**CORRELATED MATERIALS**

Systematic electronic structure in the cuprate parent state from quantum many-body simulations

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The quantitative description of correlated electron materials remains a modern computational challenge. We demonstrate a numerical strategy to simulate correlated materials at the fully ab initio level beyond the solution of effective low-energy models and apply it to gain a detailed microscopic understanding across a family of cuprate superconducting materials in their parent undoped states. We uncover microscopic trends in the electron correlations and reveal the link between the material composition and magnetic energy scales through a many-body picture of excitation processes involving the buffer layers. Our work illustrates a path toward a quantitative and reliable understanding of more complex states of correlated materials at the ab initio many-body level.

In spite of our qualitative theoretical understanding of many electronic phases of matter, there remains a deficit in the quantitative understanding of correlated electron materials (1, 2). This limits our ability to connect the atomic structure and composition to the electronic phenomena, as well as to answer fundamental physical questions related to microscopic mechanisms.

Among correlated quantum materials, the high-temperature ($T_c$) superconductors remain a fertile source of new physics (3–6). We focus on the cuprates, where one finds the highest superconducting $T_c$ in the mercury-barium cuprate family (7). Although progress has been made in understanding the universal phase diagram through numerical calculations on lattice models, the understanding of properties of individual compounds remains largely empirical, with substantial difficulties in linking the observed trends to model parameters.

In principle, a quantitative understanding is simply a matter of many-electron quantum mechanics, but solving the Schrödinger equation beyond lattice models involves three challenges: the quantum many-body correlations, the thermodynamic limit (TDL), and the high-energy degrees of freedom and long-range interactions of real materials. We adopt a pragmatic computational framework in which the challenges can be tackled simultaneously: solvers for the many-body problem beyond models (8, 9), self-consistent quantum embedding to develop phases in the TDL (10–12), and periodic quantum chemistry using local bases (13, 14) to efficiently treat long-range interactions and high-energy degrees of freedom. Each component has been individually tested in prior work, but the principal feature of our combined strategy is that the solution process bypasses models with uncontrolled parameters; the only remaining parameters are the size of the computational cell, the basis size, and the level of the many-body solver. Thus, all aspects of the calculation can in principle be controlled toward exactness.

In this work, we describe the full application of this strategy to the simulation of a family of cuprates in their parent phase at zero temperature. Although the parent phase is qualitatively simple, and elements of our framework have been used to understand exotic physics in simplified models (15), obtaining quantitative material systematics and functional relationships even in the parent phase is a major challenge and serves as a litmus test of the promise of our overall strategy. Our detailed simulations improve the level of resolution of the electronic structure and enable us to uncover direct links between the material-specific physics and composition.

**Cuprates and the parent state**

**Structure**

The main structural feature of the cuprates is the two-dimensional (2D) CuO$_2$ (formally [CuO$_2$]$^{2-}$) square lattice plane (Fig. 1A). In different cuprates, the copper-oxygen plane is surrounded by different atoms and buffer layers in the vertical direction. We consider three specific compounds, in addition to layer-stacked idealized CuO$_2$ planes (geometries are provided in table S1). The first is infinite layer CaCuO$_2$ (CCO) (Fig. 1D), where calcium counterions intercalate between the CuO$_2$ planes in an infinitely repeating structure. CCO does not itself superconduct because of difficulties in doping the material. However, high $T_c$ are observed in the related mercury-barium cuprates (the Hg–Ba–Ca–Cu–O family). There, the CuO$_2$ plane is decorated by apical oxygens, which connect to buffers of Hg and Ba ions. Unlike in CCO, the buffer layers form large spacers between the copper-oxygen layers. Different mercury-barium cuprates can be synthesized with different numbers of CuO$_2$ planes between each buffer layer, leading to single-layer cuprates, double-layer cuprates, and so on. We consider two members in this family: HgBa$_2$CuO$_4$ (Hg-1201; single-layer, $T_c = 97$ K) and HgBa$_2$CaCu$_2$O$_8$ (Hg-1212; double-layer, $T_c = 127$ K). Hg-1201 exhibits distorted octahedral Cu–O coordination (two apical oxygens per Cu) (Fig. 1B), whereas each layer of Hg-1212 contains pyramidal Cu–O coordination (one apical oxygen per Cu) (Fig. 1C). Hg-1201, Hg-1212, and CCO are compositionally related by replacing Hg-Ba-apical O layers with Ca layers.

