Higgs modes in two-dimensional spatially-indirect exciton condensates

Fei Xue,1,2,3 Fengcheng Wu,1,4 and A.H. MacDonald1

1Department of Physics, University of Texas at Austin, Austin TX 78712, USA
2Physical Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
3Institute for Research in Electronics and Applied Physics & Maryland Nanocenter, University of Maryland, College Park, MD 20742
4Condensed Matter Theory Center and Joint Quantum Institute, Department of Physics, University of Maryland, College Park, Maryland 20742, USA

(Dated: March 4, 2020)

Higgs modes in condensed matter physics have drawn attention because of analogies to the Higgs bosons of particle physics. Here we use a microscopic time-dependent mean-field theory to study the collective mode spectra of two-dimensional spatially indirect exciton (electron-hole pair) condensates, focusing on the Higgs modes, i.e., those that have a large weight in electron-hole pair amplitude response functions. We find that in the low exciton density (BEC) limit, the dominant Higgs modes of spatially indirect exciton condensates correspond to adding electron-hole pairs that are orthogonal to the condensed pair state. We comment on the previously studied Higgs-like collective excitations of superconductors in light of this finding.

PACS numbers:

I. INTRODUCTION

The standard model of particle physics posits a bosonic Higgs field that provides elementary particles with mass by breaking symmetries that would otherwise be present. The recent experimental detection1,2 of Higgs particles, the elementary excitations of the Higgs field, is therefore an important advance in fundamental physics. Partly because of their importance to the foundations of physics writ large, there has also been interest in excitations that are analogous to Higgs particles in condensed matter, especially in superconducting metals3. Indeed, the absence of massless Goldstone boson excitations in superconductors4,5 in spite of their broken gauge symmetry, played an important role historically in the theoretical work6–8 that led to the Higgs field proposal.

Emergent symmetry breaking bosonic fields are common in condensed matter, where they typically arise from interactions among underlying fermionic fields. Both electron-electron pair fields, which condense in superconductors, and electron-hole pair fields, which condense in ferromagnets and in spin or charge density wave systems, are common. An important difference between the Higgs fields of particle physics and the symmetry breaking bosonic fields in condensed matter is the absence, in the former case, of an understanding the field’s origin in terms of underlying degrees of freedom that might be hidden at present, akin to the understanding in condensed matter that the order parameter field in a superconductor measures electron-electron pair amplitudes. Such an understanding might eventually be achieved, and analogies to the observed properties of condensed matter might one again be valuable in suggesting theoretical possibilities. Motivated partly by that hope, and partly with the goal of shedding new light on the interesting literature9–17 on Higgs-like excitation in superconductors and in other condensed matter systems18–23, we address the Higgs-like excitations of two-dimensional spatially indirect exciton condensates.

Spatially indirect exciton condensates (SIXCs) are equilibrium or quasi-equilibrium states of matter that have been extensively studied over the past couple of decades in semiconductor bilayer quantum wells24–27, including in the quantum Hall regime28,29. SIXCs have recently been observed in van der Waals heterojunction two-dimensional bilayer materials both in the presence30,31 and in the absence32,33 of external magnetic fields. The bosonic order parameter field of a spatially indirect exciton condensate

$$\Delta(\vec{r}) = \left\langle \Psi_t^\dagger(\vec{r}) \Psi_b(\vec{r}) \right\rangle$$

has a non-zero expectation value in the broken symmetry ground state, which is characterized by spontaneous interlayer coherence and a suite of related anomalous transport properties34. The labels b, t on the field operators in Eq. 1 refer to electrons in the bottom
(b) and top (t) layers of a bilayer two-dimensional electron system, as illustrated schematically in Fig. 1. The SIXC state can be described approximately using a mean-field theory, analogous to the Bardeen-Cooper-Schrieffer mean-field-theory of superconductors.

Superconductors break an exact gauge symmetry related to conservation of electron number in the many-body Hamiltonian. In electron-hole pair condensates the corresponding symmetry is only approximate, but becomes accurate when the electrons and holes are selected from two different subsets of the single-particle Hilbert space whose electron numbers are approximately conserved separately. In the case of spatially indirect exciton condensates, the electrons and holes are selected from separate two-dimensional layers. Exceptionally among electron-hole pair condensates, the Hamiltonian terms that break separate particle-number conservation can be made arbitrarily weak simply by placing an insulating barrier between the two-dimensional subsystems. Phenomena associated with broken gauge symmetries can be realized as fully as desired by suppressing single-particle processes that allow electrons to move between b and t layers. The properties of spatially indirect exciton condensates are therefore very closely analogous to those of two-dimensional superconductors, as we shall emphasize again below. The main difference between the two cases is that the condensed pairs are charged in the superconducting case, altering how the ordered states interact with electromagnetic fields.

