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Excitonic Condensate in Flat Valence and Conduction Bands of Opposite Chirality

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Excitonic Bose-Einstein condensation (EBEC) has drawn increasing attention recently with the emergence of 2D materials. A general criterion for EBEC, as expected in an excitonic insulator (EI) state, is to have negative exciton formation energies in a semiconductor. Here, using exact diagonalization of multi-exciton Hamiltonian modelled in a diatomic Kagome lattice, we demonstrate that the negative exciton formation energies are only a prerequisite but insufficient condition for realizing an EI. By a comparative study between the cases of both a conduction and valence flat bands (FBs) versus that of a parabolic conduction band, we further show that the presence and increased FB contribution to exciton formation provide an attractive avenue to stabilize the EBEC, as confirmed by calculations and analyses of multi-exciton energies, wave functions and reduced density matrices. Our results warrant a similar many-exciton analysis for other known/new candidates of EIs, and demonstrate the FBs of opposite parity as a unique platform for studying exciton physics, paving the way to material realization of spinor BEC and spin-superfluidity.

Excitonic Bose-Einstein condensate (EBEC), first proposed in 1960s [1-4], has drawn recently increasing interest with the emergence of low-dimensional materials where electron screening is reduced leading to increased exciton binding energy (E_b) [5, 6]. In 1967, Jerome, et. al. [7], theoretically presented the possibility of an excitonic insulator (EI) phase in a semi-metal or a narrow gap semiconductor [7-10]. It was shown that the hybridization gap equation for excitonic condensate order parameter has non-trivial solutions, when E_b exceeds the semiconductor/semi-metal band gap (E_g). In deep semi-metallic regime with strong screening of Coulomb potential, this gap equation can be solved in analogy to Bardeen-Cooper-Schiffer (BCS) superconductor theory [7, 11]. On the other hand, in a semiconductor regime with low screening, preformed excitons may condense to form a BEC at low temperatures [7, 11].

This has led to significant theoretical [6, 12-19] and experimental [20, 32] investigations into finding an EI state in real materials. Especially, the EI state in a semiconductor provides an alternative route to realizing EBEC instead of targeting materials with long-lifetime excitons, such as optically inactive excitons in bulk Cu_2O [33-38] and indirect excitons in coupled quantum wells [39-40]. It is worth mentioning that excitonic condensation has been reported in double layer 2D heterostructures [41-51], where electrons and holes are separated into two layers with a tunneling barrier in between, and double-layer quantum Hall systems [52-56] have been shown to exhibit excitonic condensation at low temperature under a strong magnetic field. On the contrary, EIs are intrinsic, i.e., excitonic condensate stabilizes spontaneously at low temperature without external fields or perturbations.

However, experimental confirmation of EI state remains controversial [20, 32], mainly because candidate EI materials are very limited. On the other hand, some potential candidate EIs have been proposed by state-of-the-art computational studies [6, 12-19], based on calculation of single exciton formation energy. It is generally perceived that if single exciton E_b exceeds the semiconductor E_g, the material could be an EI candidate. But the original mean-field two-band model studied in Ref. [7] includes inter/intra band interactions, leading to a non-trivial condensation order parameter, which indicates the importance of multi-exciton interactions. Hence, in order to ultimately confirm new EI candidates, it is utmost necessary to analyze and establish the stabilization of multi-exciton condensate with quantum coherency in the parameter space of multiple bands with inter/intra band interactions, beyond just negative formation energy for single or multiple excitons.

In this Letter, we perform multi-exciton wave function analyses beyond energetics to directly assess EBEC for a truly EI state, namely a macroscopic number of excitons (bosons) condensing into the same single bosonic ground state [57-60]. Especially, we investigate possible stabilization of EBEC in a unique type of band structure consisting of a pair of valence and conduction flat bands (FBs) of opposite chirality. These so-called yin-yang FBs were first introduced in a diatomic Kagome lattice [61, 62] and have been studied in the context of metal-organic frameworks [63] and twisted bilayer graphene [64]. Recently, it was shown that such FBs, as modelled in a superatomic graphene lattice, can potentially stabilize a triplet EI state due to reduced screening of Coulomb interaction [6]. However, similar to other previous computational studies [16-19], the work was limited to illustrating the spontaneity of only a single exciton formation with a negative formation energy. Here, using exact diagonalization (ED) of a many-exciton Hamiltonian based on the yin-yang FBs, in comparison with the case of a parabolic conduction band, we demonstrate that
"$E_b > E_g$" is actually only a necessary but insufficient condition for realizing an EI state. While both systems show negative multi-exciton energies, only the former was confirmed with quantum coherency from the calculation of off-diagonal long-range order (ODLRO) of the many-exciton Hamiltonian. Furthermore, we show that with the increasing FBs contribution to exciton formation, the excitons, usually viewed as composite bosons made of electron-hole pairs, can condense like point bosons, as evidenced from the calculated perfect overlaps between the numerical ED solutions with the analytical form of ideal BEC wave functions.