**Parent state**

Unlike conventional superconductors, the parent state of the cuprates is an antiferromagnetic (AFM) insulator with long-range order, resulting from the strong $d-d$ electron interaction. Typical Néel temperatures for the AFM state range from about 250 K (in Nd$_2$CuO$_4$) to 450 K (in YBa$_2$Cu$_3$O$_6$) (5), and only after doping does the ground state enter the superconducting phase. It is generally thought that the antiferromagnetism is to first order approximated by 2D nearest-neighbor (NN) Heisenberg-like physics. However, the 2D NN Heisenberg model does not reproduce the dispersion of the experimental spin-wave spectrum, and questions remain as to the magnitude, sign, and material-specific origin of corrections to the NN picture.

There have been many attempts to correlate properties of the cuprates in the superconducting phase (such as $T_c$) with structure, composition, and band structure (16–21). However, without a direct ability to simulate the material $T_c$ with different parameters, distinguishing correlation from causation is difficult. Although there has been less focus on correlating properties of the parent state with physical features, many proposals relate the high Néel temperatures and strong exchange coupling in the parent state to the superconducting mechanism and other exotic physics under doping. We establish causal, quantitative relationships between the magnetic features of the parent state and the atomic-scale structural and electronic features of the materials.

**Theoretical techniques**

**Strategy**

Previous numerical work on cuprate electronic structure [with a few exceptions, such as (22, 23)] falls in two classes: (i) ab initio all-electron simulations with a modest treatment of electron correlations (24–26), which are often used to derive low-energy effective models, and (ii) accurate many-body methods applied to low-energy effective models, to obtain phase diagrams and more exotic orders (27–33).

Our strategy is to use families of methods associated with the model studies of (ii), but technologically elevated to the fully ab initio
The quantum embedding provides a framework for phases that emerge as a result of interactions (35) and includes dynamical mean-field theory and its relations (36–38) and the DMET (39, 40) used in this work. We separate the material into an impurity region and a bath that describes fluctuations out of the impurity, and their self-consistency yields emergent phases. The embedding becomes exact with increasing impurity size.

In previous work on the one-band and three-band Hubbard models, DMET has been extensively benchmarked against other methods and, for example, accurately resolves exotic order in the underdoped region (15, 41). The ability of DMET to capture exotic physics in doped lattices suggests a path from the ab initio studies of the parent state here to the physics of the doped materials. To move beyond models to the ab initio physics, we start from our recently introduced all-electron, full-cell approach (10–12). In this work, the impurity is a supercell of the cuprate that contains all atoms and orbitals, with all quartic interactions between the orbitals. In contrast to downfolded approaches with a handful of impurity orbitals and possibly simplified interactions (37, 42), our largest impurity (in Hg-1212) contains 48 atoms and close to 900 orbitals (Fig. 1E). These orbitals include many “virtual” bands, which capture quantitative electron correlation effects and screening. Part of the reason why these large impurities are feasible is the DMET formulation itself, which bypasses expensive frequency-dependent quantities. The other critical factors are the choice of solvers and the periodic quantum chemistry infrastructure based on local atomic basis sets, which compactly discretize the virtual bands for electron correlation ([34], section 1).