In this article we employ a time-dependent-mean-field weak-coupling theory description of the bilayer exciton condensate’s elementary excitations to identify Higgs-like modes, and to demonstrate that in the low-density BCS limit they have a simple interpretation as excitations in which electron-hole pairs are added to the system in electron-hole pair states that are orthogonal to the 1s pair state that is condensed in the many-body ground-state. In Section I we briefly describe some details of our theory of the SIXC’s collective excitations. In Section II we summarize and discuss numerical results we have obtained by applying this theory to bilayer two-dimensional electron-hole systems. Finally, in Section IV we conclude by commenting on similarities and differences between bilayer exciton condensates and other systems in which Higgs modes have been proposed and observed.

II. COLLECTIVE EXCITATION THEORY

The mean-field theory of the bilayer exciton condensate is a generalized Hartree-Fock theory in which translational symmetry is retained but spontaneous interlayer phase coherence, which breaks separate conservation of particle number in the two layers, is allowed. In Ref. 35 we presented a theory of the BXC’s elementary collective excitations and quantum fluctuations that accounts for quadratic variations of the Hartree-Fock energy functional. Importantly for the findings that are the focus of the present paper, the theory fully accounts for the long range Coulomb interactions among electrons and holes. Theories that do not recognize the Coulomb’s interactions long range, or which do not treat electrostatic and exchange interactions on an equal footing, make qualitative errors in describing spatially indirect exciton condensates. This comment applies in particular to the short-range interaction models that can be conveniently analyzed using Hubbard-Stratonovich transformations. In this Section we briefly summarize that theory and generalize it in a way that makes evaluation of the two-particle Greens functions that characterize the system’s particle-hole excitations particularly convenient. The SIXC’s elementary excitations energies are identified with the poles of those Greens functions and are the eigenvalues of a matrix constructed from the kernel of the quadratic-fluctuation energy functional. The character of given elementary excitations is classified by determining which particle-hole pair response functions have large residues at its poles.

A. Mean Field Theory

For simplicity we neglect the spin and valley degrees of freedom that often play a role in realistic SIXC systems. The self-consistent Hartree-Fock mean-field Hamiltonian of the broken symmetry BXC state is then

\[ H_{MF} = \sum_{\vec{k}} (a_{\vec{k}c}^\dagger a_{\vec{k}v}^\dagger - \Delta_{\vec{k}} \sigma_z) \left( \begin{array}{c} a_{\vec{k}c} \\ a_{\vec{k}v} \end{array} \right). \] (2)

Here \( a_{\vec{k}n} \) and \( a_{\vec{k}k}^\dagger \) are fermionic annihilation and creation operators for the conduction \((n = c)\) band electrons localized in the top layer and valence \((n = v)\) electrons localized in the bottom layer, \( \sigma_z \) are Pauli matrices that act in the band space, and \( \zeta_{\vec{k}} = \hbar^2 k^2 / (4m_v) \) accounts for the difference between conduction and valence band effective masses, which plays no role in the temperature \( T = 0 \), charge neutral limit that we consider. For convenience, we take \( m_v = m_h \) in the calculations described below. The dressed band parameters \( \zeta_{\vec{k}} \) and \( \Delta_{\vec{k}} \) are obtained by solving the self-consistent-field equations:

\[ \zeta_{\vec{k}} = \frac{\hbar^2 k^2}{4m} + \frac{\tilde{\mu}}{2} - \frac{1}{2A} \sum_{\vec{k}'} V_{\vec{k} - \vec{k}'} (1 - \zeta_{\vec{k}'} / E_{\vec{k}'}). \]

\[ \Delta_{\vec{k}} = \frac{1}{2A} \sum_{\vec{k}'} U_{\vec{k} - \vec{k}'} \Delta_{\vec{k}'} / E_{\vec{k}'} . \] (3)

\[ E_{\vec{k}} = \sqrt{\zeta_{\vec{k}}^2 + \Delta_{\vec{k}}^2}. \]

where \( m = m_e m_h / (m_e + m_h) \) is the reduced mass, and \( A \) is the area of two-dimensional system. In Eq. 3 \( V_{\vec{q}} = 2\pi e^2 / (\epsilon q) \) and \( U_{\vec{q}} = V_{\vec{q}} \exp( - q d) \) are the intra-layer and
inter-layer Coulomb interactions,
\[
\tilde{\mu} = \mu + 4\pi e^2 n_{ex} d/\epsilon, \\
n_{ex} = \frac{1}{2A} \sum_k (1 - \xi_k/E_k),
\]
(4)
\(\mu\) is the chemical potential for excitons, and \(n_{ex}\) is equal to both the density of conduction band electrons and the density of valence band holes. Below we refer to \(n_{ex}\) as the density of excitons, this terminology is motivated mainly by the low-density limit in which \(n_{ex}a_B^2 \ll 1\). (Here \(a_B^* = \hbar^2/\epsilon me^2\) is the Bohr radius, which is the bound electron-hole pair size in the limit of small layer separations.) The exciton chemical potential \(\mu = E_g - V_b\) can be adjusted electrically by applying a gate voltage to alter the spatially indirect band gap \(E_g\), provided that the barrier between conduction and valence band layers is sufficiently opaque, or by applying a bias voltage \(V_b\) between layers\(^2\).

