I. INTRODUCTION

Multiorbital systems in which the low-energy behavior is affected by various orbitals provide an ideal setup for the formation of rich electronic phases. One notable example is the family of layered perovskites, $\text{Sr}_n\text{Ru}_3\text{O}_{3n+1}$. Much attention has been paid to the single layer material, $\text{Sr}_2\text{Ru}_3\text{O}_7$ (214), which displays superconductivity (SC) at low temperatures [1–4]. However, despite intense studies, the nature of the superconducting state still remains a topic of debate [4, 5]. Interest in the bilayer material, $\text{Sr}_3\text{Ru}_2\text{O}_7$ (327), grew after significant anisotropy in the resistivity, signaling an electronic nematic phase, was reported [6–12]. More recently, a resistivity linear in temperature [13], as well as spin-density wave (SDW) order were found to occur at low temperatures at field values coinciding with the presumed nematic phase, bounded by metamagnetic transitions [14–17].

A longstanding puzzle in this family is the lack of SC in 327, constraining the possible superconducting mechanisms of 214. The similarity of these materials means that a model of 214 should be able to explain the lack of SC in 327 and the sensitivity to the differences between the two. The two notable differences introduced by the second layer in 327 are the bilayer coupling and the staggered rotations of the oxygen octahedra. The observation of SDW order in the presence of a magnetic field in 327 suggests a delicate competition between SC and SDW in these two sister compounds, calling for a unified picture.

Here, we address the microscopic mechanism of the evolution from SC in 214 to SDW in 327. We show that the absence of SC in 327 supports interorbital pairing in 214 which is sensitive to orbital mixing via spin-orbit coupling (SOC). Using the same parameter set for the Kanamori Hamiltonian, we find interorbital spin-triplet SC is destroyed by the staggered rotations of the octahedra, and intraorbital SDW order emerges under a magnetic field in 327. Further, we predict that ideal bilayer 327 without staggered rotations of the octahedra, denoted (327)$_0$, displays interorbital SC with a transition temperature possibly higher than 214, as shown in Fig. 1, the phase diagram of these systems in a field.

II. MICROSCOPIC THEORY

$\text{Sr}_2\text{Ru}_3\text{O}_7$ was once thought to be a leading candidate for $p + ip$ spin-triplet SC [18, 19]. However, recent NMR data showing a drop in the Knight shift below the superconducting transition temperature seems to rule out odd-parity spin-triplet pairing [20, 21]. While $p$-wave states may not explain the Knight shift reduction, spin-triplet proposals remain possible with even-parity interorbital spin-triplet states [22–29]. This pairing in the orbital basis is spin-triplet, but appears as pseudospin-singlet in the band basis due to SOC. The interorbital nature of the pairing means that significant band degeneracy near the Fermi level is required [30, 31]. Alternatively, when SOC and band separation are comparable in energy, SOC stabilizes interorbital pairing via mixing of orbitals and spin [22, 26, 28, 29, 32–36].

We adopt the Kanamori Hamiltonian, well known for multiorbital systems, to investigate competition between SC and SDW order in 214, (327)$_0$, and 327. In multiorbital
systems, the distinction between intraorbital, interorbital-singlet, and interorbital-triplet pairings must all be considered. Among all possible order parameters, inter- and intraorbital SDW, and interorbital SC occur via attractive interactions, however, interorbital SDW is small compared to intraorbital SDW and is therefore not shown. Intraorbital SC and charge-density wave channels feature repulsive interactions, but intraorbital SC is induced from interorbital SC via SOC, though is small in comparison.

Taking into account interorbital SC and intraorbital SDW, the effective Hamiltonian is found as,

$$ \frac{H_{\text{eff}}}{N} = 2(U' - J_H) \sum_{\alpha \neq b} \hat{\Delta}^\dagger_{a/b} \hat{\Delta}_{a/b} - 2U \sum_a \hat{m}_a^\alpha \cdot \hat{m}_a^\alpha - J_H \sum_{a \neq b} \hat{m}_a^\alpha \cdot \hat{m}_b^\alpha. \quad (1) $$