The quantum impurity problem in the full-cell approach is a many-body problem with hundreds of orbitals. This can be solved because many orbitals do not display strongly correlated physics. We use two impurity solvers in this work. We obtain the majority of the results using an ab initio CC singles and doubles (CCSD) (43) solver. Although approximate, they treat clusters of (arbitrarily) strongly correlated particles exactly and have previously been shown to yield accurate results in various quantum impurity problems (9–12, 44), particularly in ordered phases. To
verify the accuracy of the CC approximation, we use a second solver, the quantum chemistry DMRG (8, 46), to benchmark a subset of problems.

**Computational setup**

The $2 \times 2$ supercell impurities are shown in Fig. 1 for the different mercury-barium cuprates. In (34), section 2.1.5, we also discuss a benchmark study of lanthanum copper oxide. Every atom is represented in a valence double-$\zeta$ with polarization basis (def2-SVP) (46); for example, each Cu is represented by $[5s3p2d1f]$ shells and each O by $[3s2p1d]$ shells, and the embedding lattice is chosen to be an $8 \times 8 \times 2$ lattice of the primitive cell. Large impurities (such as in Hg-1212) were further fragmented into smaller subimpurities with up to 364 orbitals (280 impurity orbitals and 84 valence bath orbitals), and impurity solutions were obtained by using CCSD or DMRG. [Unless otherwise indicated, data are from the CCSD solver; DMRG data are in (34), section 2.1.4.] The DMET equations were then solved with self-consistency and valence-shell lattice-impurity density matrix fitting.

**Benchmarks**

Within the above strategy, the only sources of error are from the finite size of the impurity (and embedding lattice), the approximate nature of the impurity solver, and the finite size of the local atomic basis. We carried out extensive benchmarking to verify the specific approximations. In Fig. 1F, we compare results from finite impurities to the TDL (which can be estimated from a full crystal calculation within a small local atomic basis) for the energy difference between the ferromagnetic (FM) and the AFM state (proportional to the NN exchange coupling $J_i$). We also show the deviation between this energy difference estimate from the DMET and CCSD solvers in a small impurity where DMRG is tractable. Both sets of data illustrate that the TDL and many-body character of the physics are well captured within the approximations in this work. Additional benchmarks (such as basis set convergence) can be found in (34), sections 2.1 and 3.3 (figs. S4 and S5 and tables S15 to S18).

**Multiorbital electronic structure**

We start with general electronic trends across the series Hg-1201, Hg-1212, CCO, and $[\text{CuO}_2]^2$ as a baseline to understand trends in the physics in later sections.

**Order parameters and bonding**

We first extract order parameters from the $2 \times 2$ computational supercell: charge, local moment, bond orders (from the off-diagonal elements of $\gamma_{ij} = \langle a_i^\dagger a_j \rangle$, where $i, j$ label local atomic orbitals in the cell), and the spin correlation function $(S_i(0)S_j(r))$ measured across the full crystal (Fig. 2, A to E).

