I. INTRODUCTION

The concept of excitonic condensation has attracted considerable attention since its early theoretical prediction [1]. An exciton is a bound state of an electron-hole pair. This composite particle resembles the Cooper pair of superconductors and follows hard-core bosonic statistics. Early work involving semiconductors showed that in the weak-coupling limit (small $U$), near the semimetal to semiconductor transition, the system can become unstable against the formation of multiple excitons [2,3]. This can lead to a condensation into a BCS-like macroscopic state called excitonic insulator. The associated BCS wavefunction can smoothly transform into a BEC state when the gap between bands increases at fixed Hubbard repulsion $U$. In strongly correlated systems, a BCS-like excitonic insulator can also occur in the strong coupling limit (large $U$). This regime attracted theoretical investigations [4–6] and was studied using large $U$ perturbation theory, where the hopping amplitude $t$ is the small parameter.

The extended Falicov-Kimball model has often been used as a minimal theoretical model to address excitonic condensation [7–9]. Only recently, more realistic, and more difficult, three-orbital Hubbard models were explored to study this state [10–13]. On the experimental side there have been studies showing reliable signatures of the excitonic condensate in real materials, such as in transition metal dichalcogenides [14], Ta$_2$NiSe$_5$ [15], and in bilayer systems [16,17]. To better understand excitonic insulators it is important to find additional candidate materials and additional theoretical models where this state occurs and can be studied in detail. Recently, it was recognized that materials with strong spin-orbit coupling provide a new avenue to realize this physics [18,19]. For example, Sr$_2$IrO$_4$, with Ir$^{4+}$ ions and filling $n = 5$ electrons ($t_{2g}^4$), is a celebrated material due to the presence of long-range antiferromagnetic ordering in quasi-two-dimensional layers, as in superconducting cuprates [20]. Recent resonant inelastic x-ray scattering (RIXS) experiments on Sr$_2$IrO$_4$ have reported excitons as an excitation at approximately 0.5–0.6 eV [21–23]. RIXS experiments on one-dimensional stripes of Sr$_2$IrO$_4$ also indicated excitons at nearly the same energy [24]. These excitons are present in spin-orbit entangled states, hence known as spin-orbit excitons.

Another promising avenue for excitonic condensates induced by spin-orbit coupling involves the $t_{2g}$ case, which is the focus of the present paper. Theoretically, it was predicted that the three-orbital Hubbard model with a degenerate $t_{2g}$ space and in the $LS$ coupling limit ($U \gg t$, $\lambda$) leads to the BCS state of triplons (singlet-triplet excitations) [25–27]. We will show that these triplon excitations are low-energy manifestations of spin-orbit excitons. Dynamical Mean Field Theory (DMFT) calculations also supported similar findings [10,11], although it is difficult to distinguish between BCS and BEC states using this technique. A recent computational study of the ground state of a one-dimensional spin-orbit coupled three-orbital...
Hubbard model—in a nondegenerate (tetragonal) $t_{2g}$ space using the numerically-exact density matrix renormalization group (DMRG)—also reported a phase with staggered spin-orbit excitonic correlations [12]. All the above mentioned studies revealed an antiferromagnetic ordering accompanying the excitonic condensate. The $t_{2g}$ case is relevant for materials with Ir$^{5+}$ ions and other 4$d/5$d transition metal oxides with the same doping $n = 4$. The presence of triplon condensation was initially discussed for double perovskite materials, such as Sr$_2$YIrO$_6$ and Ba$_2$YIrO$_6$ with a 5$d^4$ configuration [28–33], but RIXS experiments showed that the triplon excitations bandwidth is not sufficiently large compared to $\lambda$ to develop condensation [34]. Note that recent RIXS experiments on Ca$_2$RuO$_4$ suggests that this could be a candidate material for excitonic magnetism [35], and ab initio calculations have reached the same conclusion [36].

