	ν	χ
0	0.958	0.727	0
1	-0.14	-0.273	-0.133
$\sqrt{2}$	-0.02	0.122	0
2	-0.02	-0.064	0.041

- [1] J. G. Bednorz and K. A. Müller, Possible high- T_c superconductivity in the Ba-La-Cu-O system, *Z. Phys. B* **64**, 189 (1986).
- [2] P. W. Anderson, The resonating valence bond state in La_2CuO_4 and superconductivity, *Science* **235**, 1196 (1987).
- [3] C. Gros and R. Valentí, A self-consistent cluster study of the Emery model, *Ann. Phys.* **506**, 460 (1994).
- [4] E. Dagotto, Correlated electrons in high-temperature superconductors, *Rev. Mod. Phys.* **66**, 763 (1994).
- [5] M. Imada, A. Fujimori, and Y. Tokura, Metal-insulator transitions, *Rev. Mod. Phys.* **70**, 1039 (1998).
- [6] V. J. Emery, Theory of high- T_c superconductivity in oxides, *Phys. Rev. Lett.* **58**, 2794 (1987).
- [7] F. C. Zhang and T. M. Rice, Effective Hamiltonian for the superconducting Cu oxides, *Phys. Rev. B* **37**, 3759(R) (1988).
- [8] J. H. Jefferson, H. Eskes, and L. F. Feiner, Derivation of a single-band model for CuO_2 planes by a cell-perturbation method, *Phys. Rev. B* **45**, 7959 (1992).
- [9] C. Weber, T. Giamarchi, and C. M. Varma, Phase diagram of a three-orbital model for high- T_c cuprate superconductors, *Phys. Rev. Lett.* **112**, 117001 (2014).
- [10] K. Sheshadri, D. Malterre, A. Fujimori, and A. Chainani, Connecting the one-band and three-band Hubbard models of cuprates via spectroscopy and scattering experiments, *Phys. Rev. B* **107**, 085125 (2023).
- [11] M. H. Fischer and E.-A. Kim, Mean-field analysis of intra-unit-cell order in the Emery model of the CuO_2 plane, *Phys. Rev. B* **84**, 144502 (2011).
- [12] M. H. Fischer, S. Wu, M. Lawler, A. Paramakanti, and E.-A. Kim, Nematic and spin-charge orders driven by hole-doping a charge-transfer insulator, *New J. Phys.* **16**, 093057 (2014).
- [13] C. M. Varma, Theory of the pseudogap state of the cuprates, *Phys. Rev. B* **73**, 155113 (2006).
- [14] S. Chakravarty, R. B. Laughlin, D. K. Morr, and C. Nayak, Hidden order in the cuprates, *Phys. Rev. B* **63**, 094503 (2001).
- [15] A. C. Walters, T. G. Perring, J.-S. Caux, A. T. Savici, G. D. Gu, C.-C. Lee, W. Ku, and I. A. Zaliznyak, Effect of covalent bonding on magnetism and the missing neutron intensity in copper oxide compounds, *Nat. Phys.* **5**, 867 (2009).
- [16] B. Fauqué, Y. Sidis, V. Hinkov, S. Pailhes, C. T. Lin, X. Chaud, and P. Bourges, Magnetic order in the pseudogap phase of high- T_c superconductors, *Phys. Rev. Lett.* **96**, 197001 (2006).
- [17] Y. Li, V. Balédent, N. Barišić, Y. Cho, B. Fauqué, Y. Sidis, G. Yu, X. Zhao, P. Bourges, and M. Greven, Unusual magnetic order in the pseudogap region of the superconductor $\text{HgBa}_2\text{CuO}_{4+\delta}$, *Nature (London)* **455**, 372 (2008).
- [18] L. Šmejkal, R. González-Hernández, T. Jungwirth, and J. Sinova, Crystal time-reversal symmetry breaking and spontaneous Hall effect in collinear antiferromagnets, *Sci. Adv.* **6**, eaaz8809 (2020).