The key features are the following: (i) The ground state is AFM with long-range order, with the moment in the Cu half-filled $3d_{x^2-y^2}$ orbital. Cu $4s/4p$ occupancy reduces the total moment by about 10%. The unit cell moment ranges from 0.71 in Hg-1201 to 0.55 in $[\text{CuO}_2]^2$. (ii) Charge is transferred from in-plane O to the other ions, with the degree of transfer increasing across the series. There is significant charge transfer to the Cu minority spin orbitals (as much as 0.3 electrons in $[\text{CuO}_2]^2$), (iii) Ca and Ba buffer atoms in CCO, Hg-1201, and Hg-1212 are ionic, with Hg covalently bonded to the apical oxygen through the O 2p$_z$-Hg 6s, 5d$_z$ bonds. Hg-1201 and Hg-1212 do not differ much with respect to the out-of-plane observables but do differ for their observables in the CuO$_2$ plane. (iv) In-plane $\sigma$-bonding (Fig. 2F) is predominantly Cu 4p-O 2p and does not differ much across the compounds. However, Cu 3d-O 2p bonding and out-of-plane $\pi$ bonding increase across the series, reflecting increasing in-plane 3d/4p hybridization. The change in bonding is not (solely) caused by the structural changes (for example, CCO and $[\text{CuO}_2]^2$ have the same Cu-O bond length but different bond orders) but instead reflect redistribution of charge from the buffer layers. (v) The apical oxygen bond order (Fig. 2G) decreases from Hg-1201 to Hg-1212, with the oxygen only weakly bound to Cu. Cu 4s and 4p$_z$ contribute to apical bonding, with little 3d$_{x^2}$ participation.
We obtain additional insight from the spin-resolved (\(\gamma'^{+}\)) and spin-traced (\(\gamma = \sum_{q} \gamma'^{+}\)) single-particle density matrices (equal-time Green’s functions) evaluated in the full crystal. These provide nonlocal and \(k\)-space information on correlations. We first discuss the spin-traced single-particle density matrix. The eigenvalues (the natural occupancy distribution, sometimes called the momentum distribution function) and eigenvectors (natural orbitals) illustrate the degree of symmetry breaking and highlight the important degrees of freedom near the Fermi level. The spin-traced natural occupancy distribution together with the projected atomic character of the eigenvectors is shown in Fig. 3, A and B. The most important orbitals near the Fermi level are the classic three-band orbitals: Cu 3d_{x^2−y^2} and O 2p_{x,y}. We also find no single next-most-important orbital; Cu 4s, 4p, 3d_{x,y}, and the apical oxygen and Hg orbitals all contribute to a similar degree.

The spin-resolved natural occupancies and eigenvectors indirectly reflect the nature of the quasiparticles and the importance of dynamical effects [discussion is available in (34)], section 1.3.4]. The eigenvectors with natural occupancies closest to the jump across the Fermi level can be viewed as “pseudo”-valence band maximum (VBM)–conduction band minimum (CBM) states. Defined in this way, the pseudo-VBM is dominated by O 2p_{x,y}, whereas the pseudo-CBM is dominated by Cu 3d_{x^2−y^2} (and the apical O and Hg bands in the Hg-Ba compounds) (Fig. 3C). This classifies all the compounds as charge-transfer insulators.

We further untangle the effect of interactions from pure single-particle physics by comparing the spin-resolved natural occupancies of the correlated calculation with that of a spin-polarized Hartree-Fock (HF) reference. The correlated spin-resolved natural occupancies are all quite close to 0 and 1 (fig. S15), the mean-field values; thus, dynamical effects are small. However, the orbital components of the eigenvectors are very different between the mean-field and correlated distributions (Fig. 3C), indicating strong static effects. It appears that in the AFM state the effect of interactions on the quasiparticles is mainly static rather than dynamical and can be captured largely through static screening of the interactions, correlation-driven rehybridization of the orbitals, and renormalization of their energies.

**Magnetic trends across the cuprates**

We next characterize the low-lying magnetic excitations across the series of cuprates. To do so compactly, we introduce a magnetic model (not to solve for the electronic structure, but for interpretation) and extract exchange couplings from our correlated calculations of different spin configurations: the AFM state, the FM state, and a spin-density wave state (fig. S3). From these, we derive parameters for the NN Heisenberg model (\(J\)) and a multi-\(J\) Heisenberg model in which the exchange couplings \(J_{1}, J_{2}, J_{3}\), and \(J_{c}\) are related through the perturbation expansion of the one-band Hubbard model (with only three free parameters). A 3\(\times\)3\(\times\)3 model, in which \(J_{c}\) is renormalized into the \(J_{1}, J_{2}, J_{3}\) parameters, can also be derived. In CCO, we also derive an interlayer \(J_{c}\) using two AFM layer configurations. A full discussion of all models and the spin-wave calculation is available in (34), section 1.5.2.] The parameters are illustrated in Fig. 4B and tabulated in tables S10 to S13. We display the corresponding spin-wave spectrum from linear spin wave theory in Fig. 4A.

