Excitonic insulator phase and dynamics of condensate in a topological one-dimensional model

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We employ mean-field approximation to investigate the interplay between the nontrivial band topology and the formation of excitonic insulator (EI) in a one-dimensional chain of atomic s – p orbitals in the presence of repulsive Hubbard interaction. We find that our model, in a non-interacting regime, admits topological and trivial insulator phases, whereas, in strong Hubbard interaction limit, the chiral symmetry is broken and the system undergoes a topological-excitonic insulator phase transition. The latter phase transition stems from an orbital pseudomagnetization and band inversion around \( k = 0 \). Our findings show that contrary to the topological insulator phase, electron-hole bound states do not form exciton condensate in the trivial band insulator phase due to lack of band inversion. Interestingly, the EI phase in low s – p hybridization limit hosts a Bardeen-Cooper-Schrieffer (BCS)/Bose-Einstein condensation (BEC) crossover. Irradiated by a pump pulse, our non-equilibrium findings reveal that the oscillations of exciton states strongly depend on the frequency of the laser pulse. We further explore the signatures of dynamics of the exciton condensate in optical measurements.

I. INTRODUCTION

The many-body problem of exciton formation driven by charge instability and Coulomb attraction between electron-hole pairs has triggered intensive previous and contemporary investigations in bulk and low dimensional semiconductors. Excitonic insulators (EI) arising from the condensation of electron-hole bound states, despite being conceptually introduced decades ago, have received new attention in recent years owing to their possible realization in the bulk of semiconductors.

Few layered transition metal dichalcogenide (TMD), are reputedly known to host collective exciton condensation due to low dimensionality and strong light-matter coupling. The prominent two-dimensional devices based on strong moire periodic potential in nearly aligned heterostructures of TMDs facilitate the observation of band dispersion flattening with the formation of strongly bound states. These devices are interesting platforms for various excitonic states studies, such as topological exciton bands and strongly correlated exciton Hubbard model. In nearly aligned WSe\(_2\)/WS\(_2\), within the A-exciton spectra region of WSe\(_2\) layer, the spatially localized interlayer excitons have been reported to respond differently to back gate doping. The reason is ascribed to the spatial distribution of exciton wave function and electron-exciton interactions based on the electron doping region. Thick encapsulation of TMDs with hexagonal Boron Nitride (hBN) layers leading to a weak coupling regime at nearly zero temperature can tune the coherent radiative decay rates of neutral excitons in these materials. Spontaneous photo-induced radiative recombination is believed to result in a photoluminescence spectra rise during the laser pump exposure, while the high-energy exciton relaxation to radiative states dominates the post-laser pulse exposure. In fact, the suppression of environmental dielectric constant through the hBN encapsulation results in radiative lifetime enhancement and suppression of nonradiative processes due to nearly zero temperature and the least existence of disorder and contamination.

Another intriguing platform to study the spontaneous exciton condensation is the promising quasi-one-dimensional (1D) chalcogenide Ta\(_2\)NiSe\(_5\). The large bandgap opening fingerprint in photoemission spectroscopy in a recent study, is believed to mark the enhancement of exciton order in the spatially separated Ni and Ta chains. Moreover, the newly reported novel low-frequency mode in Raman spectra was proposed as evidence for the existence of an EI phase in Ta\(_2\)NiSe\(_5\) emerging below \( \approx 328 \) K. Further analytical investigations suggested that the phase transition is associated with a Bose-Einstein condensation (BEC) in the scheme of a one-dimensional (1D) extended Falicov-Kimball model (EFKM) with an overlapping band semimetal as the normal state. Meanwhile, another plausible scenario is argued to be a spontaneous Ta-Ni hybridization based on charge instability which breaks the symmetry. Besides, the importance of structural phase transition as origin of electronic gap has been addressed in recent pump-probe and Raman spectroscopy measurements.

To date, the theorized models to understand the EI phase are mostly based on the idea of strong correlations in a semimetal with a small band overlap or a semiconductor with a small gap. Given the importance of designing new models, here, we explore the EI phase in a 1D chain of atomic s – p orbitals with the inclusion of odd parity hybridization (See Fig. 1(a)). In the non-interacting scheme, this model exhibits topological insulator (TI) and trivial band insulator (BI) phases. Adding the Hubbard interaction, our main objective is to answer the following questions. How do the latter phases change with correlations? Do TI and EI phases compete or coexist? How does the non-equilibrium dynamics of exciton condensate, coupled to a phonon bath, respond to ultrafast pulses, and what are the experimental consequences in optical measurements?