- [19] I. I. Mazin, K. Koepernik, M. D. Johannes, R. González-Hernández, and L. Šmejkal, Prediction of unconventional magnetism in doped FeSb_2 , *Proc. Natl. Acad. Sci. USA* **118**, e2108924118 (2021).
- [20] L. Šmejkal, J. Sinova, and T. Jungwirth, Emerging research landscape of altermagnetism, *Phys. Rev. X* **12**, 040501 (2022).
- [21] L. Šmejkal, J. Sinova, and T. Jungwirth, Beyond conventional ferromagnetism and antiferromagnetism: A phase with nonrelativistic spin and crystal rotation symmetry, *Phys. Rev. X* **12**, 031042 (2022).
- [22] L. Šmejkal, A. B. Hellènes, R. González-Hernández, J. Sinova, and T. Jungwirth, Giant and tunneling magnetoresistance in unconventional collinear antiferromagnets with nonrelativistic spin-momentum coupling, *Phys. Rev. X* **12**, 011028 (2022).
- [23] K.-H. Ahn, A. Hariki, K.-W. Lee, and J. Kuneš, Antiferromagnetism in RuO_2 as d -wave Pomeranchuk instability, *Phys. Rev. B* **99**, 184432 (2019).
- [24] S. Hayami, Y. Yanagi, and H. Kusunose, Momentum-dependent spin splitting by collinear antiferromagnetic ordering, *J. Phys. Soc. Jpn.* **88**, 123702 (2019).
- [25] L.-D. Yuan, Z. Wang, J.-W. Luo, E. I. Rashba, and A. Zunger, Giant momentum-dependent spin splitting in centrosymmetric low- Z antiferromagnets, *Phys. Rev. B* **102**, 014422 (2020).
- [26] H.-Y. Ma, M. Hu, N. Li, J. Liu, W. Yao, J.-F. Jia, and J. Liu, Multifunctional antiferromagnetic materials with giant piezomagnetism and noncollinear spin current, *Nat. Commun.* **12**, 2846 (2021).
- [27] V. Leeb, A. Mook, L. Šmejkal, and J. Knolle, Spontaneous formation of altermagnetism from orbital ordering, *Phys. Rev. Lett.* **132**, 236701 (2024).
- [28] I. Mazin and The PRX Editors, Editorial: Altermagnetism—A new punch line of fundamental magnetism, *Phys. Rev. X* **12**, 040002 (2022).
- [29] H. O. Jeschke, M. Shimizu, and I. I. Mazin, $\text{CuAg}(\text{SO}_4)_2$: A doubly strongly correlated altermagnetic three-dimensional analog of the parent compounds of high- T_c cuprates, *Phys. Rev. B* **109**, L220412 (2024).
- [30] A. Bose, S. Vadnais, and A. Paramakanti, Altermagnetism and superconductivity in a multiorbital $t - J$ Model, *Phys. Rev. B* **110**, 205120 (2024).
- [31] S. Giuli, C. Mejuto-Zaera, and M. Capone, Altermagnetism from interaction-driven itinerant magnetism, *Phys. Rev. B* **111**, L020401 (2025).
- [32] J. Zaanen, G. A. Sawatzky, and J. W. Allen, Band gaps and electronic structure of transition-metal compounds, *Phys. Rev. Lett.* **55**, 418 (1985).
- [33] S. Uchida, T. Ido, H. Takagi, T. Arima, Y. Tokura, and S. Tajima, Optical spectra of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$: Effect of carrier doping on the electronic structure of the CuO_2 plane, *Phys. Rev. B* **43**, 7942 (1991).
- [34] M. T. Czyżyk and G. A. Sawatzky, Local-density functional and on-site correlations: The electronic structure of La_2CuO_4 and LaCuO_3 , *Phys. Rev. B* **49**, 14211 (1994).
- [35] S. Pesant and M. Côté, DFT + U study of magnetic order in doped La_2CuO_4 crystals, *Phys. Rev. B* **84**, 085104 (2011).