To investigate the spin-orbit excitonic condensation, in this paper, we use a simple degenerate three-orbital Hubbard model. Using numerically exact DMRG simulations on one-dimensional chains, we show that an excitonic condensation is induced accompanied by antiferromagnetic ordering even in the intermediate Hubbard regime ($U/W \approx 1$). This regime is stabilized by increasing sufficiently $\lambda$ starting at the $\lambda = 0$ incommensurate spin-density wave metallic phase. These results cannot be understood using large $U$ perturbation theories. Moreover, by using the Hartree-Fock approximation (HFA) in two-dimensional ($2d$) lattices, we also found a similar excitonic insulator phase, both in the weak and intermediate $U/W$ regimes. As our main result, we show that there is a BCS-BEC crossover inside the excitonic condensate phase, both in 1$d$ and 2$d$ lattices. The BCS limit of the excitonic insulator occurs at intermediate $U/W$ (and also for weak $U/W$ in $2d$), and by increasing $\lambda$ (at fixed $U/W$) the BEC state is reached. The previously known large $U/W$ BEC state, due to triplon condensation, appears smoothly connected to the BCS excitonic insulator of intermediate $U$. We also provide the full $\lambda$ vs $U$ phase diagrams for 1$d$ and 2$d$ lattices using the many-body techniques discussed above.

The organization of this paper is as follows. In Sec. II, the model and the computational methodology used are presented. In Secs. III and IV, the main results are shown. In particular, in Sec. III, the results on 1$d$ lattices using the DMRG technique are presented. In Sec. IV, the results on 2$d$ lattices using HFA further support our main proposal of a BCS-BEC crossover in the model studied. In Sec. V, we discuss the overall results and present our conclusions.

II. MODEL AND METHOD

For our study, we use a degenerate three-orbital Hubbard model. The Hamiltonian has three primary terms: the tight-binding kinetic energy, the standard on-site multiorbital Hubbard interaction, and the on-site spin-orbit coupling (SOC): $H = H_K + H_{\text{int}} + H_{\text{SOC}}$. The tight-binding term is

$$H_K = \sum_{\langle i,j \rangle, \sigma, \gamma, \gamma'} t_{\gamma \gamma'} (c_{i \sigma \gamma}^{\dagger} c_{j \sigma \gamma'}) + \text{H.c.}. \quad (1)$$

The hopping amplitudes $t_{\gamma \gamma'}$ connect only nearest-neighbor lattice sites (in both the 1$d$ chain and 2$d$ square lattices). As discussed earlier, here we use degenerate orbitals, i.e., $t_{\gamma \gamma'} = \delta_{\gamma' \gamma} t$. We fixed $t = 0.5$ and all of our results are presented in terms of dimensionless ratios, such as $U/W$ and $\lambda/W$. The total bandwidth is $W = 4.0|t|$ and $8.0|t|$ for the 1$d$ and 2$d$ lattices, respectively. The orbitals used—labeled as 0, 1, and 2—could be associated respectively to the $d_{xz}, d_{yz}$, and $d_{xy}$ orbitals, namely the $t_{2g}$ sector. In real materials, the above hoppings can be much more complex [37], but nonetheless this simple model is useful to gain conceptual understanding. The on-site multiorbital Hubbard interaction term consists of the standard several components

$$H_{\text{int}} = U \sum_{\gamma, \gamma'} n_{\gamma \gamma} n_{\gamma \gamma'} + (U' - J_H/2) \sum_{\gamma, \gamma' \neq \gamma} n_{\gamma \gamma} n_{\gamma' \gamma'}$$

$$-2J_H \sum_{\gamma', \gamma} S_{\gamma'} \cdot S_{\gamma'} + J_H \sum_{\gamma, \gamma' \neq \gamma} (P_{\gamma}^\dagger P_{\gamma'} + \text{H.c.}). \quad (2)$$

$S_{\gamma} = \frac{1}{2} \sum_{\alpha, \beta} c_{\alpha \gamma}^\dagger c_{\beta \gamma}$ represents the spin at orbital $\gamma$ and lattice site $i$, while $n_{\gamma} = \text{Tr}S_{\gamma}$ is the electronic density operator also at orbital $\gamma$ and site $i$. The first two terms are the intra- and interorbital electronic repulsion, respectively. The Hund coupling is the third term, favoring the spins ferromagnetic alignment at different orbitals and the same site. Finally, the $P_{\gamma} = c_{\gamma}^\dagger c_{\gamma}$ operator in the fourth term (pair hopping) is the pair annihilation operator arising from the matrix elements of the “1/r” Coulomb repulsion as in the early studies of Kanamori. We used the standard relation $U' = U - 2J_H$ due to rotational invariance, and we fix $J_H = U/4$ for all the calculations as employed widely in previous efforts [12,13,38–40].