In this article, we show that the EI phase emerges out of the topological insulator phase beyond a critical interaction, while the BI phase remains remote in forming exciton condensate. Furthermore, our non-equilibrium analysis reveals that the oscillation of exciton condensate strongly depends on the frequency of driving pulse with signatures visible in optical conductivity. The paper is organized as follows. In Section II we present the theoretical model for a 1D s – p chain in equilibrium and address the nature of spontaneous exciton condensation in a topological insulator phase. Particularly, we show that the exciton formation originates from the band
inversion. Next, we address the exciton dynamics in a stimulated pump-probe situation in section III. In section IV, we present the results for the optical spectra of a $s-p$ chain in a linear response regime. Lastly, our findings are summarized in section V.

II. MODEL AND EQUILIBRIUM PHASE DIAGRAM

In this section, we introduce the 1D $s-p$ chain and present a comprehensive analysis of its equilibrium phase diagram.

A. Interacting s-p model

The non-interacting spinless model of a 1D chain of atoms with $s$ and $p$ orbitals and lattice spacing $a$, as shown in Fig. 1(a), reads

$$
H_0 = \epsilon_s \sum_j c_j^\dagger c_j + \epsilon_p \sum_j p_j^\dagger p_j - \sum_j t_s(c_j^\dagger c_{j+1} + c_{j+1}^\dagger c_j) $$

$$
+ \sum_j t_p(p_j^\dagger p_{j+1} + p_{j+1}^\dagger p_j) + V_{sp} \sum_j (c_j^\dagger p_{j+1} - c_{j+1}^\dagger p_j)$$

$$
- V_{sp} \sum_j (p_j^\dagger c_{j+1} - p_{j+1}^\dagger c_j)
$$

(1)

where $c_j^\dagger(c_j)$ and $p_j^\dagger(p_j)$ are the charge creation and annihilation operators in $s$ and $p$ orbitals of $j$th atomic site, respectively. $\epsilon_s(\epsilon_p)$ is the on-site energy, and $t_s(t_p)$ is the hopping parameter between nearest neighbors with same orbitals. Also, $V_{sp}(V_{ps})$ is the hybridization energy between $s(p)$ and $p(s)$ orbitals in a neighboring site with an odd parity, i.e. $V_{sp}(-x) = -V_{sp}(x)$. This odd parity is responsible for the band inversion that leads to TI phase formation. By Fourier transformation to momentum space, the Hamiltonian (1) becomes

$$
H_0 = \sum_k (\epsilon_s - 2t_s \cos ka) c_k^\dagger c_k + \sum_k (\epsilon_p + 2t_p \cos ka) p_k^\dagger p_k$$

$$
+ 2iV_{sp} \sum_k \sin ka c_k^\dagger p_k - 2iV_{ps} \sum_k \sin ka p_k^\dagger c_k.
$$

(2)

In a more generic model where repulsive short-range Coulomb interactions are present, the interacting Hamiltonian is $H_e = H_0 + H_U$, where

$$
H_U = U_{sp} \sum_j [(n_s - 1/2)p_j^\dagger p_j + (n_p - 1/2)c_j^\dagger c_j - \phi p_j^\dagger c_j - \phi^* c_j^\dagger p_j],
$$

(3)

where, $\phi = \langle c_j^\dagger p_j \rangle$ is the exciton order parameter. Moreover, $n_s = \langle c_j^\dagger c_j \rangle$ and $n_p = \langle p_j^\dagger p_j \rangle$ are the charge density order parameters of $s$ and $p$ orbitals, respectively. In momentum space the two-band mean-field Hamiltonian is cast as a pseudospin in a pseudomagnetic field:

$$
H_e^{MF} = \sum_{k,\gamma} B_k^\gamma S_k^\gamma
$$

(4)

where $S_k^\gamma$ is the pseudospin component,

$$
S_k^\gamma = \frac{1}{2} \xi_k^\gamma \sigma_\gamma \xi_k, \quad \xi_k^\gamma = (c_k^\dagger, p_k^\dagger)
$$

(5)

with $\sigma_\gamma(\gamma = 1-3)$ being the Pauli matrices, and $\sigma_0$, the identity matrix. In what follows, we set the parameters as $\epsilon_s = -\epsilon_p = \epsilon$, $t_s = t_p = t$ and $V_{sp} = V_{ps}$. Also, for sake of simplicity, we set $a = 1$, and $t = 1$, hereafter, unless otherwise is stated. With these identifications, the components of the pseudomagnetic field, $B_k^\gamma$s, are
\[ B_k^0 = U_{sp}(n_s + n_p - 1) \]  
\[ B_k^1 = -2U_{sp}\Re \phi \]  
\[ B_k^2 = -2U_{sp}\Im \phi - 4V_{sp}\sin k \]  
\[ B_k^3 = 2\epsilon - 4r\cos k + U_{sp}(n_p - n_s). \]  

