Interlayer excitons with tunable dispersion relation

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Interlayer excitons, comprising an electron in one material bound by Coulomb attraction to a hole in an adjacent material, are composite bosons that can assume a variety of many-body phases. The phase diagram of the bosonic system is largely determined by the dispersion relation of the bosons, which itself arises as a combination of the dispersion relations of the electron and hole separately. Here I show that in situations where either the electron or the hole has a nonmonotonic, “Mexican hat-shaped” dispersion relation, the exciton dispersion relation can have a range of qualitatively different forms, each corresponding to a different many-body phase at low temperature. This diversity suggests a platform for continuously tuning between different quantum phases using an external field.

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I. INTRODUCTION

When an electron binds to a hole in a solid state system, the resulting exciton has properties that are qualitatively different from either the electron or hole separately. For example, the electron and hole have fermionic statistics, while the exciton is a boson. Electrons and holes interact via a long-ranged Coulomb interaction, while excitons have only a short-ranged dipolar interaction. But in terms of its dispersion relation, an exciton is usually qualitatively similar to a free electron or hole. In particular, one can usually describe the center-of-mass coordinate of the exciton as an effectively free particle having mass equal to the sum of the electron and hole masses, so that the total energy $\varepsilon$ of the exciton grows as $P^2$, where $P$ is the center-of-mass momentum.

But what happens when the electron and hole have dispersion relations that are qualitatively different from each other? Which of its constituent particles does the exciton take after, in terms of its dispersion relation: the electron or the hole?

In particular, consider the case of a two-dimensional (2D) exciton for which the electron has a “Mexican hat-shaped” dispersion,

$$\varepsilon_e(k) = \frac{(|k| - k_0)^2}{2m_e}, \quad (1)$$

while the hole has the usual parabolic dispersion

$$\varepsilon_h(k) = \frac{|k|^2}{2m_h}. \quad (2)$$

Here, $m_e$ and $m_h$ are the effective masses of the electron and hole, respectively, $k$ is the momentum, and $k_0$ is a characteristic momentum that defines the width of the “brim” of the Mexican hat (see Fig. 1). One can now ask the question: What is the form adopted by the exciton dispersion relation $\varepsilon(P)$?

This seemingly quaint problem actually has a range of experimental implications. As explained below, its solution suggests the ability to engineer interlayer excitons with widely tunable dispersion relation [1]. This tunability offers the potential to realize a range of many-body quantum phases within a single device by changing an external field.

The idea of using interlayer excitons to realize a 2D Bose system goes back more than 40 years [2], but its experimental realization has been enabled only relatively recently by the development of sufficiently clean and sufficiently thin bilayer devices [3–5]. The ongoing development of new 2D materials continues to provide novel platforms for realizing bilayer exciton physics [6–9]. Of particular significance is the identification of a number of different materials that have, or can be made to have, a Mexican hat shape at low energy $\varepsilon$. Such a dispersion relation is usually associated with Rashba spin-orbit coupling [10], which can be significant in semiconductors like GaAs or InAs [11–13], at the surface of topological insulators like Bi$_2$Se$_3$ [14], or at oxide interfaces such as LaAlO$_3$/SrTiO$_3$ [15]. A Mexican hat-shaped dispersion also arises in materials such as bilayer graphene [16,17] that have an avoided crossing between two intersecting bands with opposite-sign velocity. Importantly, for each of the preceding examples in this paragraph, the brim of the Mexican hat, $k_0$, can be widely and continuously adjusted by applying a transverse electric field. As shown below, this adjustability offers the

FIG. 1. Schematic illustration of a composite boson with tunable dispersion relation. A material containing electrons (blue layer) is placed adjacent to a material containing holes (yellow layer), with the electron dispersion relation following Eq. (1) and the hole dispersion relation following Eq. (2). In this configuration the electron and hole can bind together to form an interlayer exciton, which has a dispersion relation that can take a number of qualitatively different forms, depending on the value of $k_0$. 