The mean-field ground state is
\[
|XC\rangle = \prod_k \gamma_{k,0}^\dagger |0\rangle = \prod_k (u_k^a c_k^a + v_k a_{vk}^a) |0\rangle,
\]
(5)
where
\[
u_k^c = \sqrt{\frac{1}{2}} (1 - \xi_k/E_k), \\
v_k^v = \sqrt{\frac{1}{2}} (1 + \xi_k/E_k),
\]
(6)
and \(\gamma_{k,0}^\dagger\) is the creation operator for the dressed valence band quasiparticle states that are occupied in \(|XC\rangle\). Note that we have chosen \(u_k^c\) and \(v_k^v\) to be real, and that there is family of degenerate states that differ only by a global shift in the phase difference between electrons localized in different layers.

B. Quadratic Fluctuations

We construct our theory of quantum fluctuations and collective excitations by starting from a many-body state that incorporates arbitrary single particle-hole excitation corrections to the mean-field state:
\[
|\Phi\rangle = \prod_k [Z_k + \sum_{\tilde{\mathbf{Q}}} z_{k}(\tilde{\mathbf{Q}}) \gamma_{k+\tilde{\mathbf{Q}},1}^\dagger \gamma_{k,0}] |XC\rangle,
\]
(7)
where \(\gamma_{k,1}^\dagger\) is a creation operator for a quasiparticle state in the band that is empty in \(|XC\rangle\):
\[
\gamma_{k,1}^\dagger = v_k a_{vk}^a - u_k a_k^a,
\]
(8)
and
\[
Z_k = \sqrt{1 - \sum_{\tilde{\mathbf{Q}}} |z_k(\tilde{\mathbf{Q}})|^2}
\]
(9)
is a normalization factor. The complex parameters \(z_k(\tilde{\mathbf{Q}})\) are the amplitudes of all possible single particle-hole excitations.

To characterize the quantum fluctuations of the mean-field state in a physically transparent way, we define the observables
\[
\hat{r}_\alpha = \{x,y,z\} |\tilde{\mathbf{Q}}\rangle = \frac{1}{2} \sum_k (a_k^a c_{k+\tilde{\mathbf{Q}}}^a a_{vk+\tilde{\mathbf{Q}}}^a) \sigma^a (a_k^c a_{vk}^v). \\
\]
(10)
Note that \(\langle \Phi | \hat{r}_\alpha (\tilde{\mathbf{Q}}) | \Phi \rangle = \langle \Phi | \hat{r}_\alpha (-\tilde{\mathbf{Q}}) | \Phi \rangle^*\). For the inter-layer phase choice we have made, the mean-field value of the order parameter \(\Delta^{MF}\) is real and spatially constant:
\[
\Delta^{MF} = \frac{1}{A} \sum_k u_k v_k 
\]
(11)
where \(A\) is the sample area. When fluctuations are included, the order parameter becomes
\[
\Delta(r) = \frac{1}{A} \sum_{\tilde{\mathbf{Q}}} \langle \tau_z(\tilde{\mathbf{Q}}) \rangle \cos(\tilde{\mathbf{Q}} \cdot r - \varphi_{\tilde{\mathbf{Q}}} x) + i \langle \tau_y(\tilde{\mathbf{Q}}) \rangle \cos(\tilde{\mathbf{Q}} \cdot r - \varphi_{\tilde{\mathbf{Q}}} y) 
\]
(12)
where \(\langle \ldots \rangle = \langle \Phi | \ldots | \Phi \rangle\) and \(\varphi_{\tilde{\mathbf{Q}}} \) is defined by \(\langle \tau_\alpha(\tilde{\mathbf{Q}}) \rangle = \langle \tau_\alpha(\tilde{\mathbf{Q}}) \rangle \exp(i \varphi_{\tilde{\mathbf{Q}}} \alpha)\). It follows that, to leading order, fluctuations in the order parameter magnitude are proportional to \(\langle \tau_z(\tilde{\mathbf{Q}}) \rangle\), while fluctuations in the order parameter phase are related to \(\langle \tau_y(\tilde{\mathbf{Q}}) \rangle\). \(\langle \tau_z(\tilde{\mathbf{Q}}) \rangle\) measures fluctuations in the exciton density.