Now, we adopt a self-consistent calculation to address the energy dispersion and the order parameters in equilibrium state at zero temperature. The procedure is as follows. First, we solve the eigenvalue problem \( (H_{k}^{MF} - E_{k,\pm})\ket{k,\pm} = 0 \), for the Bloch Hamiltonian driven from Eq. (5), to find the eigenvalues, \( E_{k,\pm} = [B_k^{1\pm} \pm |B_k|]/2 \), and their corresponding eigenfunctions, \( |k,\pm\rangle \). Next, we use this knowledge to evaluate the expectation values of pseudospin components at equilibrium using the following relation

\[
\langle S_k^{\gamma}(0) \rangle = \begin{cases} 
\frac{B_k^{1\gamma}(0)}{2[B_k(0)]} [f(E_{k,+}(0)) - f(E_{k,-}(0))] & (\gamma = 1 - 3) \\
\frac{1}{2} [f(E_{k,+}(0)) + f(E_{k,-}(0))] & (\gamma = 0),
\end{cases}
\]

where \( f(E_{k,+}(0)) \) is the Fermi-Dirac distribution function at equilibrium. We start with an initial guess for \( \langle S_k^{\gamma}(0) \rangle \) over all momentum \( k \). We then obtain the mean-field order parameters via

\[
\begin{bmatrix} n_0 \\ \phi \\ n_1 \end{bmatrix} = \frac{1}{N} \sum_k \left[ (S_k) \cdot \sigma + \langle S_k^0 \rangle \sigma_0 \right].
\]

Eventually, the latter values are fed into \( H_k^{MF} \) to find \( \langle S_k^{\gamma}(0) \rangle \) through Eq. (7). Until reaching the convergence, this process is iterated with the following assumption that the system is at half filling, i.e., \( n_s + n_p = 1 \).

Before we proceed any further, it is worthwhile to note a few points about the symmetry in the EI phase. In the absence of \( s-p \) orbitals hybridization, \( V_{sp} \), in Hamiltonian (1), both valence and conduction bands enjoy \( U(1) \) charge conservation symmetry separately. This implies that the overall symmetry is \( U_s(1) \times U_p(1) \) for \( s \) and \( p \) bands, which is spontaneously broken in EI phase due to condensation of complex exciton order parameter, \( \phi = |\phi|e^{i\phi} \). Here, \( \phi \) is the phase of the condensate whose fluctuations give rise to the phase mode. The net symmetry in our model, however, is explicitly broken down to \( \hat{U}(1) \) symmetry of total charge conservation by a nonzero \( V_{sp} \) (and also by coupling to phonons which we will discuss in Sec. III A). We further find that the order parameter \( \phi \) becomes real, \( \Im \phi = 0 \), at equilibrium.

### B. Equilibrium phase diagram

The equilibrium phase diagram in the plane of Hubbard \( U_{sp} \) and on-site energy for a fixed value of inter-orbital hybridization \( V_{sp} = 1/2 \) is shown in Fig. 1(b). In the non-interacting limit, i.e., \( U_{sp} = 0 \), a TI phase sets in for \( \epsilon < 2 \), while the trivial BI phase appears for \( \epsilon > 2 \), consistent with previous studies. In the TI phase, the bands undergo an inversion around \( k = 0 \) and thus the winding number becomes \( \nu = -1 \). In the BI phase, on the other hand, the valence and conduction bands are mostly of \( p \) and \( s \) character, respectively, yielding a zero winding number \( \nu = 0 \). The definition of winding number and details of calculations can be found in Appendix A.

As can be seen in Fig. 1(b), the TI phase, interestingly, shrinks as the short-range interactions become stronger, until eventually, the EI phase emerges at \( U_{sp} \approx 3 \) for nearly zero on-site energies. Therefore, in strongly correlated systems, the EI phase surpasses the TI phase and we only have exciton and band insulator states. In an intermediate coupling strength, i.e., \( 2 < U_{sp} < 3 \), all three topological phases can be reached by varying on-site potential \( \epsilon \). In both TI and BI phases, where the exciton order parameter vanishes, \( \phi = 0 \), the mean-field Bloch Hamiltonian has a chiral symmetry. This can be seen from Eq. (4) with the following Bloch Hamiltonian

\[
H_k^{MF} = \mathbf{d}(k) \cdot \mathbf{\sigma},
\]

where \( \mathbf{d}(k) = 1/2(B_k^{1x}, B_k^{1y}, B_k^{1z}) \), and also, \( B_k^1 = 0 \) when \( \phi = 0 \). Hence, since \( \langle H_k^{MF}, \sigma_x \rangle = 0 \), the system is manifestly chiral symmetric and the winding of unit vector \( \mathbf{d}(k) \equiv \mathbf{d}(k)/|| \mathbf{d}(k) || \) around the origin in the \( y-z \) plane determines \( \nu \) as one crosses the one-dimensional Brillouin zone. Therefore, we obtain \( \nu = -1 \) for TI phase and \( \nu = 0 \) for BI phase.