A tight-binding model based on diatomic Kagome lattice is considered for the kinetic energy part of the Hamiltonian, as shown in Fig. 1(a). Our focus will be on comparing the many-excitonic ground states of superatomic graphene lattice (labelled as EI$_{SG}$), which is already known to have a negative single exciton formation energy $[6]$, and the ground states of a model system (labelled as EI$_{PB}$) with a parabolic conduction band edge, in order to reveal the role of FBs in promoting an EI state. The interatomic hopping parameters for the two systems are: $t_1 = 0.532$ eV; $t_2 = 0.0258$ eV; $t_3 = 0.0261$ eV for EI$_{SG}$, benchmarked with density-functional-theory (DFT) results $[6]$, and $t_1 = 0.62$ eV; $t_2 = 0.288$ eV; $t_3 = 0.0$ eV for EI$_{PB}$. An interesting point to note here is that for EI$_{SG}$, $t_2 < t_3$. This is an essential condition to realize yin-yang FBs in a single-orbital tight-binding model as has been discussed before, which can be satisfied in several materials $[61 - 63]$. The insets in Fig. 1(c) and 1(d) show the band structures for EI$_{SG}$ and EI$_{PB}$, respectively. Coulomb repulsion between electrons is treated using an extended Hubbard model as

$$H = H_{\text{kin}} + H_{\text{int}} = \sum_n \sum_{r,r'} t_n e_r^{\dagger} c_r^\ast + \sum_n \sum_{r,r'} \frac{V_n}{r-r'} e_r^{\dagger} c_r^\ast e_r^{\dagger} c_r, \quad (1)$$

where $t_n$ is the $n^{\text{th}}$ nearest-neighbor (NN) hopping parameter, and $V_n$ is $n^{th}$ NN Hubbard parameter. Each of the $V_n$ is calculated using the Coulomb potential, $U(r > r_n) = \varepsilon^2/(4\pi\varepsilon_r r)$, with a very low dielectric constant ($\varepsilon \sim 1.02$) due to the presence of FBs in a 2D lattice $[6]$ and a cutoff ($r_n$) for onsite interactions. The Hubbard interaction terms are projected onto all three conduction and valence bands. Spin indices in the Hamiltonian are omitted. We distinguish triplet and singlet excitonic states by the absence and presence of excitonic exchange interaction, respectively $[65 - 69]$. The Hamiltonian is exactly diagonalized for a finite system size (2 x 3) for converged results $[65]$, which includes 36 lattice sites (equivalent to a 6 x 6 trigonal lattice) with 18 electrons for a half-filled intrinsic semiconductor. With $N_{\text{eh}}$ number of electrons (holes) in conduction (valence) bands, exciton density ($n_{\text{ex}}$) is defined as $N_{\text{eh}}$ divided by the total area of finite system (i.e., $A_{\text{uc}} \times 2 \times 3$). $A_{\text{uc}}$ is the area of unit-cell which we set to be same as for superatomic graphene material with lattice constant $a = 22.14$ Å as obtained form DFT calculations $[6]$. Note that the $n_{\text{ex}}$ considered in this work is of the same order of magnitude ($n_{\text{ex}} \sim 10^{13} \text{cm}^{-2}$) as the densities at which excitonic condensate was recently observed in bilayer materials $[32 - 38]$. For the ease of readability, we also sometimes use a dimensionless $\tilde{n}_{\text{ex}} = n_{\text{ex}}/(10^{13} \text{cm}^{-2})$ in the text. Throughout this work we focus on the ground state of Eqn. 1 with varying $n_{\text{ex}}$.

We first calculate the energies and wavefunctions for a single exciton, i.e., $N_{\text{eh}} = 1$, to benchmark the single-exciton results of EI$_{SG}$ with those obtained using first-principles GW-BSE method for this lattice $[6]$. Importantly, our model calculation results, especially the trends of exciton levels, match very well with GW-BSE (Fig. 1b), S2 $[65]$. One clearly sees in Fig. 1b for EI$_{SG}$ that the formation of triplet exciton is spontaneous with a negative formation energy ($E_f$), while that of singlet is positive. These key agreements validate our model for further analysis. In Fig. 1(c) and 1(d), we plot triplet excitonic density of states for EI$_{SG}$ and EI$_{PB}$, respectively. Both systems have a negative lowest triplet $E_f$, indicative of the possibility that both systems can be a triplet EI. The insets of Fig. 1(c) and 1(d) show the band excitation contribution to the lowest triplet exciton level. For EI$_{SG}$ (Fig. 1c)), as has been shown before by GW-BSE method $[6]$, all three band excitations contribute almost equally throughout the entire Brillouin zone (BZ). In contrast, for EI$_{PB}$ (Fig. 1d)), the Γ-point excitation contributes the most due to the

![FIG. 1.](attachment:image.png)
presence of parabolic conduction band edge with band minimum at $\Gamma$. In this study, we will focus on triplet excitons, which have negative $E_f$ in both systems, so unless otherwise specified, excitons below mean triplet excitons.