The orbital-singlet, spin-triplet, superconducting order parameter, $\hat{\Delta}^\dagger_{a/b}$, is written as,

$$ \hat{\Delta}^\dagger_{a/b} = \frac{1}{4N} \sum_k \hat{c}^\dagger_{k,a,x} \hat{\sigma}^\dagger_{a,x} \hat{c}^\dagger_{k,a,x} - \hat{c}^\dagger_{k,b,x} \hat{\sigma}^\dagger_{a,x} \hat{c}^\dagger_{k,b,x}, \quad (2) $$

with $l = x, y, z$ and $a \neq b$ represents the sum over the unique pairs of orbital indices in $(yz, xz, xy)$. The intraorbital SDW order parameter, $\hat{m}_q^\alpha$, is given by,

$$ \hat{m}_q^\alpha = \frac{1}{2N} \sum_k \hat{c}^\dagger_{k+q/2,\sigma} \hat{\sigma}^\dagger_{\sigma,\sigma} \hat{c}^\dagger_{k-q/2,\sigma}. \quad (3) $$

where the electron operator $\hat{c}^\dagger_{k,\sigma}$ creates an electron in orbital $a$ with spin $\sigma$.

To investigate the competition between SC and SDW order in these three systems, we must determine their tight-binding (TB) Hamiltonians. TB parameters are obtained from density functional theory (DFT) calculations, and listed in Tables AI and AII of appendix A. The SC pairing itself is insensitive to the choice of TB parameters, however, the symmetry of the pairing state depends on the details as shown in Ref. 29.

III. EVOLUTION OF SC

Self-consistent mean-field approximation (MFA) calculations are performed for the TB models of 214, (327)$_0$, and 327. (327)$_0$ uses the TB parameters from the single layer with interlayer hoppings added, referred to collectively as the bilayer coupling. In addition to bilayer coupling, interlayer hopping terms may be modified or added in 327 due to the staggered rotations of octahedra not present in the single layer.

The phase diagram for each of the three systems is shown in Fig. 1 as functions of the effective MFA interactions, $V_{SDW} = U$ and $V_{SC} = J_H - U'$. Comparing with 214, it is clear that the SC region is extended in (327)$_0$, while the SDW region is extended in 327. Since the precise values of $U$, $U'$, and $J_H$ are unknown, we set $U' = U - 2J_H$, and choose a set of $(U, J_H)$ which gives SC in 214. These same values are used in 327 and (327)$_0$ since strong variation of local interactions is not expected across these systems. An asterisk in Fig. S1 corresponds to $(U, J_H) \sim (0.8, 0.4)$, where units of $2t_{xz}^2 = 1$ in 214, given in Table S1, are used throughout this work.

For the fixed values represented by the asterisk, the phase diagram in a magnetic field perpendicular to the layers is shown in Fig. 1, where areas of finite SC are shown in red. SC exists 214 and is increased when bilayer coupling is added in (327)$_0$. The addition of staggered rotations in 327 destroys SC, which is replaced by a paramagnet. Further increasing the field eventually stabilizes SDW in 327, discussed below. This overall tendency is independent of the precise choice of $(U, J_H)$ because the evolution of SC to SDW originates from the reduction SC and enhancement of SDW in 327.

To understand why SC is enhanced in (327)$_0$, consider the effect of the interlayer hopping in a two orbital model. Interlayer hopping is orbital dependent and largest for $yz$ to $yx$ and $xz$ to $xz$ hopping, while it is minimal between $xy$ orbitals. A two-orbital model of $xz$ (or $yz$) and $xy$ gives insight into the effects of bilayer coupling. We use the basis $\Psi_k = (\psi_k^x, T \psi_k^y, T^{-1})$ where $T$ represents time-reversal, and $\psi_k^x = (\psi_{k,1}^x, \psi_{k,1}^x, \psi_{k,1}^x, \psi_{k,1}^x, \psi_{k,1}^x, \psi_{k,1}^x, \psi_{k,1}^x, \psi_{k,1}^x)$, where the top and bottom layers are represented by the subscript 1 and 2, respectively. The kinetic and SOC parts of the Hamiltonian are then given by,

$$ H_k = \xi_k \rho_{3\eta} \tau_3 \sigma_0 + \xi_k' \rho_{3\eta} \tau_2 \sigma_0 + \lambda \rho_{3\eta} \tau_2 \sigma_1 + 1 \frac{1}{2} \tau_1 \rho_{3\eta} \tau_1 (\tau_0 + \tau_1) \sigma_0, \quad (4) $$

where $\eta$, $\tau$, $\sigma$, and $\rho$ are Pauli matrices representing the layer, orbital, spin, and particle-hole bases, respectively. The orbital dispersions in $xz$ and $xy$ are represented by $\xi_{k,xz/xy} = \xi^x_k \pm \xi^x_k$ and left in this general form here. Details of these dispersions used in the full three-orbital model are found in Appendix A. The SOC is given by $\lambda$, and the interlayer hopping, $t_{1\perp}$, exists only in $xz$.