In Fig. 2(a,b) the impact of the \( s-p \) orbitals hybridization on the phase diagram is shown. The plots in panel (a) and panel (b) depict the topological behavior of \( s-p \) chain for two specific values of \( U_{sp} = 3 \) and 6, respectively. As can be seen in both panels, the EI state is limited to the low hybridization region and enhances towards larger hybridization as the short-range interactions become stronger. Thus, larger the Hubbard interaction is, a larger value of hybridization is required for the TI phase to set in.

The physical picture obtained thus far is that the EI phase emerges out of the TI phase in the strong interaction regime. Therefore, the two phases compete with each other. Moreover, we find that the EI phase is topologically trivial, and the nonzero exciton parameter leads to \( B_k^1 \neq 0 \) and, consequently, breaks the chiral symmetry. This implies that the unit vector \( \mathbf{d}(k) \) tips out of the \( y-z \) plane, and thus the closed path traveled by \( \mathbf{d}(k) \) can be shrunk to zero continuously. From a mathematical viewpoint, in the EI phase, the latter unit vector belongs to the surface of a sphere \( S^2 \), and since the first homotopy group of \( S^2 \) is trivial \( \pi_1(S^2) = 0 \), the phase is trivial.

### C. Phase transition to EI phase

In order to understand the phase transition to EI, we present an analytical study of order parameters. To this end, we build the mean-field Hamiltonian based on pseudospin components, followed by calculation of the exciton parameter, so that we
state, i.e. Eq. (8), we obtain the exciton parameter at the half filling find the criteria at which the EI phase could emerge. From Eq. (8), we obtain the exciton parameter at the half filling state, i.e. $f(E_{k,+}(0)) - f(E_{k,-}(0)) = -1$, as

$$
\phi = \frac{1}{N} \sum_k \left( \langle S_k^z \rangle + i \langle S_k^x \rangle \right) \\
= \frac{1}{N} \sum_k \left( U_{sp} \phi + 2iV_{sp} \sin k \right) / | \mathbf{B}_k |.
$$

To further simplify the above equation, we expand the norm of the pseudomagnetic field vector,

$$
[\mathbf{B}_k] = \sqrt{4U_{sp}^2 \phi^2 + 16V_{sp}^2 \sin^2 k + \left( 2\epsilon - 4t \cos k - 2U_{sp} S_3 \right)^2}
$$

which in fact, involves the evaluation of $S_3 = 1/N \sum_k \langle S_k^3 \rangle$,

$$
S_3 = \frac{1}{2\pi} \int_{-\pi}^{\pi} dk \frac{-2\epsilon + 4t \cos k + 2U_{sp} S_3}{| \mathbf{B}_k |},
$$

that practically plays the role of a pseudomagnetic field parameter. Since $| \mathbf{B}_k |$ is an even function of $k$, Eq. (10) simplifies to

$$
\phi = \frac{1}{N} \sum_k \frac{U_{sp} \phi}{| \mathbf{B}_k |},
$$

which is not dissimilar to the Bardeen-Cooper-Schrieffer (BCS) superconductor gap equation. Eventually, we have two equations for $\phi$ and $S_3$ that could be solved self-consistently to obtain a concrete condition over which the exciton parameter becomes nonzero. The results are displayed in Fig. 2(c,d) for the on-site energy $\epsilon = 1/2$, and inter-orbital hopping parameter $V_{sp} = 1/2$ (panel (c)), and $V_{sp} = 1$ (panel (d)). As can be seen, a slope discontinuity occurs in $z$ component of pseudospin, $S_3$, which presents the difference between $s$ and $p$ orbitals charge density $(n_s - n_p)$, acting as an effective orbital pseudomagnetization. The susceptibility (i.e. the derivative of the pseudomagnetization) at a critical pseudomagnetic field becomes discontinuous at a critical value of interaction alluding a phase transition to an ordered phase, i.e EI. The nonzero $\phi$ amounts to developing an $x$-component of pseudomagnetization. This is exactly the tipping of $\mathbf{d}(k)$ out of the $y-z$ plane as we mentioned in the preceding subsection. Note that the creation of $x$-component is set by hybridization of $s-p$ orbitals which is facilitated in the band-inverted TI phase. In the BI phase, however, no band mixing occurs which is detrimental in forming nonzero $\phi$ even for strong interactions.

D. BCS-BEC crossover in the EI phase

As we showed above, the strong Hubbard interaction can establish the EI phase in the $s-p$ model. Our results further reveal that there is a BCS-BEC crossover within the EI phase by varying interaction and hybridization strength. In the BSC regime, the minima of the bandgap appear away from $k = 0$, while in the BEC regime bottom of the valence band and top of the conduction band lie at $k = 0$. In Fig. 3 we present a phase diagram in $U_{sp}$-$V_{sp}$ plane where the domain of BCS and BEC phases is indicated. BCS phase can be only found in a low $s-p$ hybridization energy limit and by increasing the short-range interaction strength, the bands are flattened until a BCS-BEC crossover occurs. In Appendix B we present...
the details of band evolution, which clearly shows how the crossover takes place.