- [36] J. W. Furness, Y. Zhang, C. Lane, I. G. Buda, B. Barbiellini, R. S. Markiewicz, A. Bansil, and J. Sun, An accurate first-principles treatment of doping-dependent electronic structure of high-temperature cuprate superconductors, *Commun. Phys.* **1**, 11 (2018).
- [37] M. Biemann, P. Schwaller, P. Ruffieux, O. Gröning, L. Schlapbach, and P. Gröning, AgO investigated by photoelectron spectroscopy: Evidence for mixed valence, *Phys. Rev. B* **65**, 235431 (2002).
- [38] C. Lane, J. W. Furness, I. G. Buda, Y. Zhang, R. S. Markiewicz, B. Barbiellini, J. Sun, and A. Bansil, Antiferromagnetic ground state of La_2CuO_4 : A parameter-free *ab initio* description, *Phys. Rev. B* **98**, 125140 (2018).
- [39] H. Eschrig and K. Koepernik, Tight-binding models for the iron-based superconductors, *Phys. Rev. B* **80**, 104503 (2009).

- [40] K. Koepnick and H. Eschrig, Full-potential nonorthogonal local-orbital minimum-basis band-structure scheme, *Phys. Rev. B* **59**, 1743 (1999).
- [41] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [42] C. Weber, C. Yee, K. Haule, and G. Kotliar, Scaling of the transition temperature of hole-doped cuprate superconductors with the charge-transfer energy, *Europhys. Lett.* **100**, 37001 (2012).
- [43] C. Weber, C.-H. Yee, K. Haule, and G. Kotliar, Scaling of the transition temperature of hole-doped cuprate superconductors with the charge-transfer energy, [arXiv:1108.3028](https://arxiv.org/abs/1108.3028).
- [44] J. Knolle, I. Eremin, and R. Moessner, Multiorbital spin susceptibility in a magnetically ordered state: Orbital versus excitonic spin density wave scenario, *Phys. Rev. B* **83**, 224503 (2011).
- [45] N. Kaushal and M. Franz, Altermagnetism in modified Lieb lattice Hubbard model, [arXiv:2412.16421](https://arxiv.org/abs/2412.16421).
- [46] L. L. Miller, X. L. Wang, S. X. Wang, C. Stassis, D. C. Johnston, J. Faber, and C.-K. Loong, Synthesis, structure, and properties of $\text{Sr}_2\text{CuO}_2\text{Cl}_2$, *Phys. Rev. B* **41**, 1921 (1990).
- [47] A. Cuccoli, T. Roscilde, R. Vaia, and P. Verrucchi, Detection of *XY* behavior in weakly anisotropic quantum antiferromagnets on the square lattice, *Phys. Rev. Lett.* **90**, 167205 (2003).
- [48] B. J. Suh, F. Borsa, L. L. Miller, D. C. Johnston, D. R. Torgeson, and M. Corti, Evidence for crossover effects in the spin dynamics of the two-dimensional antiferromagnet $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ from ^{35}Cl nuclear magnetic resonance, *J. Appl. Phys.* **79**, 5084 (1996).
- [49] K. Katsumata, M. Hagiwara, Z. Honda, J. Satooka, A. Aharony, R. J. Birgeneau, F. C. Chou, O. Entin-Wohlman, A. B. Harris, M. A. Kastner, Y. J. Kim, and Y. S. Lee, Direct observation of the quantum energy gap in $S = 1/2$ tetragonal cuprate antiferromagnets, *Europhys. Lett.* **54**, 508 (2001).
- [50] B. O. Wells, Z.-X. Shen, A. Matsuura, D. M. King, M. A. Kastner, M. Greven, and R. J. Birgeneau, *E* versus *k* relations and many body effects in the model insulating copper oxide $\text{Sr}_2\text{CuO}_2\text{Cl}_2$, *Phys. Rev. Lett.* **74**, 964 (1995).
- [51] A. N. Yaresko, A. Y. Perlov, R. Hayn, and H. Rosner, Exchange integrals of $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ and $\text{Ba}_2\text{Cu}_3\text{O}_4\text{Cl}_2$ from LDA + *U* calculations, *Phys. Rev. B* **65**, 115111 (2002).