**Spin-wave spectrum**

In CCO, the full experimental dispersion is available, whereas for Hg-1201 and Hg-1212, only part of the dispersion near the \(\Gamma\) point has been measured. As expected, the NN Heisenberg model does not capture dispersion away from the \(\Gamma\) point; however, the derived NN \(J\) agrees well with that derived from experiment by fitting near the \(\Gamma\) point. For example, in CCO, the NN \(J\) fit to DMET data yields \(J = 155\) meV, compared with \(J = 142,158\) meV (the two numbers are from different experiments) (47, 48). The multi-\(J\) model

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**Fig. 3. Natural occupancy distribution (eigenvalues of the single-particle density matrix) and quasiparticle character.** (A) Occupancy of natural orbitals around the Fermi level (dashed line), from the spin-traced density matrix (\(\gamma'^{+}\)) in Hg-1201, Hg-1212, and CCO. Orbital character is indicated with colors and labels. (B) Half-filling index of the different local orbitals (definition is provided in eq. S50), measuring their importance in the most correlated orbitals of the calculation. (C) Orbital component analysis of the spin-resolved mean-field (HF) and correlated (DMET) top valence (V) and bottom conduction (C) bands of CCO at different \(k\) points (averaged from the eight bands near the Fermi level). \(\Gamma\): (0, 0); \(X\): (\(\frac{1}{2}, 0\)); \(M\): (\(\frac{1}{2}, \frac{1}{2}\)).
with ab initio parameters yields improved agreement across the experimental dispersion, illustrating the importance of long-range exchange. The discrepancies are largest near the X point (\( \frac{1}{4}, \frac{3}{4}, 0 \)) likely owing to finite size effects in the embedding, although there are also confounding factors from the experimental setting in Hg-1201 and Hg-1212 ([34]), section 2.3.5. Compared with CCO, the Hg-Ba compounds display flatter dispersions, and we capture this in our derived spin-wave spectrum.

**Magnetic parameters**

Trends in the magnetic couplings of the multi-J Heisenberg model among the four compounds are shown in Fig. 4B. Across the series Hg-1201, Hg-1212, CCO, CuO2\(^{2-}\), all couplings \( J_1, J_2, J_3 \), and \( J_c \) increase substantially. \( J_1 \) roughly doubles, and \( J_c \) increases by a factor of 5, illustrating (i) the importance of the buffer layers in the long-range exchange coupling and (ii) the increasing “delocalization” across the series of compounds. A recent resonant inelastic x-ray scattering (RIXS) experiment ([49]) suggests that \( J_1 \) increases significantly (by about 20 to 30%) from the single-layer

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**Fig. 4. Spin wave dispersion of Hg-1201, Hg-1212, and CCO.** (A) The 2D magnetic Brillouin zone is sampled along \( \Gamma, (0, 0); X, (\frac{1}{4}, 0); R, (\frac{1}{4}, \frac{1}{4}, 0) \); \( k_z \) is fixed at 0.46 to match the experimental conditions in CCO and fixed at 0 for Hg-1201 and Hg-1212. NN Heisenberg (1J) and multi-J model curves are shown. The multi-J model includes a quantum renormalization factor of 1.219 ([52]). Experimental RIXS data are extracted from ([49], [53]) for Hg-1201 and Hg-1212 and ([47]) for CCO. (B) Trends in the multi-J model parameters across the cuprate family. Hybrid density functional (PBE0, HSE06, and B3LYP) results for the first two Hg compounds are also shown with symbols. Details are available in ([34]), section 2.3.