The BCS-BEC crossover has been addressed very recently in a three-orbital Hubbard model with the inclusion of spin-orbit coupling (SOC)\(^{38}\). It’s been shown that the SOC-originated condensation of BCS type EI at intermediate Hubbard interaction region crosses over to a BEC type EI by increasing SOC strength. This suggests that SOC in their model acts analogously to the hybridization of the \(s\) and \(p\) orbital in our model. Furthermore, in a very recent study, the Ta\(_2\)NiSe\(_5\) phase transition that was observed as an anomaly in the resistivity at \(T_c = 328\)K\(^{1}\), has been modeled via 1D EFKM where a BEC type condensation occurs even though the normal state is an overlapping band semimetal\(^{27}\). This in fact alludes to the notion of BCS-BEC crossover.

III. EXCITONS IN NON-EQUILIBRIUM STATE

Motivated by recent pump-probe measurements on flakes of Ta\(_2\)NiSe\(_5\)\(^{28}\), in this section we study the non-equilibrium dynamics of the EI phase of the \(s-p\) model described in the preceding section. The non-equilibrium dynamics of excitons in a 1D \(s-p\) chain generated with a laser pump provides an excellent playground for understanding the collective behaviors. We first present the details of the studied non-equilibrium model, then we elaborate on the real-time evolution of exciton condensate. Note that, for the sake of simplicity, we neglect any plausible induction of dipole excitation.

![Diagram](https://via.placeholder.com/150)

FIG. 3. The exciton order of a 1D \(s-p\) chain at equilibrium with on-site energy of \(\epsilon = 1/2\). The BCS-BEC crossover is visible at low values of \(V_{sp}\). The details of band dispersion change with Coulomb coupling enhancement is shown in Fig. 7.

A. Coupling to phonons and laser pulses

We consider the charge interactions with a bath of optical phonons with \(\hbar \omega_{ph}\) energy. The Hamiltonian is modified as \(H = H_e + H_{ph}\), where

\[
H_{ph} = \hbar \omega_{ph} \sum_j b_j^\dagger b_j + g \sum_j (b_j^\dagger b_j) (c_j^\dagger p_j + p_j^\dagger c_j), \tag{14}
\]

\(g\) is the electron-phonon coupling, and \(b_j^\dagger (b_j)\) is the creation (annihilation) operator for phonons. The charge-phonon interaction in mean-field approximation reduces to

\[
H_{ph}^{MF} = \hbar \omega_{ph} \sum_j b_j^\dagger b_j + gX \sum_j (c_j^\dagger p_j + p_j^\dagger c_j) + g \sum_j (b_j^\dagger + b_j)(\phi + \phi^*), \tag{15}
\]

where \(X = \langle b_j^\dagger + b_j \rangle\) is the phonon displacement. The total Hamiltonian in momentum space thus can be written as the summation of Fourier transform of electron-phonon mean-field Hamiltonian and Eq. (4),

\[
H^{MF} = \tilde{H}_e^{MF} + \hbar \omega_{ph} \sum_j b_j^\dagger b_j + 2g\text{Re} \phi \sum_j (b_j^\dagger + b_j) \tag{16}
\]

with \(\tilde{H}_e^{MF}\) being the electronic part of the mean-field Hamiltonian with a slightly modified pseudomagnetic field component \(B_j^e = -2U_{sp} \text{Re} \phi + 2gX\).

Next, we model an optical laser pulse pulse impinging on the system. We assume the induced time-dependent electromagnetic vector potential as a Gaussian function

\[
A(t) = \Theta(t) A_0 e^{-\frac{(t - t_p)^2}{\sigma_p^2}} \sin \Omega t / \hbar. \tag{17}
\]

in which, \(t_p = 100\)ps and \(\sigma_p = 30\)ps are the duration and width of the pulse, respectively. Here, we set the pulse amplitude \((A_0)\) to 0.05 and \(\hbar = 1\) throughout this paper. The non-equilibrium state could be modeled by a Peierls phase in the mean-field Hamiltonian\(^{15}\). The Heisenberg equation of motion provides the time evolution of electron-hole pairs,

\[
\frac{\partial \langle S_k(t) \rangle}{\partial t} = B_k(t) \times \langle S_k(t) \rangle \tag{18a}
\]

\[
\frac{\partial \langle S_k^2(t) \rangle}{\partial t} = 0 \tag{18b}
\]

\[
\frac{\partial X(t)}{\partial t} = \omega_{ph} P(t) \tag{18c}
\]

\[
\frac{\partial P(t)}{\partial t} = -\omega_{ph} X(t) - 4g\text{Re} \phi. \tag{18d}
\]