- [52] A. de la Torre, K. L. Seyler, L. Zhao, S. D. Matteo, M. S. Scheurer, Y. Li, B. Yu, M. Greven, S. Sachdev, M. R. Norman, and D. Hsieh, Mirror symmetry breaking in a model insulating cuprate, *Nat. Phys.* **17**, 777 (2021).
- [53] K. W. Plumb, A. T. Savici, G. E. Granroth, F. C. Chou, and Y.-J. Kim, High-energy continuum of magnetic excitations in the two-dimensional quantum antiferromagnet $\text{Sr}_2\text{CuO}_2\text{Cl}_2$, *Phys. Rev. B* **89**, 180410(R) (2014).
- [54] D. Betto, R. Fumagalli, L. Martinelli, M. Rossi, R. Piombo, K. Yoshimi, D. Di Castro, E. Di Gennaro, A. Sambri, D. Bonn, G. A. Sawatzky, L. Braicovich, N. B. Brookes, J. Lorenzana, and G. Ghiringhelli, Multiple-magnon excitations shape the spin spectrum of cuprate parent compounds, *Phys. Rev. B* **103**, L140409 (2021).
- [55] G. Kresse and J. Furthmüller, Efficiency of *ab-initio* total energy calculations for metals and semiconductors using a plane-wave basis set, *Comput. Mater. Sci.* **6**, 15 (1996).
- [56] J. Hafner, *Ab-initio* simulations of materials using VASP: Density-functional theory and beyond, *J. Comput. Chem.* **29**, 2044 (2008).
- [57] V. Wang, N. Xu, J.-C. Liu, G. Tang, and W.-T. Geng, VASPKIT: A user-friendly interface facilitating high-throughput computing and analysis using VASP code, *Comput. Phys. Commun.* **267**, 108033 (2021).
- [58] S. L. Dudarev, G. A. Botton, S. Y. Savrasov, C. J. Humphreys, and A. P. Sutton, Electron-energy-loss spectra and the structural stability of nickel oxide: An LSDA+*U* study, *Phys. Rev. B* **57**, 1505 (1998).
- [59] T. C. Sterling and D. Reznik, Effect of the electronic charge gap on LO bond-stretching phonons in undoped La_2CuO_4 calculated using LDA + *U*, *Phys. Rev. B* **104**, 134311 (2021).
- [60] E. Pavarini, I. Dasgupta, T. Saha-Dasgupta, O. Jepsen, and O. K. Andersen, Band-structure trend in hole-doped cuprates and correlation with T_{cmax} , *Phys. Rev. Lett.* **87**, 047003 (2001).
- [61] N. Barišić and D. K. Sunko, High- T_{c} cuprates: a story of two electronic subsystems, *J. Supercond. Novel Magn.* **35**, 1781 (2022).
- [62] L. Martinelli, K. Wohlfeld, J. Pellicciari, R. Arpaia, N. B. Brookes, D. Di Castro, M. G. Fernandez, M. Kang, Y. Krockenberger, K. Kummer, D. E. McNally, E. Paris, T. Schmitt, H. Yamamoto, A. Walters, K.-J. Zhou, L. Braicovich, R. Comin, M. M. Sala, T. P. Devereaux *et al.*, Collective nature of orbital excitations in layered cuprates in the absence of apical oxygens, *Phys. Rev. Lett.* **132**, 066004 (2024).
- [63] R. D. Gonzalez Betancourt, J. Zubáč, K. Geishendorf, P. Ritzinger, B. Růžicková, T. Kotte, J. Železný, K. Olejník, G. Springholz, B. Büchner *et al.*, Anisotropic magnetoresistance in altermagnetic MnTe, *npj Spintronics* **2**, 45 (2024).
- [64] T. Osumi, S. Souma, T. Aoyama, K. Yamauchi, A. Honma, K. Nakayama, T. Takahashi, K. Ohgushi, and T. Sato, Observation of a giant band splitting in altermagnetic MnTe, *Phys. Rev. B* **109**, 115102 (2024).