We quantize fluctuations in the XC state by constructing the Lagrangian:
\[
L = \langle \Phi | i\hbar \partial_t - H | \Phi \rangle \approx B - \delta E^{(2)},
\]
(13)
where \(\delta E^{(2)}\) is the harmonic fluctuation energy functional\(^3\) and \(B = \langle \Phi | i\hbar \partial_\mathbf{r} | \Phi \rangle\) is the Berry phase term which enforces bosonic quantization rules on the \(z_k(\tilde{\mathbf{Q}})\).
fluctuation parameters. The energy functional is obtained by taking the expectation value of the many-body Hamiltonian and has the form

$$\delta E^{(2)} = \langle \Phi | H | \Phi \rangle = \sum_{\vec{Q}, \vec{k}, \vec{p}} \{ E_{\vec{k}, \vec{p}}(\vec{Q}) z_{\vec{k}}^{*}(\vec{Q}) z_{\vec{p}}(\vec{Q}) + \frac{1}{2} \Gamma_{\vec{k}, \vec{p}}(\vec{Q}) [z_{\vec{k}}(\vec{Q}) z_{\vec{p}}^{*}(-\vec{Q}) + z_{\vec{k}}^{*}(\vec{Q}) z_{\vec{p}}(-\vec{Q})] \}. \quad (14)$$

$$\delta E^{(2)}$$ accounts for variations in band kinetic energy and Hartree and exchange interaction energy as the many-electron state fluctuates. Explicit forms for the matrices $E$ and $\Gamma$ are given in Appendix A.

We separate the fluctuation Hamiltonian into amplitude and phase fluctuation contributions by making the change of variables:

$$z_{\vec{k}}(\vec{Q}) = \frac{1}{\sqrt{2}} (x_{\vec{k}}(\vec{Q}) + iy_{\vec{k}}(\vec{Q})), \quad (15)$$

$$z_{\vec{k}}^{*}(-\vec{Q}) = \frac{1}{\sqrt{2}} (x_{\vec{k}}(-\vec{Q}) - iy_{\vec{k}}(-\vec{Q})). \quad (16)$$

Note that although $(\tau_{x}(\vec{Q}))$ and exciton density $(\tau_{z}(\vec{Q}))$ fluctuations are both related to $x_{\vec{k}}(\vec{Q})$, they have different $\vec{k}$-dependent weighting factors. For each wavevector transfer $\vec{Q}$ we define vectors of $x$ and $y$ variables, $X(\vec{Q}) \equiv (x_{\vec{k}_{1}}(\vec{Q}), \ldots, x_{\vec{k}_{i}}(\vec{Q}), \ldots)$ and $Y(\vec{Q}) \equiv (y_{\vec{k}_{1}}(\vec{Q}), \ldots, y_{\vec{k}_{i}}(\vec{Q}), \ldots)$, whose elements are labelled by the particle-hole pair’s hole-momentum. In terms of these vector variables the action

$$S = \int dt (B - \delta E^{(2)}) = \frac{1}{2} \sum_{\vec{Q}} \int dt \left( \hbar \nabla^{\dagger}(\vec{Q}) \partial_{t} X(\vec{Q}) - \hbar X^{\dagger}(\vec{Q}) \partial_{t} Y(\vec{Q}) - X^{\dagger}(\vec{Q}) \mathcal{K}^{(+)} X(\vec{Q}) - Y^{\dagger}(\vec{Q}) \mathcal{K}^{(-)} Y(\vec{Q}) \right), \quad (18)$$

where $\mathcal{K}^{(\pm)}(\vec{Q}) = (E_{\vec{k}, \vec{p}} \pm \Gamma_{\vec{k}, -\vec{p}})(\vec{Q})$ is real and symmetric for both sign choices and we have used the fact that $\int dt Y^{\dagger}(\vec{Q}) \partial_{t} X = - \int dt X \partial_{t} Y^{\dagger}$.

Minimizing the action yields the following equations of motion:

$$\hbar \partial_{t} X = \mathcal{K}^{(-)} Y(\vec{Q})$$

$$\hbar \partial_{t} Y = -\mathcal{K}^{(+)} X(\vec{Q}) \quad (19)$$

This theory of fluctuations is equivalent to time-
dependent Hartree-Fock theory for the exciton condensate state response functions. It is in the same spirit as auxiliary field functional integral theories of harmonic quantum fluctuations, but unlike those approaches treats Hartree and exchange energy contributions on an equal footing, an attribute that is necessary if the bilayer exciton condensate is to be described directly.