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ability to continuously tune the dispersion relation of interlayer excitons.

The remainder of this paper is dedicated to deriving the exciton dispersion relation (Sec. II), and to discussing the different many-body phases that can be obtained by tuning $k_0$ and the exciton density $n$ (Sec. III). For simplicity, the amplitude of interlayer tunneling is considered everywhere to be negligibly small.

II. SINGLE-EXCITON DISPERSION RELATION

Before giving a detailed calculation of the exciton dispersion, it is worth outlining how the dispersion relation can be understood qualitatively using the following simple scaling arguments, which for simplicity assume small interlayer separation $d$. The key idea is to compare $k_0$ with the typical internal momentum $k_e$ of the electron within a stationary exciton. When $k_0$ is much smaller than $k_e$, the (small) Mexican hat feature in the electron dispersion is irrelevant, since the electron wave function uses primarily much larger momentum components. When $k_0 \gg k_e$, on the other hand, the Mexican hat structure in $\varepsilon_e(k_e)$ is reflected in the exciton dispersion relation, since the energy of the electron is strongly reduced when the exciton acquires enough momentum that the electron momentum approaches $k_0$.

Consider first the case where the hole mass is heavy, $m_h \gg m_e$. In this case the characteristic radius $r_h$ of the hole wave function is much smaller than that of the electron, $r_e$. Consequently, $r_h$ is similar to the radius of a 2D hydrogen atom with a stationary nucleus, $r_h \approx \kappa/m_e e^2 \equiv a_e$ (the electron effective Bohr radius, in Gaussian units with $\hbar = 1$; here $\kappa$ is the dielectric constant). This expression for $r_h$ is roughly correct even when $k_0$ is large, since large $k_0$ leads only to a logarithmic renormalization of $r_e$ [18]. The typical electron momentum is therefore $k_e \sim 1/a_e$. If $k_0$ is small enough that $k_0 \ll 1/a_e$, then the nonmonotonic part of the electron dispersion at small $k$ is smeared out by the internal motion of the electron around the hole, and the exciton dispersion is parabolic. Thus, it is only at larger $k_0$, such that

$$k_0 a_e \gg 1, \quad (m_h/m_e \gg 1), \quad (3)$$

that the exciton dispersion has a Mexican hat shape.

On the other hand, when the hole mass is light enough that $m_h \ll m_e$, the hole wave function acquires a large size $r_h \approx \kappa/m_h e^2$, while the electron wave function is relatively compact. The spatial extent of the electron wave function in this case can be found by considering that the electron sits in the bottom of a parabolic potential well created by the Coulomb potential of the hole. The shape of this potential well is $\varepsilon(r) \sim e^2 \rho^2/\kappa r_h^3$, where $\rho$ is the displacement of the electron from the center of the potential well, and the corresponding size of the electron wave function is that of the ground state of the 2D harmonic oscillator—namely, $r_e \sim (\hbar^2 k_0^2)^{1/4}$. (As in the previous case, this expression for $r_e$ is correct even for large $k_0$.) The Mexican hat shape of the dispersion relation remains only when $k_0 \gg 1/r_e$, which is equivalent to the condition

$$k_0 a_e \gg \left(\frac{m_h}{m_e}\right)^{3/4}, \quad (m_h/m_e \ll 1). \quad (4)$$