Here, \(P(t) = i \langle b_j^\dagger b_j \rangle\) is phonon momentum. We solve the set of above equations, (18a)-(18d), numerically by Runge-Kutta fourth-order method where we insert the self-consistent results as the initial value for exciton order parameter. We assume that the system is at zero temperature and thus the initial phonon momentum vanishes \(P(0) = 0\). From Eq. (18d),
FIG. 4. Time evolution of the exciton parameter for diverse $s-p$ chain systems. (a,b) illustrates the results for a $s-p$ chain with $\epsilon = 1/2, V_{sp} = 1/2, U_{sp} = 2.8$, while panel (c,d) shows those for $\epsilon = 1/2, V_{sp} = 1, U_{sp} = 3.8$. (e,f) are exciton dynamics for $\epsilon = 1/4, V_{sp} = 1/2, U_{sp} = 4$. Black dashed line indicates the exciton order time evolution for laser frequency $\Omega = E_g$. For both EI phases, (a,b,e,f) BCS and (c,d) BEC, the exciton order melts down during the exposure of laser pump and oscillates afterward. In panel (e,f) the exciton dynamics in $t \in [300, 600]$ps and $\text{Re}\phi(t) \in [0.26, 0.28]$ window are magnified to ease the visualization. The left (right) column insets are the corresponding band dispersion (oscillation frequency). The dashed (solid) lines depict energy dispersion for non-interacting $U_{sp} = 0$ (interacting $U_{sp} \neq 0$) $s-p$ chain in the inset. Phonon-charge interactions addressed in III A, are considered in preparation of this figure with the following assumptions. $g = \sqrt{\lambda \omega_{ph}/2}$ is the charge-phonon interaction factor. $\lambda = 0.1$ is the effective electron-phonon coupling parameter, and $\omega_{ph} = 0.1$ is the optical phonon energy.

This assumption yields $X(0) = -4g\phi(0)/\omega_{ph}$. At equilibrium ($t \leq 0$) the pseudomagnetic field component discussed below...
Eq. (16), becomes \( B^1 = -2(U_{sp} + 2\lambda)\text{Re}\phi \) with \( \lambda \equiv 2q^2/\omega_{ph} \) being the electron-phonon coupling constant. Hence, at equilibrium the latter interaction only shifts \( U_{sp} \). In the following we discuss the evolution of system for \( t > 0 \) irradiated by the pump pulse (17).

### B. Real-time evolution of EI condensate

The time evolution of the exciton order is illustrated in Fig. 4. Panels (a) and (b) show a BCS type EI dynamics, whilst panel (c) and (d) depict those for a BEC EI. All EI phases demonstrate an exciton order melt-down after the laser pump exposure as a consequence of photo-induced breaking of exciton bound states. In each panel, we show the real-time evolution of \( \text{Re}\phi \) for different values of pulse frequency \( \Omega \).

The black dashed line shows \( \text{Re}\phi(t) \) when \( \Omega \) equals the EI gap energy. From panel (b) we see that, when the pulse frequency is lower than the gap, the oscillation of condensate is almost the same for all. However, for \( \Omega \) being larger than the gap, the oscillations do depend on pulse frequency. This behavior can be ascribed to the type of band structure associated with the BCS condensate illustrated in the inset of panel (a). The variation of band structure near \( k = 0 \) introduces different resonance energy scales for the condensate. For frequencies well above the gap, these variations are smeared out; the same reasoning holds for frequencies below the gap as shown in panel (b).

The oscillation frequency \( \tilde{\omega} \) of \( \text{Re}\phi(t) \) with respect to pulse frequency is depicted in the inset of panel (b). Note that in all latter cases, \( \tilde{\omega} \) of exciton condensate coincides with the phonon frequency, i.e., with the oscillation of \( \chi(t) \). We note that in the absence of electron-phonon coupling the atoms oscillate with their natural optical frequency which we set to be \( \omega_{ph} = 0.1 \) throughout. However, when coupled to exciton condensate the phonon frequencies change. The coupling between phonons and excitons has been argued to be crucial in understanding the recent pump-probe measurements on Ta\(_2\)NiSe\(_5\)\(^{23,26,36}\).

The results of the condensate dynamics for BEC type EI are shown in Fig. 4(c,d). The band structure depicted in panel (c) clearly shows that the direct gap is located at \( k = 0 \). Again in this case the exciton order parameter is quenched by the pump pulse and oscillates afterward. The main observation now is that for pulse frequencies either below or above the bandgap, the exciton frequencies \( \tilde{\omega} \) changes only mildly in contrast to the BCS case discussed above. The reason can be ascribed to the band structure of the BEC with only one direct bandgap at \( k = 0 \).