C. Particle-Hole Correlation Functions

Collective modes give rise to poles in particle-hole channel Greens functions. As in the case of BCS superconductor\cite{31,30}, those collective modes that have large residues in \((\hat{\tau}_x, \hat{\tau}_z)\) particle-hole Greens functions can be identified as Higgs-like modes. Because the fluctuation Hamiltonian \(\delta E^{(2)}\) is the sum of quadratic contributions in the \(X\) and \(Y\) fields, which are canonically conjugate, we can apply the generalized Bogoliubov transformation\cite{29} described in details below to write the fluctuation Hamiltonian as a free-boson form:

\[
H = E_0 + \sum_{\vec{Q}} \sum_i h\omega_i(\vec{Q})B_i^\dagger(\vec{Q})B_i(\vec{Q}).
\]

where \(h\omega_i(\vec{Q})\) is an excitation energy and \(B_i^\dagger(\vec{Q})\) and \(B_i(\vec{Q})\) are linear combinations of the \(x_k\) and \(y_k\) fields. To evaluate correlation functions involving the \(\tau_\alpha(\vec{Q})\) fields we reexpress \(x_k(\vec{Q})\) and \(y_k(\vec{Q})\) in Eqs.\cite{17} in terms of these free boson fields. The character of collective excitations is revealed by the residues of response functions at poles that lie below particle-hole continua.

To carry out this procedure explicitly, we start from the assumptions that the amplitude/density kernel \(\mathcal{K}^{(+)}\) is positive definite at any \(\vec{Q}\), and that the phase kernel \(\mathcal{K}^{(-)}\) is positive definite for \(\vec{Q} \neq 0\) and positive semi-definite \(\vec{Q} = 0\). These assumptions are satisfied whenever the mean-field condensate is metastable. The zero eigenvalue of \(\mathcal{K}^{(-)}\) at \(\vec{Q} = 0\) arises from the broken U(1) symmetry associated with spontaneous interlayer phase coherence. Because \(\mathcal{K}^{(+)}\) is real symmetric and positive definite, it is possible\cite{28} to perform a Cholesky decomposition for each \(\vec{Q}\) by writing

\[
\mathcal{K}^{(+)} = LL^T,
\]

and then to diagonalize

\[
\Gamma = L^T\mathcal{K}^{(-)}L = \Gamma^T.
\]

Writing \(\Gamma = SAS^T\) where \(S\) is an orthogonal matrix and \(\Lambda\) is a diagonal matrix, we define new fields

\[
\Psi = S^T L^{-1} Y,
\]

\[
\Pi = S^T L^T X,
\]

where \(X, Y\) are the density and phase fluctuation vectors labeled by wavevector \(\vec{k}\) introduced above. The transformed Hamiltonian is

\[
\mathcal{H} = \frac{1}{2}(X^T \mathcal{K}^{(+)} X + Y^T \mathcal{K}^{(-)} Y) = \frac{1}{2}(\Pi^\dagger \Pi + \Psi^\dagger \Lambda \Psi)
\]

\[
= \frac{1}{2} \sum_i (|\Pi_i|^2 + \hbar^2 \omega_i^2 |\Psi_i|^2).
\]

Noting that the eigenvalues of \(\Gamma\) are identical to the eigenvalues of \(\mathcal{K}^{(+)}\mathcal{K}^{(-)}\), and that the equation of motion for phase fluctuations can be written in the form

\[
- \hbar^2 \partial_x^2 Y = \mathcal{K}^{(+)}\mathcal{K}^{(-)} Y,
\]

we have identified them as the squares \(\omega_i^2\) of the elementary excitation frequencies.

When expressed in terms of the normal mode fields, the action in Eq.\cite{18} has the form

\[
S = \frac{1}{2} \sum_{\vec{Q}} \int dt \left( \hbar \Psi^\dagger \partial_t \Pi - \hbar \Pi^\dagger \partial_t \Psi - \Pi^\dagger \Pi - \Psi^\dagger \Lambda \Psi \right) \vec{Q}.
\]

The time-ordered Green function at each \(\vec{Q}\) can be calculated directly from the action of fields \(\phi\),

\[
G(\omega) = \left( \begin{array}{cc} -\Lambda & i\hbar \omega \\ -i\hbar \omega & -1 \end{array} \right)^{-1}
\]

\[
= (\det(\Lambda - \hbar^2 \omega^2))^{-1} \left( \begin{array}{cc} -\Lambda & -i\hbar \omega \\ i\hbar \omega & -\Lambda \end{array} \right).
\]