The interorbital-singlet spin-triplet pairing, $\Delta^z_{x/z}/xy$, with a $d$-vector in the $x$-direction is written as,

$$ H_{SC} = \Delta_{x/z/xy} z_1 \rho_{1\eta} \tau_2 \sigma_1. \quad (5) $$

Transforming $H_{SC}$ to the band basis, i.e., the basis in which $H_k$ is diagonal, it is clear how intraband pairing forms, and how the various terms affect the gap size; the magnitude
of the intraband gaps in the two bands is,

$$|\Delta_{\text{band}}^\pm| = \frac{2\Delta_{xy}^{xy}}{\sqrt{(\xi_k - \frac{1}{2}t_{\perp})^2 + 4\lambda^2}}. \quad (6)$$

This shows first that SOC is responsible for the formation of the intraband gap, consistent with previous works when $t_{\perp} = 0$ [28, 29]. Second, it relates the interorbital SC pairing, $\Delta_{xy}^{xy}$, to the pairing on the band, $\Delta_{\text{band}}$, where the pairing on the band is affected by the band dispersion $\sqrt{(\xi_k - \frac{1}{2}t_{\perp})^2 + 4\lambda^2}$ or $\sqrt{(\xi_k - \frac{1}{2}t_{\perp})^2 + 4\lambda^2}$. The pairing on one of the bands is increased by $t_{\perp}$ if the degeneracy of the bands is increased. Therefore, when the bilayer coupling brings two bands closer together near the Fermi level, the resulting bands feature a more significant mixture of the two orbitals with appropriate spin character via SOC, and thus are more ideal for interorbital SC to form.

The effect of the bilayer coupling on the orbital and spin mixing, shown for 214 in Figs. 2(a) and (b), is another way to understand the effect on interorbital SC pairing. This displays the trend of orbital and spin mixing via SOC, measured by $2L_{xy}S_{xy}$ or $2L_{y}S_{y}$, favorable for $\Delta_{xy}^{xy}$ or $\Delta_{xy}^{xy}$, within each of the bands at the Fermi level. Red areas are the most mixed, ideal for SC. Note that not all bands exhibit the increase in such mixing due to the bilayer, i.e., the bilayer can in principle decrease the band degeneracy, which works against the SC. For the 214 TB model though, we found that it works in favor of SC.

Next, we consider the effect of the staggered rotations present in 327. While the bilayer coupling may bring band energies closer together and increase degeneracy, orbital hybridization is detrimental to the formation of interorbital-singlet spin-triplet SC [28, 33]. The hybridization from staggered rotations in 327 is included in Fig. 2(c), showing that areas of maximum mixing in the single layer and ideal bilayer have disappeared, destroying SC. This is consistent with the phase diagram of 327 in Fig. 1, obtained by self-consistent MFA calculations.

### IV. SPIN-DENSITY WAVE ORDERING

In addition to interorbital SC, the Kanamori Hamiltonian also provides the intraorbital SDW instability. Specifically, with the Hubbard interaction, SDW order appears when an appropriate nesting vector exists. Previous studies on the magnetic susceptibility of 214 have found strong intraband nesting for $\mathbf{q} \approx (\pm \frac{\pi}{2}, \pm \pi)$ to $(\pm \frac{\pi}{2}, \pm \pi)$ depending on TB parameters [25, 41]. However, no SDW order is found in unstrained 214 at low temperatures, suggesting that this nesting is not sufficient for SDW order. Similar nesting is present in 327, with $\mathbf{q} \approx (\pm \frac{\pi}{2}, 0)$ to $(\pm \frac{\pi}{2}, 0)$ due to the unit cell doubling. In addition, there also exists the same vector rotated by $\frac{\pi}{4}$ (i.e., along the $y$-direction instead). As in the case of 214, this nesting alone is not sufficient, as no SDW order forms at zero field. It was shown that SDW order is suppressed by SOC in 327 by reducing nesting [42], but the importance of SOC in this material has been recognized in explaining the dependence of the metamagnetic transition on the orientation of the field [42]. Additionally, experiments show a strong dependence of the formation of SDW order on the orientation of the field [16].