The SOC term is

$$H_{\text{SOC}} = \lambda \sum_{\gamma, \gamma', \gamma''} \langle \gamma | L_{\gamma'} | \gamma'' \rangle \cdot \langle \sigma | S_{\gamma} | \sigma' \rangle c_{\gamma''}^{\dagger} c_{\sigma'} , \quad (3)$$

where the coupling $\lambda$ is the SOC strength.

Using the model described above, we performed calculations on one-dimensional chains employing the DMRG [41,42] for various system lengths $L = 16, 24$, and 32 sites. We kept up to 1600 states for the DMRG process and maintained a truncation error below $10^{-8}$ throughout the finite algorithm sweeps. We used the corrected single-site DMRG algorithm [43] with correction $a = 0.001–0.008$, and performed 35 to 40 finite sweeps to gain proper convergence depending on the system size. After this convergence, we calculated the spin-structure factor $S(q)$, orbital-structure factor $L(q)$, excitonic momentum distribution function $\Delta_{m=1/2}(q)$, and coherence length $\xi_{\text{coh}}$. From all these observables, we constructed the phase diagram. To calculate spectral functions with the DMRG, we used the correction vector method [44], with the Krylov-space approach [45]. We obtain the single-particle spectral function $A(q, \omega)$ and the excitonic pair-pair susceptibility $\Delta_{m=1/2}(q, \omega)$. These frequency-dependent observables require considerable computational time, and multiple compute nodes. In our DMRG, we target the total $F_{\text{eff}}$ of the system to reduce the computational cost [12]. Details of the Hartree-Fock calculations are in Ref. [46].
FIG. 1. Visual representation of our main results, all supported by actual DMRG and Hartree Fock calculations. In (a), the single-particle excitations of the $j_{\text{eff}} = 3/2$ and $j_{\text{eff}} = 1/2$ bands are shown at intermediate $U$, where near the chemical potential a gap opens due to the formation of excitons (note electron and hole have the same $m$). The $j_{\text{eff}} = 3/2$ and $j_{\text{eff}} = 1/2$ bands open gaps near momentum $q = 0$ and $q = \pi$, respectively. In (b), the real-space perspective of the excitonic state (at intermediate $U$) is shown, where the exciton’s mean radius (characterized by the coherence length) decreases by increasing $\lambda$. In (c), the excitonic condensation in strong coupling is depicted. The local exciton creation operator leads to the creation of both a triplon and a quintuplon when acting on $|\text{exciton}\rangle$. This exciton condensation opens a gap in the distribution functions of the local excitations in the BCS and BEC limits in one dimension. The excitonic operator $\Delta_{1/2,m}^{1/2,0} = \langle j_{1/2,m} j_{1/2,m} \rangle$ in the single-atom LS coupling limit can be written using triplon and quintuplon excitations (see Supplementary [46]), corresponding, respectively, to $T_{\text{eff}}^\dagger |\text{eff}\rangle = 0, J_{\text{eff}} = 0 = |J, 1\rangle$, and $Q_{\text{eff}}^\dagger |\text{eff}\rangle = 0, J_{\text{eff}} = 0 = |J, 2\rangle$, where $n \in \{\pm 1, 0\}$ and $l \in \{\pm 2, \pm 1, 0\}$. Calculating the pair-pair susceptibility of excitons $\Delta(q, \omega)$ for chains, in the strong-coupling nonmagnetic phase, we found bands of triplon and quintuplon excitations, with minima at $q = \pi$ but both bands are gapped. Decreasing $\lambda$ drives the system to the BEC state, where $\Delta(q, \omega)$ has gapped triplon and quintuplon excitations only near $q = 0$ but mainly a continuum of excitations at other momenta above the Goldstone-like modes emerging from $q = \pi$, as sketched in Fig. 1(c).