Next, we discuss many-exciton calculations. A BEC superfluid flows with minimal dissipation $[58]$. Statistically, the BEC state is characterized with a Poisson particle distribution manifesting a non-interactive nature $[67]$. In other words, even in the presence of interactions, there should be a minimal change in the average formation energy ($E_f$) of a superfluid when more particles are condensed. To reveal such effect of exciton-exciton interactions, we exactly diagonalize (1) for $N_{ch} > 1$. In Fig. 2(a), and Fig. 2(b), we show the average ground-state $E_f$ of excitons with increasing $n_{ex}$ for $E_{IG}$, and $E_{IPB}$, respectively, namely the multi-exciton ground-state $E_f$ divided by $N_{ch}$. Note that both plots have the same scale to facilitate a direct comparison.

In both cases, the ground-state excitons have negative formation energies at all $n_{ex}$, but importantly the nature of exciton-exciton interactions are different. For $E_{IG}$, the excitons experience a very slight repulsive exciton-exciton interaction, indicated by a very small positive slope of their $E_f$ curve (Fig. 2(a)). From $n_{ex} = 0.39$ to $n_{ex} = 2.35$, $E_f$ increases by only $0.47\%$. Differently for $E_{IPB}$, excitons experience a strong effective repulsion from each other (Fig. 2(b)); $E_f$ increases by $21.9\%$ from $n_{ex} = 0.39$ to $n_{ex} = 2.35$. Consequently, we make the following inferences. First, the excitons in $E_{IG}$ are likely forming a BEC superfluid in the ground state because the effect of exciton-exciton interactions on $E_f$ is negligible. In the sense of weak exciton-exciton repulsion, the low-lying excitons for $E_{IG}$ appear like composite bosons, similar to weakly repulsive bosons in helium-II $[68]$. Secondly, the existence of negative exciton formation energy alone is possibly insufficient to establish a coherent BEC state. The multi-exciton ground state of $E_{IPB}$ has also negative formation energies, but judging from the strong exciton-exciton interaction excitons seem to unlikely form a condensate. In order to confirm this argument, however, one has to further assess directly the nature of exciton-exciton interaction and confirm quantum coherence of multi-exciton wavefunctions as we do next.

Since excitons are composite bosons made of electron-hole pairs like Cooper pairs of two electrons, we calculate eigenvalues of reduced two-body density matrix as a definitive signature of BEBEC based on the concept of off-diagonal long-range order (ODLR), which was first introduced to characterize superfluidity of Cooper pairs $[69, 70]$. Similarly, the reduced two-body density matrix for excitons can be written as $[69]$.\[\rho^{(2)}(k, k'; \mathbf{R}, \mathbf{R}') = \langle \Psi | \psi^\dagger_c(k) \psi_c(k') \psi^\dagger_v(k') \psi_v(k) | \Psi \rangle,\]

where $\psi^\dagger_c(k)$ creates a conduction (valence) electron at reciprocal lattice point $k$, and $| \Psi \rangle$ is the many-exciton wavefunction. We calculate the eigenvalues of $\rho^{(2)}$ and normalize it by $N_{ch}$ as a function of $n_{ex}$, then the existence of a single normalized eigenvalue close to 1 is a signature of EBEC $[69]$. We also calculate the ratio of the first two eigenvalues to check for fragmentation $[70]$ of multi-exciton ground state. Ideally, this ratio should be close to zero; if it is close to 1, it indicates fragmentation of the condensate.