These qualitative predictions can be verified in a quantitative way as follows. The exciton dispersion $\varepsilon(P)$ is found by solving the Schrödinger equation $H \psi = \varepsilon \psi$ with the Hamiltonian

$$H = \hat{\varepsilon}_e + \hat{\varepsilon}_h - V(r_{eh}), \quad (5)$$

where $\hat{\varepsilon}_e$ and $\hat{\varepsilon}_h$ are the kinetic energy operators for the electron and hole, respectively, and

$$V(r_{eh}) = -\frac{e^2}{\kappa \sqrt{r_{eh}^2 + d^2}} \quad (6)$$

is their mutual Coulomb energy. Here $r_{eh} = r_e - r_h$ is the displacement vector between the electron and hole, with $r_e$ being the electron coordinate and $r_h$ the hole coordinate. The wave function with fixed total momentum $P$ is written as

$$\psi(r_e, r_h) = \exp \left[i \left(\mathbf{k}_e^0 \cdot r_e + \mathbf{k}_h^0 \cdot r_h\right)\right] \psi(r_{eh}), \quad (7)$$

where $\mathbf{k}_e^0$ and $\mathbf{k}_h^0$ are wave vectors such that $\mathbf{k}_e^0 + \mathbf{k}_h^0 = P$ and $(d\varepsilon_e/dk)_|_{\mathbf{k}=\mathbf{k}_e^0} = (d\varepsilon_h/dk)_|_{\mathbf{k}=\mathbf{k}_h^0}$. These two conditions ensure that the electron and hole together have total momentum $P$, and that they both have the same group velocity. (Equivalently, $\mathbf{k}_e^0$ and $\mathbf{k}_h^0$ are the values of momentum that minimize the total kinetic energy $\varepsilon_e(\mathbf{k}_e^0) + \varepsilon_h(\mathbf{k}_h^0)$ under the constraint $\mathbf{k}_e^0 + \mathbf{k}_h^0 = P$.) One can say that the distribution of electron momenta is centered around $\mathbf{k}_e^0$, while the hole has momenta in the neighborhood of $\mathbf{k}_h^0$. For concreteness, below I take $P$ to be in the $x$ direction, so that $\mathbf{k}_e^0$ and $\mathbf{k}_h^0$ are also in the $x$ direction.

The function $\psi(r_{eh})$ represents the wave function for the relative motion of the electron and hole around each other. Such motion is, in general, anisotropic, corresponding to a hydrogenlike state that is elongated in one direction. One can see how this anisotropy arises by considering the special case in which $k_0$ is large and $m_e$ is small. In this case the electron kinetic energy has a sharp minimum at $|\mathbf{k}| = k_0$, and $\mathbf{k}_e^0$ is very close to $k_0 \delta$. The dispersion relation for the electron in the neighborhood of $\mathbf{k}_e^0$ is therefore parabolic in the $x$ direction with mass $m_e$, and nearly flat in the $y$ direction. Consequently, the electron wave function acquires a shape that is tightly confined in the $y$ direction (extended in momentum space) and more extended in the $x$ direction (compact in momentum space).

In order to account for this anisotropy, one can use the variational wave function

$$\varphi(x, y) = \sqrt{\frac{2\beta^2 \lambda}{\pi}} \exp[-\beta \sqrt{x^2 + (\lambda y)^2}] \quad (8)$$

(following, for example, Refs. [19,20]). Here, $\beta$ and $\lambda$ are variational parameters, with $\beta$ corresponding to the inverse size of the wave function in the $x$ direction, and $\lambda > 1$ being the dimensionless anisotropy of the wave function. In the limiting case $k_0 \to 0$ and $d \to 0$, Eq. (8) reproduces the ground state wave function of the 2D hydrogen atom at $\lambda = 1$. More generally, the energy of the exciton is approximated by the minimum value of $\langle \psi | H | \psi \rangle$ over all values of the variational parameters. That is,

$$\varepsilon(P) \simeq \min_{\beta, \lambda} \left(\varepsilon_\lambda + \varepsilon_e\right), \quad (9)$$
FIG. 2. The dispersion relation \( \varepsilon(P) \) of an interlayer exciton with \( m_h = m_e = 1 \) and \( d/a_e \to 0 \). Different curves are labeled by the corresponding value of \( k_0 \). Units of energy on the vertical axis are \( e^2/\kappa a_e \). The feasibility of this transition can be checked numerically, for example, in Ref. [24]. In this approach, each lattice site (say \( P \)) is taken to be equivalent to a 2D layer exciton such high densities are generally not accessible: at much smaller densities \( n \sim 1/(\max(a_e, d)^2) \) the exciton wave functions overlap strongly with each other, and the individual excitons dissociate to form a state that resembles a uniform electron gas parallel to a uniform hole gas. Thus, at \( k_0 = 0 \) only liquid phases are typically possible—including, for example, a Bose-Einstein condensate at small enough \( d \) [23].