A. Magnetic properties and staggered excitonic correlations

Now consider the results for magnetism in one-dimensional chains. We choose $U/W = 2.0$ and $10.0$ as representative points for the intermediate and strong coupling regions, respectively. First, consider the intermediate coupling region where at $\lambda = 0$ we found an incommensurate spin-density wave (IC-SDW) via the spin-spin correlation $\langle S_i \cdot S_j \rangle$ and $\langle L_i \cdot L_j \rangle$, as shown in Figs. 2(a,c). Block
magnetic states as in previous DMRG studies [12,38–40], do not appear in our model at \( \lambda = 0 \). Increasing \( \lambda \) drives the system towards antiferromagnetism with staggered spin-spin and orbital-orbital correlations, see Figs. 2(b) and 2(d). For additional confirmation, we show the spin structure factor \( S(q) = (1/L) \sum_{j,m} e^{i q (j-m)} (S_j \cdot S_m) \), and the orbital structure factor \( L(q) = (1/L) \sum_{j,m} e^{i q (j-m)} (L_j \cdot L_m) \) for various \( \lambda \) values. As \( \lambda \) increases, the incommensurate peak in \( S(q) \) in Fig. 2(e) shifts to lower \( q \) values. The antiferromagnetic tendencies, shown by the \( q = \pi \) peak, starts only near \( \lambda = 0.22W \), and on further increasing \( \lambda \) the \( q = \pi \) peak grows and the incommensurate peak is reduced. At larger \( \lambda \)'s, the \( S(q = \pi) \) peak decreases as the system transitions into the nonmagnetic state. The orbital structure factor \( L(q) \) displays similar behavior at \( q = \pi \). \( L(q) \) starts with a flat plateau near \( q = \pi \), then grows when increasing \( \lambda \), and eventually the \( L(q = \pi) \) peak vanishes for very large \( \lambda \), see Fig. 2(f).

In strong coupling and at \( \lambda = 0 \), the spins align ferromagnetically and orbital-orbital correlations show a \( +−+−... \) pattern, with peaks at momentum \( q = 0 \) and near \( q = 2\pi/3 \) in \( S(q) \) and \( L(q) \), respectively (Fig. 3). Recent DMRG calculations on the low-energy \( S = 1 \) and \( L = 1 \) model showed similar results in the orbital correlations [47] at \( \lambda = 0 \). As \( \lambda \) increases, the system enters into the phase where orbital ordering becomes staggered, as shown by a peak at \( q = \pi \) for \( \lambda \approx 0.05W \) in Fig. 3(a). However, the spin ordering is dominantly ferromagnetic with secondary antiferromagnetic tendencies leading to a small peak at \( q = \pi \) [Fig. 3(b) for \( \lambda = 0.05W \)]. Further increasing \( \lambda \), both \( L(q = \pi) \) and \( S(q = \pi) \) grow while \( S(q = 0) \) decreases, and ultimately \( L(q = \pi) \) and \( S(q = \pi) \) also start decreasing when the system enters into the nonmagnetic phase with exponentially decaying spin and orbital correlations.

In our DMRG calculations, the development of antiferromagnetic correlations in the spin and orbital channels is always accompanied by staggering in the exciton-exciton correlations, both at intermediate and strong coupling. We estimate the amount of staggering in excitonic correlation for our chains using:

\[
\Delta_m = \frac{1}{L} \sum_{|i-j|>0} (-1)^{|i-j|/2} (\Delta_{1/2,m}(i) \Delta_{1/2,m}(i') - \Delta_{1/2,m}(i) \Delta_{1/2,m}(i')^*),
\]

where \( m \in \{±1/2\} \). In Fig. 3(c), the evolution of \( \Delta_{1/2} \) is shown when changing \( \lambda \), where each panel belongs to a different \( U/W \). We found that smaller \( \lambda \)'s are needed as \( U/W \) increases to obtain staggered excitonic correlations. A finite range of \( \lambda \) where the excitonic correlations are staggered is present for \( U/W \) as low as 1.5, although the magnitude of \( \Delta_{1/2} \) decreases as \( U/W \) decreases, and for \( U/W \lesssim 1.0 \), we were unable to identify—within our numerical accuracy—the region with staggered excitonic correlations. Nonetheless, it is interesting that we find staggered excitonic correlations at intermediate \( U \), where small \( \lambda \) shows IC-SDW metal (for evidence of metallicity see Sec. III C). Perturbation theories at \( U \gg 1 \), \( \lambda \) cannot explain these results.