In Fig. 2(c), we plot the eigenvalue spectra (for $n_{ex}$) of $\rho^{(2)}$ for the many-body ground state of excitons for $E_{IG}$ at $n_{ex} \sim 1.17$, in a descending order, i.e., $\lambda_n$ being the $n^{th}$ largest eigenvalue. Similar results are found for all $n_{ex}$ (see Fig. S4 $[64]$). Clearly, there appears a high degree of condensation for $n_{ex} \sim 1.17$. It can also be seen from Fig. 2(c), where the ratio $\lambda_2/\lambda_1$, indicative of fragmentation of the condensate, is very low for all $n_{ex}$. For comparison, in Fig. 2(d), we plot the $\lambda_n$ spectra for the many-body ground state of excitons for $E_{IPB}$ at $n_{ex} \sim 1.17$. Again, similar results are found for other $n_{ex}$ (see Fig. S5 $[69]$). The excitons in this case, however, are clearly not condensing even though they have also negative $E_f$ as shown.
acting composite-bosonic system is the presence of one large eigenvalue of $\rho^{(2)}$, as discussed above. On the other hand, for non-interacting single-body bosons (free boson gas), condensation implies macroscopic occupation of the single-particle bosonic ground state. One can form a similar non-interacting BEC wavefunction for excitons\footnote{\cite{57,58,65,67}.},

$$|\phi_{\text{BEC}}\rangle = \frac{1}{\Omega} \left| \hat{b}_{\text{ex}}^\dagger \right|^N |0\rangle,$$

where $\hat{b}_{\text{ex}}^\dagger$ is the creation operator for the single triplet level obtained from ED with $N_{eh} = 1$, $\Omega$ is the normalization constant and $N$ is the number of electrons (holes) in conduction (valence) bands. Let $|\phi_{\text{ED}}\rangle$ be the ED solution with $N$ electrons (holes) in conduction (valence) bands. Next, we calculate the overlap, $OV = |\langle \phi_{\text{BEC}} | \phi_{\text{ED}} \rangle |$ for the multi-exciton ground states (Fig. 3(b)), which can be considered as an indicator of the one-body vs composite nature of excitons. In other words, if $OV$ is close to 1, excitons behave as non-interacting single-body bosons, while if $OV$ is much smaller than 1, excitons behave as composite bosons.

Moreover, the above comparative study suggests that FB is preferable to enhance exciton coherence, as opposed to parabolic band. Interestingly, in our tight-binding model of a diatomic Kagome lattice, it is possible to increase the relative FB contribution to exciton formation by tuning the hopping parameters. Specifically, to increase the band gap between the yin yang FB\footnote{\cite{65}} to the lowest excitonic state, as exemplified in Fig. 3(a) using the hopping parameters: $t_1 = 1.92$ eV; $t_2 = 0.0$ eV; $t_3 = 0.93$ eV (labelled as $EI_{FB}$), where we plot the single excitonic energy levels and band excitation contributions (inset) to the lowest triplet level of $EI_{FB}$. Note that even with a small $E_b$ in this case, excitons have a large $E_b$ because FBs host massive carriers, leading to a very small dipole matrix element between them\footnote{\cite{91}}, which enables a low-bandgap system to still have a very low screening\footnote{\cite{91}}. The lowest exciton level of $EI_{FB}$ has a negative $E_l$ and FB excitations contribute the most to this level.

Similar to the above analyses for $EI_{SG}$ and $EI_{FB}$, we have used ODRO calculation to confirm that multi-exciton ground state of $EI_{FB}$ is an $E_l$ state\footnote{\cite{65}} with a slight fragmentation at higher $n_{ex}$ (see Fig. S6, S7\footnote{\cite{64}}). An interesting point to note here is the presence of superfluidic excitonic order in FBs, implying mobile FB excitons even though the individual electrons and holes are inherently immobile due to localization of FB wavefunctions and infinite effective mass of the carriers. Similar behavior was recently theoretically studied for FB Cooper pairs\footnote{\cite{72}}. Detailed investigation into this fascinating feature is left for future work. Here, we instead provide another compelling evidence towards this behavior. A general criterion for condensation in inter-
eters producing the desired band structure with valence and conduction FBs of opposite chirality, and hence is general. We also do a similar many-excitonic analysis for the conventional semiconductor case where both conduction and valence band edges are parabolic (Section V in SM [65]). Our results indicate that a strong exciton-exciton repulsion in this case leads to positive formation energies of many-excitonic states even though a single exciton has a negative formation energy implying an excitonic instability. Also, at low exciton density, although average exciton formation energy could still be negative, analysis of ODLRO indicates fragmentation of the condensate.

Last but not least, the yin-yang FB model and the material system of superatomic graphene studied in this work has been recently experimentally realized (albeit using a different name of triangulene-Kagome lattice), where excitonic instability was confirmed using spectroscopic measurements [74]. Moreover, flat valence and conduction bands are being increasingly realized experimentally in moiré heterostructures [75]. Similarly, bilayer FB materials could be interesting platforms to realize FB BEC by tuning the Fermi-level so that carriers in each layer occupy a FB. In addition, the stabilization of triplet EI state, as illustrated here for FBs of opposite chirality, paves the way towards material realization of exotic phases like anomalous bilayer quantum Hall states [65], fractional excited spin Hall effect [63], spin-I bosonic condensate [76] [77] and spin superfluidity [78] [79].

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