Although the nesting in 327 at zero field is not enough for SDW order to form, the presence of a van Hove singularity (vHS) was experimentally identified in the bilayer in the presence of a field [43], and it was suggested that the metamagnetic and nematic transitions are driven by the vHS [44–49]. We therefore consider the effect of nesting and vHSSs together on the formation of SDW order in 327. Analysis of the 327 TB model obtained via DFT reveals a peak in the DOS just below the Fermi level. Introducing a magnetic field splits the peak in two, leading to a significant change in the magnetization once one peak reaches the Fermi level. The Fermi surface under a field, shown in the inset of Fig. 3(a), shows the additional bands crossing the Fermi level near $(\pm \pi, 0)$ and $(0, \pm \pi)$.

Considering possible ordering wave vectors, the intraorbital, intralayer transverse susceptibility is shown in Fig. 3(a). Importantly, the peak in the susceptibility increases when a field is present due to the additional bands crossing the Fermi level. This ordering wave-vector corresponds to $\mathbf{q} \approx (0.3\pi, 0)$ in the reduced Brillouin zone, but slightly shifts as the field is changed. Details of the susceptibility calculations are found in Appendix B. To obtain numerical values, we perform self-consistent MFA calculations using Eq. (1) under a magnetic field. The region of SDW order is shown in blue in Fig. 1 where, for the choice of interaction and TB parameters, $B_{\perp} \approx 2.3 \times 10^{-3}$ and $B_{\parallel} \approx 6.4 \times 10^{-3}$ with $\mu_B$ set to 1. Setting $t_{\perp}^{2}$ to the DFT obtained value, $B_{\parallel}$ is of order 10 T. However, these values are sensitive to the location of the vHSS in energy and the values of hopping parameters which can be further reduced by correlation effects beyond the MFA. Thus the precise field values require further study beyond the scope of the current work.

The SDW order parameters and magnetization are shown in Fig. 3(b), where the spin component is in the $y$-
V. DISCUSSION AND SUMMARY

The absence of SC in 327 must be considered when explaining SC in 214 due to the similarity of the two materials. The most significant differences, bilayer coupling and staggered rotations, primarily affect interorbital SC and therefore provide a natural explanation for the lack of SC in 327. The presence of SDW order in 327 under a field is another piece of the ruthenate puzzle. The Kanamori Hamiltonian provides a consistent framework for both SC and SDW order. The Hund’s coupling plays an important role in stabilizing interorbital SC, while the Hubbard repulsion and Hund’s coupling together lead to SDW order. These interactions plus the increase in the DOS occurring in a field, experimentally observed as a metamagnetic transition, allow SDW ordering to form. The interactions presented here are limited to on-site, but further neighbor interactions have been considered in interorbital SC studies of 214 [50]. Their effect on SDW order remains to be studied.

Some limitations of our theory arise from the finite size of the TB model and MFA. Experiments detect multiple metamagnetic transitions and two SDW phases with different ordering wave-vectors all separated by less than 1T [51]. Electron correlations beyond the MFA were shown to be important in the formation of the heavy bands [52], important for reproducing the narrow range over which the metamagnetic transitions and SDW phases occur. The interorbital SC presented here requires $U' < J_H$ within the MFA. It was shown by DMFT and other studies that such a requirement is not necessary beyond the MFA [23–25, 53, 54]. To corroborate the earlier numerical studies, we calculated pairing correlators via exact-diagonalization on a small cluster and found interorbital pairing tendency occurring when $J_H$ is about 20% of $U$. This tendency is related to the changes of charge configurations via Hund’s coupling, shown in Appendix D.

An area of recent development requiring consideration going forward is possible SDW order in strained 214 [55]. While no experimental observations have been reported about the ordering wave-vector or spin direction, this appears similar to 327 since SDW order occurs near a vHS. The shift of the vHS however, is induced by strain, so while the $q_x$ and $q_y$ ordered states are degenerate in the bilayer, only one of these is chosen by the uniaxial strain. Further work is required to fully differentiate the two cases. Other recent experiments have studied surface layers of 214 [56], and the trilayer, Sr$_3$Ru$_2$O$_{10}$ [57], both of which feature staggered rotations. No SC is found in either, consistent with the current theory. However, future works are needed to estimate surface effects including broken inversion symmetry.