B. BCS-BEC crossover and IC-SDW metal to BCS-Excitonic insulator transition

Now we will discuss the main result of this paper, where we present the numerical evidence for the BCS-BEC crossover in the excitonic condensate. At intermediate \( U/W \)—the value

![FIG. 2. DMRG results at \( U/W = 2 \). (a) and (b) show the real-space spin-spin correlations at \( \lambda/W = 0 \) and \( \lambda/W = 0.29 \), respectively. In (c) and (d), the real-space orbital-orbital correlations are shown at \( \lambda/W = 0.0 \) and \( \lambda/W = 0.29 \), respectively. The spin structure factor \( S(q) \) and orbital structure factors \( L(q) \) are in (e) and (f), respectively, for various \( \lambda \). In (a)–(d), the system size was \( L = 32 \) and one site is fixed at the center, i.e., \( i = 15 \). A system size \( L = 16 \) is used for (e,f).](image-url)
$U/W = 1.75$ is chosen merely for simplicity—there is a finite range of $\lambda/W \in [0.28, 0.4)$ where staggering in excitonic, spin, and orbital correlations is present. We calculate the coherence length $r_{coh}(m)$ using the widely employed formula [4,48,49]

$$r_{coh}(m) = \sqrt{\sum_{i,j} |i-j|^2 |a_i^\dagger a_j^\dagger a_j a_i|^2} \sqrt{\sum_{i,j} |a_i^\dagger a_j^\dagger|^2},$$

(5)

where $m \in \{\pm 1/2\}$, for the points lying inside the excitonic condensate region at fixed $U/W = 1.75$, namely, the Path-1 shown in Fig. 6. Note that $r_{coh}(1/2) = r_{coh}(-1/2)$. Interestingly, we found that as $\lambda$ increases, $r_{coh}$ decreases from nearly one lattice spacing $\approx a$ to $\approx 0.2a$, see Fig. 4(c). This reduction in the coherence length of electron-hole pairs resembles the BCS-BEC crossover already discussed in semiconductors near the semi-metal to semiconductor transition [1-3]. We also calculate $r_{coh}$ for various values of $\lambda$ and $U$ when transitioning from the intermediate to the strong coupling limits, while being still inside the excitonic condensate, which we call Path-2 (see Fig. 6), as shown in Fig. 4(d). Here again we found that $r_{coh}$ decreases from $O(a)$ to $O(0.1a)$. The ordered $(\lambda/W, U/W)$ points chosen for Path-2 are $(0.3, 1.75), (0.29, 2.0), (0.29, 2.25), (0.287, 2.3), (0.285, 2.35), (0.283, 2.4), (0.28, 2.5), (0.279, 2.525), (0.277, 2.55), (0.275, 2.6), (0.27, 2.65), (0.25, 2.8), (0.24, 3.0), (0.23, 3.5), (0.21, 4.0), (0.2, 4.5), (0.17, 6.0), (0.15, 7.0), (0.13, 9.0), (0.11, 10.0)). Note that for $U/W \approx 2.75$, the coherence length is small as expected because at these values of $U$, the system is already in strong coupling for $\lambda = 0$.