Note that the Green function is a 2 by 2 matrix in the basis of fields \(\{\Psi, \Pi\}\). The \(\tau_\alpha\) fields can be expressed in terms of the normal mode fields for each \(\vec{Q}\) using

\[
\tau_x = \sum_i (T_x(L^T)^{-1} S)_i \Pi_i \equiv \tau_{x,i} \Pi_i
\]

\[
\tau_z = \sum_i (T_z(L^T)^{-1} S)_i \Pi_i \equiv \tau_{z,i} \Pi_i
\]

\[
\tau_y = \sum_i (T_y(L^T) \Pi)_i \equiv \tau_{y,i} \Pi_i
\]

where the \(T_\alpha\) on the right-hand side of these equations are the matrix forms of Eq.\cite{17}. The linear response functions are related to the time-ordered Green functions,

\[
\chi_{AB} = -\frac{i}{\hbar} \langle T[\hat{A}(t), \hat{B}(t')]\rangle
\]

The response functions of operators expressed in terms of fields \(\Pi_i\) can be evaluated by performing the average in Eq.\cite{29} using the quadratic action weighting factor with the result that

\[
\chi_{AB} = \sum_i \frac{\omega_i}{2} \left( A_{0i} B_{0i} - \frac{A_{0i} B_{0i}}{\omega - \omega_i + i\eta} - \frac{A_{0i} B_{0i}}{-\omega + \omega_i + i\eta} \right),
\]

where \(A_{mn}\) is the matrix element in a complete set of fields \(\Pi_i\). We identify Higgs modes by finding isolated
eigenvalues $|\omega_i|^2$ with large values of $|\tau_{xx,i}|^2$ in the imaginary part of the response functions for positive frequencies:

$$\text{Im} \chi_{xx}(\omega) = -\pi \sum_i \frac{\omega_i}{2} |\tau_{xx,i}|^2 \delta(\omega - \omega_i).$$  \hspace{1cm} (31)$$

An alternative approach to obtain the same results is to map the diagonalized action in Eq. (20) to that of a set of independent harmonic oscillators, defining the oscillator ladder operators $B_i$ by

$$\Pi_i = i \sqrt{\frac{\hbar \omega_i}{2}} (B_i^\dagger - B_i)$$
$$\Psi_i = \sqrt{\frac{1}{2\hbar \omega_i}} (B_i^\dagger + B_i),$$  \hspace{1cm} (32)$$

where the $B_i$ satisfy $[B_i, B_j^\dagger] = \delta_{i,j}$. The fluctuation Hamiltonian for each $\vec{Q}$ is then

$$\mathcal{H} = E_0 + \sum_i \hbar \omega_i B_i^\dagger B_i.$$  \hspace{1cm} (33)$$

From the general linear response theory, the Lehmann representation of the response function is

$$\chi_{AB}(\omega) = \frac{1}{\hbar} \sum_{m,n} \frac{P_m - P_n}{\omega - \omega_{nm} + i\eta} A_{mn} B_{nm},$$  \hspace{1cm} (34)$$

where $P_n = \frac{e^{-\beta E_n}}{\sum_m e^{-\beta E_m}} (\beta = 1/k_B T)$ is the occupation probability, $\omega_{nm} = (E_n - E_m)/\hbar$ is the excitation frequency, and $A_{mn} \equiv \langle \psi_m | A | \psi_n \rangle$ is the matrix elements in a complete set of exact eigenstates $|\psi_n\rangle$ of $\mathcal{H}$. At zero temperature, the imaginary part of response function is:

$$\text{Im} \chi_{AB} = -\frac{\pi}{\hbar} \sum_{m,n} P_m A_{mn} B_{nm} \delta(\omega - \omega_{nm})$$
$$- A_{mn} B_{nm} \delta(\omega + \omega_{nm})$$
$$= -\frac{\pi}{\hbar} \sum_{n,m} (A_{mn} B_{nm} \delta(\omega - \omega_{nm}) - A_{nm} B_{mn} \delta(\omega + \omega_{nm}))$$  \hspace{1cm} (35)$$

III. RESULTS

We now apply the theory outlined above to bilayer exciton condensates. The length and energy units we use in our calculations are those appropriate for Coulomb interactions, the Bohr radius $a_B^* = e^2/(\hbar^2/m^*)$ and the effective Rydberg $Ry^* = e^2/((2\alpha_B^*)^2)$. Typical values of these parameters in transition metal dichalcogenides bilayers are $a_B^* \approx 10\AA, Ry^* \approx 100\text{meV}$, while typical values for GaAs bilayer quantum wells are $a_B^* \approx 100\AA, Ry^* \approx 5\text{meV}$. For all the numerical calculations, we assume $m_e = m_h$, and use $d/a_B^* = 0.5$. Because our momentum-space grids are necessarily discrete, corresponding to applying periodic boundary conditions to a finite area system, the number of particle-hole pairs at a given excitation momentum is finite. The distinctions between particle-hole continua and isolated collective modes made below are qualitative, but for the most part unambiguous.