Consider, however, that as \( k_0 \) is increased from zero by the application of a perpendicular electric field, the dispersion relation of the excitons begins to flatten, as illustrated in Fig. 2. This flat dispersion implies that the quantum confinement energy associated with forming a crystalline state is much reduced. Correspondingly, a Wigner crystal state becomes energetically favored over a uniform state at sufficiently low density as \( k_0 a_e \) approaches unity. In other words, one can drive a liquid-to-solid transition at fixed density by increasing the strength of a transverse electric field.

The feasibility of this transition can be checked numerically in a simple way by examining a trial many-body wave function of dipolar bosons arranged on a triangular lattice (as employed, for example, in Ref. [24]). In this approach, each lattice site is taken to be the locus of a Gaussian wave packet of particle density, and the width \( w \) of the wave packet is used as a variational parameter. For such a wave function, the phase of the system can be estimated by examining the Lindemann ratio \( \eta = w/\ell \), where \( \ell = (2/\sqrt{3})^{1/2} n^{-1/2} \) is the lattice spacing.
energy below that of a simple Bose-Einstein condensate or Wigner crystal at low density, and so there must be some kind of unconventional state at large \( k_0 \).

IV. CONCLUDING REMARKS

This paper has demonstrated that the dispersion relation of an interlayer exciton can take a range of qualitatively different forms when one of its constituent particles (electron or hole) has a Mexican hat dispersion. This relatively simple result suggests the exciting possibility of realizing a 2D bosonic system for which the particles’ dispersion relation can be tuned continuously using a transverse electric field. Such tuning would allow one to realize novel types of quantum phase transitions.

At present, the full range of possible phases for this system remains to be completely explored. The analysis of Sec. III has demonstrated an instability toward forming a solid phase with increasing \( k_0 \) at a fixed density, but the regime of even larger \( k_0 \) is likely associated with more unconventional phases. The nature of these phases may provide fruitful ground for future studies, both theoretical and experimental. Indeed, while the majority of experimental proposals so far for realizing systems of “Mexican hat bosons” have involved optically driven Floquet bands or cold atomic gases \([10,30]\), the present work suggests that solid state bilayers or interfaces may be used as easily tunable platforms for studying the same physics. Particularly promising candidate materials include bilayer graphene \([17]\), the LaAlO\(_3/\)SrTiO\(_3\) interface \([15]\), or the surface states of Bi\(_2\)Se\(_3\) \([14]\), each of which has been shown to have a large, tunable Mexican hat effect when a transverse electric field is applied. A heterointerface between one of these materials and a conventional semiconductor can, in principle, house the field-tunable excitons discussed here.

Of course, the quantitative applicability of the calculations in this paper to a given solid state system require that Eqs. (1) and (2) are valid descriptions of the electron and hole dispersion relations. In particular, Eq. (1) should hold for all momenta of order \( k_e \), and smaller, where \( k_e \sim (1/a_e) \min\left\{1, (m_n/m_e)^{3/4}\right\} \) is the typical internal momentum of the electron (as discussed in Sec. II). The single band approximation implicit in Eqs. (1) and (2) also requires that the exciton binding energy be smaller than the band gap for either the electron or hole; otherwise one cannot describe the dispersion relation of the electron or hole without taking into account multiple bands. This single band condition is usually easily met in wide-bandgap semiconductors, while in gapped bilayer graphene it is generally limited to the case of small \( \alpha = \varepsilon^2/(\kappa \hbar v) \), where \( v \) is the graphene Dirac velocity \([18]\).