In Figs. 4(a) and 4(b), we show the excitonic momentum distribution function $\Delta(q) = (1/L) \sum_{i,j} \langle e^{iq(i-j)} a_{i+q}^\dagger a_j \rangle$ for two points $(\lambda/W = 0.3, U/W = 1.75)$ and $(\lambda/W = 0.15, U/W = 7.0)$, at the intermediate $U$ BCS and strong $U$ BEC regions. In the BCS limit, $\Delta(q = \pi)$ grows very slowly with system size $L$. However, in the BEC region we found nearly linearly increasing $\Delta(q = \pi)$ with $L$, implying either a very slow power-law decay or even true long-range order. Such a true long-range order in our one dimension is allowed as the $U(1)$ symmetry related to the excitonic condensation is explicitly broken by a finite Hund coupling [13]. The analysis above also clearly implies that as we increase the system size and hence increase the number of excitons, these excitons can condense also at momentum $q \neq \pi$ in the BCS limit, whereas in the BEC limit excitons condense only at $q = \pi$. For completeness, note that a similar BCS-BEC crossover has
also been reported in the extended Falicov-Kimball model in one-dimensional chains [7].

As discussed before, the exciton creation (i.e., electron-hole pair excitation) becomes the triplon and quintuplon excitation in the atomic LS limit. We calculated the excitonic pair-pair susceptibility

$$\Delta(q, \omega) = \langle \Psi_0 | \Delta^{13/2,1/2}_0(q) | \omega + i \eta - H + E_0 \Delta^{3/2,1/2}_0(q) | \Psi_0 \rangle$$ (6)

at large $U/W = 10.0$ on one-dimensional chains to study the effect of the kinetic energy. The broadening $\eta$ was 0.05eV. We choose two values $\lambda/W = 0.20$ and $\lambda/W = 0.11$ in the nonmagnetic insulator and BEC regions, respectively [Figs. 4(e) and 4(f)]. For the nonmagnetic state, we found the pair-pair susceptibility shows two cosine-like bands with minima at $q = \pi$, where the lower band belongs to $\Delta J_{\text{eff}} = 1$ (triplon) and the upper band represent $\Delta J_{\text{eff}} = 2$ excitations, as in previous analytical studies [27]. The BEC is expected to occur by reducing $\lambda$, when the lower band of triplons becomes gapless. In our BEC state, $\Delta(q, \omega)$ shows features very different from those in the nonmagnetic state: after removing the elastic peak we found that two gapped bands appear only near $q = 0$, and the spectrum at $q = \pi$ is now gapless with emerging Goldstone-like modes. We also found continuum-like features for $q \in \{ -\frac{\pi}{4}, \frac{3\pi}{4} \}$.

In the noninteracting limit, $\lambda$ only splits the $j_{\text{eff}} = 3/2$ and $j_{\text{eff}} = 1/2$ bands, see Fig. 5(a), driving a metal to band-insulator transition. Only at finite $U$ the excitonic condensation happens: as shown in our DMRG results, $U/W \gtrapprox 1.0$ is required to obtain a noticeable staggering in the excitonic correlations. To further investigate the intermediate $U$ region, we calculated the single-particle spectral function $A_{jm}(q, \omega)$ (see Ref. [46]) at $U/W = 2.0$, using both $\lambda = 0.0$ and $\lambda/W = 0.29$ corresponding to the IC-SDW and excitonic condensate with $r_{\text{coh}} \approx 1.0$ (BCS limit), respectively. In Fig. 5(b), we show $A_{jm}(q, \omega - \mu)$ for $\lambda = 0$: at this point all $\{j_{\text{eff}}, m\}$ states are degenerate. Comparing to the noninteracting limit band structure, a renormalization of the bands is clearly visible, having two minima structure and a local maxima at $q = \pi$, as a consequence of the Hubbard repulsion. In this phase, the nesting vector at the chemical potential $\mu$ decides the ordering vector of the incommensurate spin-density wave. We also show the single-particle density of states $\rho_{jm}(\omega - \mu)$, Fig. 5(c), which indicates that the system has a finite density of states at $\mu$ suggesting this phase is metallic.