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**Fig. 5. Effects of buffer layers.** (A and B) Representative out-of-plane orbitals (isosurfaces) in (A) Hg-1201 and (B) CCO. (C) The effect of freezing fluctuations to out-of-plane orbitals on the NN magnetic coupling \( J_1 \) and cyclic exchange coupling \( J_c \). (D) Excitations relevant to exchange pathways in cuprates. (i) Superexchange is facilitated by excitations from in-plane oxygen orbitals to empty copper states; (ii) in Hg-1201, substantial excitations from the copper-oxygen plane to the buffer layer reduce superexchange. Double-excitation pathways (iii) and (iv) involving buffer layers are also shown. The numbers \( \Delta T_1, \Delta T_2 \) reflect the change in excitation weight upon unfreezing the buffer orbitals. (E) Influence of apical Cu–O distance on exchange coupling \( J_1 \) and \( J_c \) at the mean-field (HF) and correlated (DMET) level.
Hg-1201 to the double-layer Hg-1212, which is similar to the increase in $T_c$. We find quantitative agreement with our correlated calculations, in which Hg-1212 shows an increase in $T_c$ by about 18%.

**Effect of interactions**

To understand the effect of interactions, we can compare to the mean-field HF results. These give almost flat dispersion curves because the $J$ couplings are very small (for example, $J_1 \sim 40$ meV), and the magneton energy at the $\Gamma$ point is also lower than the experimental value. Thus, the observed magnetic energy scales require a careful treatment of electron correlations. As suggested above, a large part of the effect of interactions can be captured by a renormalization of the low-energy band structure and interaction. Choosing a density functional treatment or Hubbard $U$ parameter can mimic this; however, we do not find that a single choice of functional or Hubbard $U$ consistently or accurately reproduces the material trends. For example, moving from Hg-1201 to Hg-1212 should yield a substantial increase in the exchange couplings, but $J_1$ decreases with the B3LYP functional and increases only marginally with HSE06 (5%) and PBE0 (6%) (Fig. 4B, symbol data). In addition, $J_3$ is overestimated by all of the above functionals.

**Untangling layer effects**

We now connect the microscopic correlated electronic structure with the trends in the magnetic physics observed above to derive mechanistic insights. Changing the buffer layer leads to large changes in the exchange couplings (particularly for the nonlocal terms). However, this effect does not appear at the HF mean-field level. To verify that it is caused by fluctuations (electron correlations) coupling the CuO$_2$ plane with the buffer layers (and not simply the effect of the electrostatic potential of the buffer layer on electron correlation within the cuprate plane), we devise a procedure that allows us to switch electron correlation with the buffer layer orbitals on and off. To do so, we explicitly freeze excitations involving out-of-plane orbitals in the correlated impurity solver calculations (the impurity wave function excludes particle-hole excitations from the HF determinant involving out-of-plane orbitals) ([34], section 2.4.1). Any changes from freezing and unfreezing these fluctuations therefore directly reflect the influence of electron correlation with the orbitals of the buffer plane.

Representative out-of-plane orbitals of Hg-1201 and CCO are shown in Fig. 5, A and B. The out-of-plane impurity orbitals consist of empty outer valence shells on Ca, Hg, and Ba apical oxygen orbitals and other orbitals that originate from the adjacent copper-oxygen plane. The Ca- and Ba-centered localized orbitals (4s and 6s) are similar in CCO and Hg-1201.