In summary, we showed that interorbital SC provides a natural explanation for both the presence of SC in 214 and lack of SC in 327. In 327, SC is destroyed by orbital hybridization introduced by staggered rotations, not present in 214. This same model finds intraorbital SDW ordering in 327 when the vHS reaches the Fermi level via a magnetic field. We believe that the lack of SC in 327 is an important piece of the puzzle to understand SC in 214, and therefore interorbital SC should remain among the promising candidates. Additionally, we predict that ideal (327)$_0$ can feature increased orbital degeneracy by tuning the bilayer coupling strength, and therefore can also exhibit interorbital SC with a possibly higher transition temperature.

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Appendix A: Tight-binding model

To obtain tight-binding parameters, we perform density functional theory calculations with the Vienna ab initio simulation package (VASP) [58], using the projector augmented-wave potential [59] and the Perdew-Burke-Ernzerhof exchange-correlation functional [60], with an energy cutoff of 400 eV. The relevant TB parameters are obtained using WANNIER90 [61], and are listed in Tables I and II.
\[ H_k = \sum_{k,l} t_{k,l} (c_{k,\sigma}^{\dagger} c_{l,\sigma} + c_{l,\sigma}^{\dagger} c_{k,\sigma}) + \sum_{k,l} \epsilon_{k,\sigma} c_{k,\sigma}^{\dagger} c_{l,\sigma} + \sum_{k} t_{1D} (c_{k,\sigma}^{\dagger} c_{k+1,\sigma} + c_{k,\sigma}^{\dagger} c_{k-1,\sigma}) + \sum_{k} t_{xy} (c_{k,\sigma}^{\dagger} c_{k+y,\sigma} + c_{k+y,\sigma}^{\dagger} c_{k,\sigma}) \]

where \( a \in \{yz, xz, xy\} \) represents the orbitals, and \( l \in \{t, b\} \) is the layer index. The intraorbital dispersions are,

\[ \epsilon_{k}^{xz(yz)} = -\mu_{1D} - 2t_{x,y}^{xz} \cos k_{x}(y) - 2t_{y,z} \cos k_{y}(x) \]

\[ + 4t_{x,y}^{xz} \cos k_{x} \cos k_{y} - 2t_{x,z}^{xz} \cos 2k_{x}(y) \]

\[ - 2t_{y,z}^{xz} \cos 3k_{x} - 4t_{x,z}^{xz} \cos 2k_{y}(x) \cos k_{y}(x), \]

\[ \epsilon_{k}^{xy} = -\mu_{xy} - \sum_{n} \left[ 2t_{x,y}^{x,y} \cos nk_{x} + \cos nk_{y} \right] \]

\[ + 4t_{x,y}^{x,y} \cos nk_{x} \cos nk_{y} \]

\[ - \sum_{m \neq n} 4t_{m,x,y}^{x,y} \cos mk_{x} \cos nk_{y} + \cos nk_{x} \cos mk_{y} \]

where \( m \) and \( n \) are the integers listed in Table II describing the intraorbital hopping within the xy orbitals between sites separated by \( m \hat{x} + n \hat{y} \) as well as \( n \hat{x} + m \hat{y} \) by symmetry. The dispersion of the xy orbital includes further neighbor hopping terms to model the flatness of the bands near \((\pm \pi, 0)\) and \((0, \pm \pi)\) of the Brillouin zone in the ideal bilayer material. The orbital mixing is given by,

\[ t_{k} = -4t_{1D} \sin k_{x} \sin k_{y}. \]

The interlayer hoppings, \( t_{\perp}^{z,l} \) and \( t_{\perp}^{x,y} \), represent hopping between layers within orbitals directly above or below each other, while \( t_{\perp}^{x,z} \) is hopping in the xz(yz) orbital between layers and one lattice site over in the x(y)-direction, and \( t_{\perp}^{x,y/z} \) hopping between layers, one lattice site over in the \( y(x) \)-direction, between \( xy \) and \( xz(yz) \) orbitals. The staggered hopping, \( t_{stag}^{\perp} \), allowed by the staggered rotation, represents the hopping between NN 1D orbitals within a layer and changes sign between layers due to the opposite sense of the octahedra rotations between the layers, while \( t_{stag}^{\perp} \) occurs between 1D orbitals directly above or below each other. Finally, the SOC Hamiltonian is,

\[ H_{SOC} = i\lambda \sum_{i} \sum_{a,b} \epsilon_{ab} c_{i,\sigma}^{\dagger} \sigma_{i,\sigma}^{a} \sigma_{i,\sigma}^{b} \]

where the strength of the SOC used is set to \( \lambda = 62.5 \text{ meV} \) throughout this work.