Figures 5(d) and 5(e) show $A_{jm}(q, \omega - \mu)$ for $j_{\text{eff}} = 3/2$ (all $m \in \{\pm 3/2, \pm 1/2\}$ are degenerate) and $j_{\text{eff}} = 1/2$ (both $m \in \{\pm 1/2\}$ are degenerate), respectively. The gaps at $\omega$, at wavevectors $q \approx 0$ and $q \approx \pi$ in the spectral functions of $j_{\text{eff}} = 3/2$ and $j_{\text{eff}} = 1/2$, respectively, are clearly present. These gaps appear due to the formation of bound states of electrons and holes, arising from the $q \approx 0$ and $q \approx \pi$ states of the $j_{\text{eff}} = 3/2$ and $j_{\text{eff}} = 1/2$ bands, respectively. This leads to excitons creation with net momentum $\pi$ (indirect excitons). We also noticed a nontrivial suppression of the

FIG. 5. The single-particle spectral function calculated using DMRG. The noninteracting band structure is shown in (a). (b) contains $A_{jm}(q, \omega)$ for $\lambda = 0$ and $U/W = 2.0$, in the IC-SDW metallic phase. In (d) and (e), $A_{jm}(q, \omega)$ at $\lambda/W = 0.29$ and $U/W = 2.0$ is shown inside the excitonic insulator phase ($r_{\text{coh}} = 1.019$). (c) and (f) show the single-particle DOS [$\rho_{jm}(\omega)$] at $\lambda = 0.0$ and 0.29, respectively. The scaling of the charge gap is in inset (g) for ($U/W = 2, \lambda/W = 0$) and ($U/W = 2, \lambda/W = 0.29$).

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spectral function near $q = \pi$ for $j_{\text{eff}} = 3/2$, below $\mu$, but the explanation for these small features requires further work. However, it is evident that the gap opens due to the formation of indirect excitons and eventually leads to a BCS-like state. In Fig. 5(f), the $j$-resolved density-of-states is shown to illustrate the suppression near $\mu$ for both the $j_{\text{eff}} = 3/2$ and $1/2$ bands. A small but finite density-of-states at $\mu$ is present because of the broadening $\eta$ used. To confirm the transition from metal (at $\lambda = 0$) to excitonic insulator (at $\lambda/W = 0.29$), we performed finite-size scaling of the charge gap $\Delta_c = E_G(N + 1) + E_G(N - 1) - 2E_G(N)$ for both points, see Fig. 5(g). At $\lambda = 0.29$, using system sizes $L = 8, 12, 16, 24, 32$, and 42, we found the charge gap is quite robust 0.55 eV. Sizes $L = 8, 16, 20, 28, 32$, and 40 were used for the scaling at $\lambda = 0$, which indicates that $\Delta_c$ scales to $\approx 0$ eV in the thermodynamic limit.

Figure 6 ends this section by displaying the full $\lambda$ vs $U$ phase diagram for our one-dimensional systems. The CPU-costly DMRG calculations were performed for all small circles shown using a system size $L = 16$. After obtaining the ground state, then spin-spin correlations, orbital-orbital correlations, and exciton-exciton correlations were calculated to analyze the properties of each phase. The $(j_{\text{eff}}, m)$-resolved local electronic densities were also studied to identify the relativistic band insulator phase. The dashed line inside the excitonic condensate region (green region) is a guide to the eyes to show the BCS and BEC limits of the excitonic condensate.

IV. HARTREE-FOCK RESULTS IN TWO-DIMENSIONS

In this section, we will present and discuss the results obtained in two-dimensional lattices by performing mean-field calculations in real-space. Hartree-Fock approximations are appropriate to study excitonic condensates as shown in the original publications that started this field [1], as well as in recent numerical calculations on Falicov-Kimball models [8,50].

In Fig. 7(a), we show the full $\lambda$ vs $U$ phase diagram for the square lattice, calculated using the Hartree-Fock approximation. In (b) and (c), the momentum distribution function of excitons $\Delta(q)$ is shown at $\lambda = 0.53$ and $\lambda/W = 0.592$, respectively, for fixed $U/W = 0.5$ and a $24 \times 24$ system. The coherence length for various values of $\lambda$, at fixed $U/W = 0.5$, for system sizes $8 \times 8$, $16 \times 16$, and $24 \times 24$ are in (d). The $N(\pi, \pi)$, $S(\pi, \pi)$, and $L(\pi, \pi)$ are in (e) for various values of $\lambda$, at fixed $U/W = 0.5$ and using a $24 \times 24$ cluster.
metal to excitonic insulator at intermediate $U$, by calculating single particle density-of-states (see Ref. [46]). It must be noted that excitonic magnet, at large $U$, found in Hartree-Fock calculations cannot be described by condensation of triplons. In the intermediate $U$ region, the band insulator is present at large $\lambda$ with excitons as local objects and the band gap is larger than the binding energy of excitons. As $\lambda$ decreases, the excitonic magnet emerges due to the condensation of these conventional excitons. The above described scenario can be captured in mean-field theory calculations leading to the excitonic condensate state, as discussed earlier [1].