Finally, it is worth mentioning that in principle one can create a tunable excitonic dispersion relation even if the value of \( k_0 \) is fixed, provided that \( k_0 \) is large enough and that the small-\( k \) part of the electron spectrum can be tuned using some other parameter. For example, consider a system in which the electrons experience a strong Rashba spin-orbit coupling. If an external magnetic field is applied to such a system, the spin-split kinetic energy bands hybridize and the sharp maximum in energy in the lower band is reduced, as illustrated in Fig. 5. If this mechanism is used to tune the electron dispersion relation, then a similar tunability can be achieved for the exciton dispersion relation \([32]\).
A kinetic and potential energy of a single exciton

This appendix presents the derivations of Eqs. (10) and (11) of the main text, which describe the expectation values of the kinetic and potential energies of the exciton. The exciton wave function is given by Eqs. (7) and (8). The corresponding wave function \( \tilde{\psi}(k_e, k_h) \) in momentum space, defined by

\[
\psi(r_e, r_h) = \frac{1}{(2\pi)^3} \int d^2k_e d^2k_h \tilde{\psi}(k_e, k_h) \exp[i(k_e \cdot r_e + i k_h \cdot r_h)], \tag{A1}
\]

is

\[
\tilde{\psi}(k_e, k_h) = \frac{16(2\pi)^{5/2} \beta^2}{\sqrt{\lambda}} \times \frac{\delta^2(k_e + k_h - k_0^e - k_0^h)}{[4\beta^2 + (k_e - k_h - k_0^e + k_0^h)^2 + (k_h - k_0^h)^2]^{3/2}}. \tag{A2}
\]

Here, \( k_e = k_{ex} \hat{x} + k_{ey} \hat{y} \) is the electron momentum, and \( k_h = k_{hx} \hat{x} + k_{hy} \hat{y} \) is the hole momentum.

The expectation value of the kinetic energy is given by

\[
\epsilon_k = \int d^2r_e d^2r_h [\hat{\epsilon}_e(r_e, r_h) \psi^*(r_e, r_h) (\hat{\epsilon}_e + \hat{\epsilon}_h) \psi(r_e, r_h)], \tag{A3}
\]

where \( \hat{\epsilon}_e \) and \( \hat{\epsilon}_h \) are the kinetic energy operators for the electron and hole, respectively. Inserting Eq. (A1) into this expression, and using the fact that plane waves are eigenstates of the kinetic energy operators—i.e., that

\[
\hat{\epsilon}_e \exp[i(k \cdot r_e)] = \epsilon_e(k) \exp[i(k \cdot r_e)],
\]

\[
\hat{\epsilon}_h \exp[i(k \cdot r_h)] = \epsilon_h(k) \exp[i(k \cdot r_h)]
\]

gives the following general expression for the kinetic energy:

\[
\epsilon_k = \frac{1}{(2\pi)^2} \int d^2k d^2k_h [\epsilon_e(k_e) + \epsilon_h(k_h)] |\tilde{\psi}(k_e, k_h)|^2. \tag{A4}
\]

Inserting Eq. (A2) into Eq. (A4) and evaluating one of the two momentum integrals gives Eq. (10).

The potential energy of the exciton is given generally by

\[
\epsilon_c = -\int d^2r_e d^2r_h |\psi(r_e, r_h)|^2 \frac{e^2}{\kappa \sqrt{(r_e - r_h)^2 + d^2}} = -\int d^2r_{eh} |\varphi(r_{eh})|^2 \frac{e^2}{\kappa \sqrt{r_{eh}^2 + d^2}}. \tag{A5}
\]

Inserting Eq. (8) into this expression gives Eq. (11). In the special case \( d = 0 \), the integral expression for \( \epsilon_c \) can be evaluated analytically, giving

\[
\epsilon_c = -\frac{2e^2\beta}{\pi \kappa} [K(1 - 1/\lambda^2) + \lambda K(1 - \lambda^2)], \tag{A6}
\]

where \( K(x) \) is the complete elliptic integral of the first kind. An equivalent expression to Eq. (A6) has been derived previously, for example in Refs. [20,33].