In calculations which feature a magnetic field in the z-direction, the additional term included in the Hamiltonian is,

\[ H_{B} = -B_{z} \sum_{i} (2S_{i}^{z} + L_{i}^{z}). \]

The total z-magnetization calculated in the presence of the field is given by,

\[ M_{z} = \sum_{i} (2S_{i}^{z} + L_{i}^{z}) \]
when a field is introduced. a paramagnet at zero field and only exhibits SDW order conductivity exists in 214 and (327)

\[ t_{x,y}^z \begin{array}{cccc} 270.2 & 12.1 & -15.1 & -29.8 & -10.1 \\ 434.6 & 12.6 & 282.9 & 281.6 & 90.6 \\ 294.3 & 137.2 & 32.5 & 13.6 & 22.1 \end{array} \]

\[ t_{x,y}^2 \begin{array}{cccc} -3.6 & 12.6 & -12.5 & 10.3 & 1.34 \end{array} \]

\[ t_{x,y}^3 \begin{array}{cccc} -1.2 & 1.1 & 1.3 & -3.76 \end{array} \]

\[ t_{3x,y}^4 \begin{array}{cccc} 400.7 & 18.6 & 13.6 & 84.5 & 64.3 \end{array} \]

TABLE II. Tight-binding parameters obtained for the real bilayer, 327. All values are shown in units of meV. Note that the chemical potential listed here is slightly below a filling of 2/3 to ensure that the vHS is close enough to the Fermi level to produce the metamagnetic transition at a field value near the experimentally observed 7.9 T. The chemical potential used corresponds to a filling fraction of approximately 0.657.

Appendix B: Susceptibility

Defining the components of the spin operator for orbital \( \alpha = (yz, xz, xy) \) and layer \( l = (t, b) \) as,

\[
S^a_{\alpha l}(q) = \sum_{k,\sigma,\sigma'} c^a_{k\alpha\sigma} \frac{\sigma^a_{\alpha l\sigma}}{2} c_{k+q\alpha l\sigma'},
\]

\[
\chi^{ab}_{\alpha l}(q, i\omega_n) = -\frac{1}{4N} \sum_{k,s,s',\sigma_1,\sigma_2,\sigma_3,\sigma_4} u^*_{\alpha l\sigma_1,s\sigma_2} u_{\alpha l\sigma_2,s'}(k+q) u^*_{\beta m\sigma_4,s'}(k+q) u_{\beta m\sigma_3,s}(k)
\]

Below we show the intra-layer contribution to the transverse susceptibility from each orbital, \( \sum_{\alpha}[\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q) \), the intra-layer contribution summed over the orbital index, \( \sum_{\alpha,\beta l}[\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q) \), and the inter-layer contribution summed over the orbital index, \( \sum_{\alpha,\beta, l \neq m}[\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q) \).

Appendix C: Full Phase Diagram

To show the behaviour of all three models considered for various sets of interaction parameters, the full phase diagrams are shown in Fig. 5. The superconducting region expands when the bilayer coupling is added to the single layer model, however, the superconducting region shrinks and the spin-density wave region is expanded when the staggered rotations are added. The asterisks correspond to the values used in the main text, where superconductivity exists in 214 and (327)\( _b \), while 327 remains a paramagnet at zero field and only exhibits SDW order when a field is introduced.