- [65] N. Doiron-Leyraud, C. Proust, D. LeBoeuf, J. Levallois, J.-B. Bonnemaison, R. Liang, D. Bonn, W. Hardy, and L. Taillefer, Quantum oscillations and the Fermi surface in an underdoped high- T_{c} superconductor, *Nature (London)* **447**, 565 (2007).
- [66] S. E. Sebastian, N. Harrison, and G. G. Lonzarich, Towards resolution of the Fermi surface in underdoped high- T_{c} superconductors, *Rep. Prog. Phys.* **75**, 102501 (2012).
- [67] Z.-X. Li, H. Zhou, X. Wan, and W. Chen, Diagnosing altermagnetic phases through quantum oscillations, *Phys. Rev. B* **111**, 125119 (2025).
- [68] A. McCollam, S. R. Julian, P. M. C. Rourke, D. Aoki, and J. Flouquet, Anomalous de Haas–van Alphen Oscillations in CeCoIn_5 , *Phys. Rev. Lett.* **94**, 186401 (2005).
- [69] V. Leeb and J. Knolle, Theory of difference-frequency quantum oscillations, *Phys. Rev. B* **108**, 054202 (2023).
- [70] V. Leeb, N. Huber, C. Pfeleiderer, J. Knolle, and M. A. Wilde, A field guide to non-Onsager quantum oscillations in metals, [arXiv:2408.15092](https://arxiv.org/abs/2408.15092).
- [71] J. A. Ouassou, A. Brataas, and J. Linder, dc Josephson effect in altermagnets, *Phys. Rev. Lett.* **131**, 076003 (2023).
- [72] C. Sun, A. Brataas, and J. Linder, Andreev reflection in altermagnets, *Phys. Rev. B* **108**, 054511 (2023).
- [73] F. Ferrari and R. Valentí, Altermagnetism on the Shastry-Sutherland lattice, *Phys. Rev. B* **110**, 205140 (2024).
- [74] D.-F. Shao, S.-H. Zhang, M. Li, C.-B. Eom, and E. Y. Tsymbal, Spin-neutral currents for spintronics, *Nat. Commun.* **12**, 7061 (2021).

- [75] R. González-Hernández, L. Šmejkal, K. Výborný, Y. Yahagi, J. Sinova, T. Jungwirth, and J. Železný, Efficient electrical spin splitter based on nonrelativistic collinear antiferromagnetism, *Phys. Rev. Lett.* **126**, 127701 (2021).
- [76] S.-B. Zhang, L.-H. Hu, and T. Neupert, Finite-momentum Cooper pairing in proximitized altermagnets, *Nat. Commun.* **15**, 1801 (2024).
- [77] D. Zhu, Z.-Y. Zhuang, Z. Wu, and Z. Yan, Topological superconductivity in two-dimensional altermagnetic metals, *Phys. Rev. B* **108**, 184505 (2023).
- [78] D. Chakraborty and A. M. Black-Schaffer, Zero-field finite-momentum and field-induced superconductivity in altermagnets, *Phys. Rev. B* **110**, L060508 (2024).
- [79] G. B. Sim and J. Knolle, Pair density waves and supercurrent diode effect in altermagnets, *Phys. Rev. B* **112**, L020502 (2025).
- [80] M. Dürrnagel, H. Hohmann, A. Maity, J. Seufert, M. Klett, L. Klebl, and R. Thomale, Altermagnetic phase transition in a Lieb metal, *Phys. Rev. Lett.* **135**, 036502 (2025).
- [81] B. Brekke, A. Brataas, and A. Sudbø, Two-dimensional altermagnets: Superconductivity in a minimal microscopic model, *Phys. Rev. B* **108**, 224421 (2023).
- [82] B. Lau, M. Berciu, and G. A. Sawatzky, High-spin polaron in lightly doped CuO₂ planes, *Phys. Rev. Lett.* **106**, 036401 (2011).