The changes in the $J_1$ and $J_2$ from unfreezing the out-of-layer orbitals are shown in Fig. 5C. In both compounds, the exchange couplings are decreased by freezing, but in CCO, the effect is stronger, and $J_3$ is especially strongly influenced by freezing, decreasing by as much as 71% in CCO. To understand this, we analyze the correlated impurity wave functions in CCO and Hg-1201. Shown in Fig. 5D are the changes in the weights of single-particle excitations $\Delta T_1$ and connected two-particle excitations $\Delta T_2$ upon unfreezing the buffer layer in the two compounds. Generally speaking, when the buffer layer is unfrozen, the increased excitation manifold increases screening and decreases the energetic penalty to excite from filled to empty states, such as the empty Cu and buffer-layer states. In CCO and Hg-1201, we find that this increases the $O \rightarrow Cu$ excitation associated with superexchange [Fig. 5D, process (i)], increasing the exchange couplings. However, in Hg-1201, we see in addition a significant increase in excitations from in-plane Cu, O orbitals to the empty Hg, apical O states [Fig. 5D, process (ii)]. This change in the copper plane to buffer excitation is more than twice as large in Hg-1201 as in CCO, and it depletes the ground configuration associated with in-plane exchange and reduces the effective nonlocal hopping by rehybridizing the Cu empty states (16), canceling the enhancement of in-plane $O \rightarrow Cu$ excitations and yielding an aggregate small change in exchange coupling upon unfreezing the buffer orbitals ([34], section 2.4.2). This also explains why the exchange couplings of Hg-1212 lie in between those of Hg-1201 and CCO: The buffer suppression of in-plane superexchange occurs through a single buffer layer in Hg-1212 versus two on either side in Hg-1201. The analysis also reveals (smaller) differences between the compounds in the connected two-particle fluctuations involving the buffer [Fig. 5D, processes (iii) and (iv)]; these are material-specific effects that cannot be folded into a static renormalization. Last, in Fig. 5E we show the effect on the exchange coupling of increasing the apical oxygen distance in Hg-1201, both at the mean-field level and at the correlated level. Consistent with the above mechanism, we find that increasing apical oxygen distance removes the buffer suppression effect in the correlated calculation (increasing the exchange coupling) but makes little difference in the mean-field calculation because fluctuations must first renormalize the energies of the empty states for them to be accessible.

**Discussion**

We have demonstrated that through a numerical strategy that combines quantum embedding, ab initio quantum solvers, and periodic quantum chemistry, we can determine material-specific correlated electron structure in the parent state of the cuprates at the many-body level. This reveals trends in the multi-orbital bonding and correlation effects in the Fermi distribution and quasiparticles and gives a quantitative description of the low-energy magnetic excitations. Across a series of homologous mercury-barium and calcium cuprates, the systematic trends in the nature of the magnetic exchange can be explained through the analysis of the many-body state, which uncovers a competition between superexchange and plane-to-buffer excitation processes.

A general observation is that although the interactions are strong, many of their effects in the parent state can be renormalized into a static low-energy theory. This supports the long-standing practice of interpreting physics in this region through simple band structures and static interaction parameters. However, we also find that empirical approaches to determine this renormalization do not have the accuracy to capture the trends among the materials, unlike the controlled many-body approaches used here.

A strength of the many-body approach is that we can interrogate individual electronic processes, and our ab initio formulation allows us to trace these processes beyond models to the individual atomic orbital level. We used this capability to untangle the links between layer composition and magnetic exchange. In present work, it was conjectured that the range of magnetic exchange is related to electronic processes involving an effective apical conduction band and that this further correlates with the superconducting transition temperature (16). We now have a direct picture of the first part of this conjecture, with rich atomic-scale and many-body resolution.

Components of the numerical strategy in this work have previously been used to describe exotic phases in models. The success of the current ab initio realization for cuprate parent states thus gives hope that a similar approach may eventually yield a quantitative picture of more complex cuprate phases. If that is the case, we may be able to answer the second part of the above and similar conjectures about superconducting properties, through a direct ab initio simulation of the superconducting orders and the energy scales of the cuprates in their doped states.

**REFERENCES AND NOTES**

1. E. Dagotto, Science 309, 257–262 (2005).
2. R. M. Martin, L. Reining, D. M. Ceperley, Interacting Electrons (Cambridge Univ. Press, 2005).
3. E. Dagotto, Rev. Mod. Phys. 66, 763–840 (1994).
4. J. Orenstein, A. J. Millis, Science 288, 468–475 (2000).
5. N. Paikida, High-Temperature Cuprate Superconductors: Experiment, Theory, and Applications, vol. 166 (Springer, 2010).
6. X. Zhou et al., Nat. Rev. Phys. 3, 452–465 (2021).
7. A. Schilling, M. Cantoni, J. D. Guo, H. R. Ott, Nature 363, 56–58 (1993).
8. H. Zhai, G. K.-L. Chan, J. Chem. Phys. 154, 224116 (2021).
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