Finally, in Fig. 4(e,f), we present the results for non-equilibrium dynamics of condensate in an EI with large bandgap, \( \approx 3.41 \). Two observations are manifest. First, we see that the condensate is less influenced by the laser pulse than the other aforementioned cases. That is, \( \text{Re}\phi \) drops to a small fraction of its initial value; it changes from \( \text{Re}\phi \approx 0.34 \) to \( \text{Re}\phi \approx 0.26 \) even at the resonance with the gap energy. Second, in stark contrast to previous cases, no oscillation occurs for a full range of pulse frequencies below and above the EI gap. Compared to dynamics in panels (a-d), one can see that the effective coupling of condensate to phonons only takes place in the EI phase with a narrow gap.

Before moving to the next section, a remark is in order. The condensate dynamics saturates to a mean steady value after a melt-down and does not decay, which can be attributed to the lack of damping mechanism induced by phonons, contamination, doping, and defects in general. This is actually the motivation behind the recent time-resolved photoluminescence measurements reporting a high exciton lifetime in encapsulated TMDs\(^{30}\). In our model the decay could be considered by adding a damping term as \( -\gamma P(t) \) phenomenologically\(^{32}\) to the right side of Eq. (18d), which leads to decay of oscillations.

### IV. OPTICAL CONDUCTIVITY

This section aims to answer the last question posed in introduction seeking the signature of EI phases in optical measurements. In a linear response regime at zero temperature, the longitudinal optical conductivity can be evaluated by Kubo formula,
FIG. 6. Optical response of (a-c) BCS, (d-f) BEC EI systems introduced in Fig. 4, for finite time after the laser pulse exposure. Each panel presents the optical conductivity for specific laser pump frequency ($\omega$). Black bottom line in each panel is the optical spectra for the corresponding EI at equilibrium. The energy dispersion at times shown in panels, are plotted for comparison (See top right of each panel). The dashed line indicates the energy dispersion at equilibrium.

\[
\sigma_{xx} = i \frac{N}{\omega} \sum_k \left( \frac{1}{\omega - \omega_k + i0^+} - \frac{1}{\omega + \omega_k + i0^+} \right) |J|^2, \tag{19}
\]

where $\omega_k = E_{k\uparrow} - E_{k\downarrow}$, $\omega$ is the laser probe frequency, and $J$ is the current matrix element between the conduction and valence bands,

\[
J = -2e \left( t \sin(k) \frac{B_1^1 + iB_2^1}{|B_k|} + V_{sp} \cos(k) \frac{B_3^1}{|B_k|} \right). \tag{20}
\]

From Eq. (20) we see that, the matrix element is a function of exciton parameter ($B_1^1 + iB_2^1$) and the $s - p$ orbitals hybridization parameter ($V_{sp}$). Also, via setting $V_{sp} = 0$, one can reproduce the relation for current density operator of a 1D EFKM\textsuperscript{35},

\[
J = -2e \sin(k) \frac{B_1^1 + iB_2^1}{|B_k|}. \tag{21}
\]

One observation is that when both $\phi$ and $V_{sp}$ are zero, optical conductivity vanishes, since $J = 0$. However, when the exciton condensation is formed, the matrix element $J$ becomes nonzero and optical response acquire finite values as a function of measured frequencies.

The real part of the $\sigma_{xx}$ is shown in Fig. 5 for diverse values of Hamiltonian parameters at equilibrium. Panel (a) and (b), present the longitudinal optical spectra, for BEC and BCS EIs, respectively. Note that both plots demonstrate condensations in the TI phase. From the optical spectra, one can clearly see that more than one excitonic bound state form, including at exact bandgap energy transitions ($E_{\alpha}$), and the BZ edge transitions ($E_{\beta}$ in (c) and (d)). Thus, the exciton order, at equilibrium state, is a superposition of all bound states. There also exists an additional peak in the optical absorption of BCS type EI which stems mostly from the zero momentum transitions ($E_{\beta}$). Overall, the exciton condensation fingerprints in optical response are originated from optical transitions at almost flat bands in the electronic dispersion, where the joint density of states becomes extremely large. Therefore, the optical conductivity measurements provide a valuable tool for EI type
recognition in a topological excitonic insulator phase.

We now utilize a similar procedure to evaluate the optical absorption spectra in a photo-induced excited regime (non-equilibrium state after the imposition of a laser pump). Fig. 6 represents the real part of longitudinal optical conductivity for (a-c) BCS and (d-f) BEC EI previously discussed in Fig. 5. Each column depicts the optical response to a specific laser pump energy ($\Omega$). The bottom plot marked in black in each panel depicts the equilibrium optical response for the corresponding system.