To proceed with our discussion, we fix $U/W = 0.5$ ($W = 8\pi$) where the excitonic condensation is present in a narrow but finite range of spin-orbit coupling, while for smaller $\lambda$’s the IC-SDW phase is present. Similar to our one-dimensional DMRG calculations, in two dimensions we found that inside the excitonic condensate regime (at a fixed weak or intermediate $U/W$ values), $r_{coh}$ decreases on increasing $\lambda$, as shown in Fig. 7(d) depicting the BCS-BEC crossover. We have calculated $r_{coh}$ for system sizes $8 \times 8$, $16 \times 16$, and $24 \times 24$. We found that in the BCS limit, $r_{coh}$ increases as the system size increases and $r_{coh}$ can reach values as high as $\approx 15.0a$ for the $24 \times 24$ lattice. This clearly supports our claim for the presence of the BCS state above the IC-SDW region, as in our DMRG chain calculations.

On the other hand, in the BEC limit, below the relativistic band insulator in the phase diagram, $r_{coh}$ is $O(0.1a)$, as shown in Fig. 7(d). Figure 7(e) displays $S(\pi, \pi)$ and $L(\pi, \pi)$, for $U/W = 0.5$, showing that only for a finite range of $\lambda$ the antiferromagnetic ordering develops. We also show the momentum distribution function of excitons $\Delta_{1/2}(q)$ at $\lambda/W = 0.53$ and $\lambda/W = 0.592$ in the BCS and BEC limits, respectively. In the BEC limit, $\Delta(q)$ is much sharper near $q = (\pi, \pi)$ than in the BCS limit, as expected because in BEC a larger ratio of excitons is expected at the condensation momentum than other momenta. To further investigate the above claim we calculated the ratio of excitons at wave vector $q = (\pi, \pi)$ and at other wave vectors using $N(\pi, \pi) = \Delta_{1/2}(\pi, \pi)/\Delta_{1/2}(q \neq (\pi, \pi))$, where $\langle \Delta_{1/2}(q) \neq (\pi, \pi) \rangle = \frac{1}{L^2} \sum_{\mathbf{q} \neq (\pi, \pi)} \Delta_{1/2}(\mathbf{q})$. It is evident from Fig. 7(e) that $N(\pi, \pi)$ increases as we transition from the BCS to the BEC limits.

V. CONCLUSIONS

In this publication, we studied the degenerate $(t_{2g})^4$ multiorbital Hubbard model in the presence of spin-orbit coupling, using one-dimensional chains and numerically exact DMRG and also using two-dimensional clusters within the Hartree-Fock approximation. In both calculations, we provide evidence for a BCS-BEC crossover in the spin-orbit excitonic condensate, in the regime of robust Hund coupling fixed as $J_H = U/4$. Within our accuracy, we established that in this model and at intermediate $U/W$, the system transits from an IC-SDW metallic phase to the BCS limit of an antiferromagnetic excitonic condensate. Further increasing $\lambda$ the coherence length of electron-hole pairs decreases rapidly as the system crossovers to the BEC regime. This BEC regime ends as eventually the system transits to the relativistic band insulator on increasing further $\lambda$. Our work provides the first indications of a BCS-BEC crossover in the excitonic magnetic state at intermediate $U/W$, a region of couplings that cannot be explored within approximations developed in the large $U/W$ regime.

We hope our results encourage further theoretical and experimental investigations on $(t_{2g})^4$ compounds with robust spin-orbit coupling. Although our study is performed using degenerate orbitals, we believe that our findings could be generic and relevant for materials showing magnetic excitonic condensation at intermediate values of the Hubbard repulsion and spin-orbit coupling.

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