![FIG. 2: (Color online) Spectra of the imaginary part of $\tau_x - \tau_y$ and $\tau_z - \tau_x$ response functions. Black dots represent the positive collective excitation energies. Blue bars denote the $\chi_{xx}$ response amplitude and red bars denote the $\chi_{xx}$ response amplitude. The purple dashed line denotes the location of electron-hole continuum (minimun of $E_{ex}$ and $E_{ex+Q}$). (a) and (b) show the results of $Q = 0$ at low exciton density ($n_{ex}a_B^* = 0.008$) and high exciton density ($n_{ex}a_B^* = 0.08$) respectively. (c) and (d) show similar results at finite center of mass momentum. Bilayer exciton condensates have a BEC-BCS crossover that can be tuned by varying not the strength of interactions, as in cold atom systems, but the Fermi energy of the underlying electrons and holes. In two dimensions the binding energy of a single electron-hole pair is $4Ry^*$, and the Fermi energy in Ry$^*$ units is $2\pi n_{ex} a_B^*$. The bilayer exciton condensate therefore approaches a BEC limit for small values of $n_{ex} a_B^*$ and approaches a BCS limit for $n_{ex} a_B^* \gtrsim 0.1$. In Fig 2(a) and (c), we plot $I m \chi_{xx}$ (red bars) as a function of collective mode energies (black dots) for $\vec{Q} = 0$ and $\vec{Q} a_B^* = 1.5$ for a low exciton density in the BEC regime, $n_{ex} a_B^* = 0.08$. These calculations identify a collective mode at an energy below the particle-hole continuum (purple dashed line) that has a large weight in the ($\tau_x, \tau_z$) pair-amplitude (red bars in Fig. 2(a)). This result is reminiscent of the finding in earlier work that for superconductors there is a collective mode at the edge of the excitation continuum with a large residue in the pair-amplitude response function. At finite excitation wavevector $\vec{Q}$ additional modes have significant pair amplitude character. We interpret these findings more fully below.]

The corresponding results for $\chi_{zz}$ (exciton density) are presented as blue bars in Figs 2(c). As we see by comparing the locations of blue and red bars, the collective modes that have large weights are quite different in the two cases. In particular, the exciton-density response always has biggest weight in the lowest excitation Goldstone mode, which in the BEC regime has low energy compared to the particle-hole continuum gap even for large excitation wavevectors. Although both responses
which more fully, we examine the low carrier density limit in correlation function methods. In the dilute limit the matrices $u$ that to lowest order in $\tau x - \tau s$ response are neutral, whereas it has a finite energy in the three-dimensional electron-electron pair case of superconductors because of the divergence in the Coulomb interactions as $Q \to 0$. The collective modes that have large weight in the $\chi_{xx}$ response function are entirely different in character. At $Q = 0$, Eq. (31) forbids any response of Goldstone mode because $\omega_{GS} = 0$. However, a few peaks appear in the $\tau x - \tau x$ response below the particle-continuum and they are Higgs modes identified by our correlation function methods.

To understand the character of the Higgs-like modes more fully, we examine the low carrier density limit in which $u_k$ has small values at all $\vec{k}$. From Eq. (17) it follows that to lowest order in $u_k$:

$$K^{(\pm)} = \delta_{\vec{k},\vec{p}}(E_{\vec{k}} + E_{\vec{k}+\vec{Q}}) - \frac{1}{A} U(\vec{k} - \vec{p}),$$

$$\tau_{x,y}(\vec{Q}) = \sum_k x_k(\vec{Q}),$$

$$\tau_z(\vec{Q}) = \sum_k (u_k^* + u_{\vec{k}+\vec{Q}}) x_k(\vec{Q}).$$

In the dilute limit the matrices $K^{(+)} = K^{(-)}$ reduce to the two-particle electron-hole relative motion Hamiltonian matrices at center of mass wavevector $\vec{Q}$. This very dilute exciton condensate limit becomes a standard two-dimensional hydrogen-like problem where each excitation can be characterized by atomic-like orbitals, such as $1s$, $2s$, $2p$, etc. Note that in this limit, the $\chi_{zz}$ response weighting factor projects out relative-motion states that are orthogonal to the the pair state that is macroscopically occupied in the ground state - 1s hydrogenic pair states in the Coulomb interaction case. We have computed the momentum space eigenvectors for the Higgs collective mode and compared them with the analytically known 2D hydrogenic excited state solutions. In this way we have found that the large weight amplitude response correspond to the addition of an electron-hole pair to the system, not in the the 1s pair state which is condensed, but in a higher energy orbitals. The lowest energy high weight state in the BEC limit at $Q = 0$ corresponds to adding an electron-hole pair in a 2s state, which in 2D has a binding energy relative to the particle-hole continuum that is smaller by a factor of nine. (The second highest weight state corresponds to the 3s state and higher $n$ excitations are not fully identifiable only because of the finite density of the momentum space grids used in our calculations.) In a SIXC, therefore, the gapped Higgs modes are excitations in which one electron-hole pair is added in a state that is orthogonal to the pair-state present in the condensate. As we see in Figs 2(c), at finite $\vec{Q}$, the Goldstone mode also makes a nonzero contribution to the pair-amplitude response. The 2s-like Higgs mode still has very large weight in the spectra and is located below the electron-hole continuum even in the large wave-vector $Q a_B^2 = 1$ case. In the low exciton density limit, the wavevector dependence of the Goldstone collective mode is consistent with the Bogoliubov theory of weakly interacting bosons. On the other hand, the 2s-like Higgs mode has a dispersion vs. $\vec{Q}$ that can be fit using a quadratic function as shown in Fig. 3.