As previously discussed, three peaks appear in the equilibrium optical response of a BCS EI stemming from the optical transitions in the BZ center, edges, and the midway between them (black bottom line in panel (a-c)). At a finite time far away from the laser pump exposure time ($t_p$), we see that the BZ edge transitions with the least amplitude compared to its counterparts remain almost intact indicating that the energy bands are not significantly altered at BZ edges. On the contrary, we observe an energy difference displacement between the central BZ transitions and a slight red-shift of these two peaks (See Fig. 6(c)). This indicates the slight variation of energy dispersion and consequently the joint density of states (JDOS) with the laser pump exposure. Interestingly, an additional peak emerges in the leftmost of the optical response ($\Delta$JDOS) with the laser pump exposure. Interestingly, an additional peak emerges in the leftmost of the optical response which marks the splitting of the first peak into two strongly bound condensates by lifting the degeneracy of otherwise local band minima. This is clearly seen in the band dispersion shown in the top right of each panel: the energy dispersion (solid lines) becomes asymmetric compared to one at equilibrium (dashed line). Note the time scale over which the exciton order parameter irradiated by a pump pulse and coupled to an optical phonon bath. We find that exciton dynamics clearly depend on the driven laser frequency, and of course on the nature of EI phases, BCS vs BEC condensates. Also, the fingerprint of photo-induced bandgap shrinkage is observable as an energy redshift in the optical spectra of both EI types. More interestingly, the emergence of an additional peak in the optical spectra of BCS EI is understood to be a peak splitting driven by degeneracy lifting of optical transitions.

VI. ACKNOWLEDGMENTS

The authors would like to acknowledge support from the Sharif University of Technology under Grant No. G960208 and Iran’s National Elite Foundation.

Appendix A: Winding number

In this appendix we present the details of computation of winding number for non-interacting Hamiltonian (2). The Bloch Hamiltonian of Eq. (2), transforms under the following unitary transformation,

$$U = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 1 \\ 1 & -1 \end{bmatrix}, \quad (A1)$$

to,

$$H_0(k) = \frac{\epsilon_s + \epsilon_p}{2} - t_s \cos k + t_p \cos k \mathbb{I}_{2 \times 2} + \mathbf{d}(k) \cdot \mathbf{\sigma}, \quad (A2)$$

where

$$d_s(k) = \frac{\epsilon_s - \epsilon_p}{2} - t_s \cos k - t_p \cos k, \quad d_p(k) = 2V_{sp} \sin k, \quad \mathbf{d}(k) = 0. \quad (A3)$$

For $\epsilon_s = -\epsilon_p = \epsilon > 0$, $t_s = t_p = t = 1$, the $H_0(k)$ is chiral symmetric, since $[H_0(k), \sigma^z] = 0$. Thus, the winding number can be written as

$$v = \frac{1}{2\pi} \int_{-\pi}^{\pi} dk \frac{d}{dk} \ln \mathbb{d}(k) = \frac{\bar{V}}{2\pi} \int_{-\pi}^{\pi} dk \left( \frac{\bar{\epsilon} \cos k - 1}{(\bar{\epsilon} - \cos k)^2 + \bar{V}^2 \sin^2 k} \right), \quad (A5)$$

where we have used,

$$\mathbb{d}(k) = d_s(k) + id_p(k) = |\mathbb{d}(k)| e^{i\phi}, \quad (A6a)$$
FIG. 7. The evolution of band dispersion for a 1D \( s - p \) chain with increment of \( U_{sp} \) from 3 to 7. Top row plots are for a \( s - p \) chain with \( \epsilon = 1/2, V_{sp} = 1/2 \), and the bottom row plots for one with \( \epsilon = 1/2, V_{sp} = 1 \). Here, we clearly see a BCS-BEC crossover with enhancement of Hubbard \( U_{sp} \).

\[
|\mathbf{d}(k)| = \sqrt{(\epsilon - 2t \cos k)^2 + 4V_{sp}^2 \sin^2 k}, \quad (A6b)
\]

\[
\varphi_k = \tan^{-1}(d_y(k)/d_x(k)), \quad (A6c)
\]

\[
\bar{\epsilon} = \epsilon/2t, \quad (A6d)
\]

\[
\bar{V} = V_{sp}/t. \quad (A6e)
\]

Therefore, for a non-interacting \( s - p \) chain (\( U_{sp} = 0 \)), when \( \epsilon > 2 \Rightarrow d_x(k) > 0 \), and the winding number becomes zero. In fact, in a high on-site energy regime, \( s \) and \( p \) orbitals are energetically separated leading to an effectively insignificant hybridization between \( s \) and \( p \) orbitals, thus a semiconductor forms. On the other hand, in a small \( \epsilon \) regime, the \( s - p \) hybridization significantly contributes to band inversion and leads to a TI phase forming.

Appendix B: BCS-BEC crossover

In the low \( s - p \) hybridization energy limit, the band structure of a BCS type EI evolves as the Hubbard interaction is enhanced. The valence and conduction bands are flattened at \( k = 0 \) with \( U_{sp} \) increment until eventually, a BCS-BEC crossover occurs. Fig. 7 illustrates the gradual evolution of band dispersion proportional to \( U_{sp} \) strength.