APPENDIX B: ENERGY OF A TRIAL MANY-BODY STATE OF DIPOLAR BOSONS

In Sec. III, the feasibility of a liquid-to-solid transition driven by increasing \( k_0 \) was examined using a simple trial wave function. Here I provide some details about this calculation, which largely follows the one presented in Ref. [24].

In this approach, the interlayer excitons are treated as bosonic particles with a dipolar interaction law and the dispersion relation \( \epsilon(k) \) that results from the procedure described in Sec. II. The many-body wave function for the system is taken to be a product of Gaussian wave packets \( \phi_i(r) \), each centered around some point \( r_{ij} \) on the triangular lattice. These wave packets are given by

\[
\phi_i(r) = \frac{1}{\sqrt{\pi} w^2} \exp\left[-\frac{|r - r_{ij}|^2}{2w^2}\right]. \tag{B1}
\]

so that the corresponding uncertainty in position for each particle is \( \sqrt{r^2} = w \). The Fourier transform of this wave packet is given by

\[
\tilde{\phi}(k) = \sqrt{4\pi w^2} \exp[-k^2 w^2/2], \tag{B2}
\]

and the corresponding kinetic energy per particle is

\[
\epsilon_{kin} = \frac{1}{(2\pi)^2} \int d^2k \epsilon(k) |\tilde{\phi}(k)|^2. \tag{B3}
\]

The value of \( \epsilon_{kin} \) is determined by numerically evaluating this integral for each value of \( w \) and each instance of the dispersion relation \( \epsilon(k) \). For the case where \( \epsilon(k) = (k - k_0)^2/2m \), the

FIG. 5. Schematic illustration of an alternate scheme for realizing a tunable exciton dispersion. For an electron dispersion with strong Rashba spin-orbit coupling (top left), the energy at small \( k \) can be modified by application of a magnetic field. (Here, the lower curves correspond to increasingly high magnetic field values.) If such an electron dispersion is combined with a parabolic hole dispersion (lower left), the resulting exciton can have a dispersion relation \( \epsilon(P) \) that has a range of different forms (right).

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kinetic energy is given by
\[ \varepsilon_{\text{kin}} = \frac{1}{2mw^2} [1 + k_0w(k_0w - \sqrt{\pi})]. \]

The interaction energy (Hartree energy) per particle is [34]
\[ \varepsilon_{\text{int}} = \frac{n}{2} \sum_{q \in G} \tilde{V}(q) \exp[-q^2 w^2/2] - \frac{1}{2(2\pi)^2} \int d^2 k \tilde{V}(q) \exp[-q^2 w^2/2]. \]  
(B4)

Here, \( n \) is the particle density, \( G \) labels the set of all reciprocal lattice vectors of the triangular lattice, and
\[ \tilde{V}(q) = \frac{4\pi e^2}{kq} (1 - \exp[-qd]) \]  
(B5)

is the Fourier-transformed interaction law between dipoles with a dipole arm \( d \). The second term on the right-hand side of Eq. (B4) removes the self-interaction term from the sum in the first term. The set of reciprocal lattice vectors are defined by
\[ q_{ij} = 2\pi i + 4j \sqrt{3} \]  
(B6)

where the indices \( i \) and \( j \) run over all integers.

In the variational method, the value of the positional uncertainty \( \sqrt{\langle r^2 \rangle} \) is estimated to be equal to the value of \( w \) which minimizes the total energy per particle, \( \varepsilon_{\text{kin}} + \varepsilon_{\text{int}} \).

[1] This tunability is similar in spirit to the problem of mobile magnetic impurities in a superfluid [35].