\[
\chi^{ab}_{\alpha l}(q, i\omega_n) = \sum_{\alpha,\beta, l, m} [\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q, i\omega_n),
\]

\[
[\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q, i\omega_n) = \frac{1}{N} \int_{0}^{\beta} d\tau e^{i\omega_n\tau} (T_{\alpha l}\sigma_{\alpha l}(q, \tau)\sigma_{\beta m}(q, 0))
\]

where \( \beta = 1/\tau \), \( \tau \) is imaginary time and \( T_{\alpha l} \) is the imaginary time-ordering operator. To evaluate this we use the transformation to the band basis given by, \( c_{kn} = \sum_{s} u_{ns}(k)c_{ks} \), where \( n = (\alpha, l, \sigma) \), \( s \) indexes the bands, and \( u_{ns}(k) \) are the coefficients of the unitary transformation. Evaluating the correlation function in terms of the Green’s functions in the band basis and subsequently the Matsubara sum via contour integration, with the analytic continuation \( \omega_n \rightarrow w + i\eta \), the components of the static susceptibility are given by

\[
\chi^{ab}_{\alpha l}(q, i\omega_n) = \sum_{\alpha,\beta, l, m} [\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q, i\omega_n),
\]

\[
[\chi^{+\alpha -\alpha}_{\alpha l}]_{\beta m}(q, i\omega_n) = \frac{1}{N} \int_{0}^{\beta} d\tau e^{i\omega_n\tau} (T_{\alpha l}\sigma_{\alpha l}(q, \tau)\sigma_{\beta m}(q, 0))
\]

B(2)

Appendix D: Exact Diagonalization Correlators

To extend the consideration of SC beyond the MFA, in Fig. 6, we show an exact diagonalization calculation for a three-site cluster as shown by the dots below the curve Fig. 6, we show an exact diagonalization calculation for a three-site cluster as shown by the dots below the curve.

\[
\Delta_{xz/xy}^x \Delta_{yz/xy}^y \]

operators start to increase at \( J_H/U \approx 0.18 \), and are matched by an increase in charge fluctuations away from the \( d_{1}^{x}d_{2}^{y}d_{3}^{z} \) configuration to configurations such as \( d_{1}^{x}d_{2}^{y}d_{3}^{z} \).
Figure 4. Transverse susceptibility of the 327 model in the presence of a finite field. The intraorbital, intralayer susceptibilities are individually plotted, which provide the primary contributions to the ordering. The total intralayer susceptibility including intraorbital and interorbital contributions is also shown. Additionally, the total interlayer susceptibility is plotted, which, when included may modify the ordering wave vector due to the differing locations of its peaks.

Figure 5. Full phase diagram of Eq. (1) from the main text for each of the TB sets in Tables I and II, where $V_{SDW} = U$ and $V_{SC} = 3J_H - U$. All calculations are performed without a field. In the presence of a field, the SDW region of the 327 model expands due to the vHSs crossing the Fermi level. The asterisks correspond to the values used in the calculations presented in the main text.

where $d_i^n$ refers to $n$ electrons at site $i$. The weight of the $d_1^4d_2^4d_3^4$ and $d_1^1d_2^5d_3^3$ configurations in the ground state wavefunction are shown in the upper left inset of Fig. 6, denoted $a_{444}$ and $a_{453}$, respectively. Other configurations also contribute to the ground state wavefunction, but are omitted here. The inclusion of electron fluctuations as allowed by these calculations shows that the strict requirement of $J_H/U > 1/3$ is only necessary in the MFA where the effects of charge configurations such as $d_1^4d_2^5d_3^3$ combined with Hund’s coupling cannot be taken into account.
Figure 6. Pairing correlators from three-site exact diagonalization calculations as a function of $J_H/U$, calculated for the 214 tight-binding parameters shown in Table I, and the Kanamori Hamiltonian using $U = 3.5$. The correlators are finite before the MFA required minimum ratio of $J_H/U > 1/3$, suggesting that electron fluctuations can reduce the required Hund’s coupling necessary for SC to form. The electron configurations are shown in the upper left inset, where $a_{n_1n_2n_3}$ refers to the weight of $d_{n_1}^1d_{n_2}^2d_{n_3}^3$ configuration in the wavefunction, where $n_i$ represents the number of electrons at site $i$. The pairing correlators become finite at the interaction ratio corresponding to the sudden drop of the weight of the $d_{1}^1d_{2}^1d_{3}^3$ configuration and increase in the $d_{1}^1d_{2}^2d_{3}^3$ configuration, boosted by Hund’s coupling. The $a_{444}$ and $a_{453}$ weights are shown, but other configurations also contribute to the wavefunction. The geometry of the three-site configuration is shown under the curve.