As the BCS regime at large exciton densities is ap-
proached, the spectra of the Higgs modes change. In Fig. 2(b) and (d), we find that different collective modes have larger weight in the response function as wave-vector increases. At zero wave-vector, we identify the first Higgs mode shown in Fig. 2(b) and find that its qualitative interpretation as the excitation of a non-condensed pair is unchanged. As \( \vec{Q} \) varies, another peak belonging to a different excitation away from the particle-hole continuum appears, and the new Higgs mode eventually has larger weight at the large \( \vec{Q} \) illustrated in Fig. 2(d). We plot the frequencies of the two competing modes with large weight in the \( \tau_x - \tau_1 \) response function as a function of wave-vector in Fig. 3. We can see that the first Higgs mode enters the continuum and has its weight spread over a broader range of energies at large \( \vec{Q} \), but the second Higgs mode always lies below the continuum. We nevertheless find that even in the BCS regime the SIXC supports Higgs modes below the particle hole continuum. This behavior contrast with the case of the BCS models commonly used for superconductors in which Higgs modes are located exactly at the edge of the particle-hole continuum \( 2\Delta \). The Higgs modes here are distinct modes, higher in energy than the Goldstone modes but still in the excitation gap. This source of the difference is the nature of long-range of the Coulomb interaction between electrons and holes. If we replace our interlayer Coulomb potential with a BCS-like \( \delta \)-type attractive potential, we obtain Higgs modes sitting at the particle-hole continuum edge, in agreement with earlier work.

In contrast to the different behaviors of Higgs modes in BEC and BCS limits, Goldstone modes always contribute most to \( \chi_{zz} \) response at any \( Q \) (except \( Q = 0 \)) shown as blue bars in Fig. 2.

IV. DISCUSSION

In this manuscript we have applied time-dependent mean-field theory to spatially-indirect exciton condensates with the goal of identifying collective modes associated with quantum fluctuations in the electron-hole pair amplitude. We find that in the low-exciton density BEC regime the strongest response to Higgs-like perturbations are ones in which an electron-hole pair is added in a state that is orthogonal to the pair state present in the ground state condensate. This interpretation retains qualitative validity when the exciton density is increased and the BCS limit is approached. These findings shed new light on previous work that has studied Higgs modes in superconductors, in which the Higgs response appears, mysteriously perhaps, at the edge of the particle-hole continuum. In the light of the present calculations it is clear that this property just reflects the absence in the BCS models used for these studies of a higher energy electron-electron pair bound state, and begs the question as to whether or not higher energy bound states do exist in some superconductors. Since Higgs excitations in superconductors change the total electron number, they can be observed only indirectly. One possible strategy to detect these higher energy bound states where they are suspected is to look for resonant features in the bias voltage dependent subgap currents of Josephson junctions.

Two different cases need to be distinguished in discussing the detection of Higgs modes. When a spatially indirect exciton condensate is formed from equilibrium populations of electrons and holes in two-separate layers, the operator \( \tau_x \) corresponds to tunneling between layers. The presence of a spatially indirect exciton condensate, or incipient condensate, then appears as an anomaly in the interlayer tunneling current-voltage relationship near zero bias. We anticipate that Higgs modes will appear as finite-bias voltage anomalies at energies below the particle-hole continuum.

The case in which an exciton condensate is formed in a quasi-equilibrium systems of electrons and holes, either in the same layer or in adjacent layers, generated by optical pumping is perhaps simpler experimentally. In this case the coherent excitons are routinely examined by measuring the photo-luminescence (PL) signal. Emitted photons with energy \( \hbar \omega \) can be generated by transitions between initial \( N \)-exciton states and final \( N - 1 \) exciton states which satisfy

\[
\hbar \omega = E_i(N) - E_j(N - 1) = \mu_{ex} + (E_i(N) - E_0(N)) - (E_j(N - 1) - E_0(N - 1)).
\]

(37)

The matrix elements for these processes are proportional to the operator \( \tau_x \), which change the number of electron-hole pairs present in the system by one, and can therefore generate Higgs excitations. In Eq. 37, \( \mu_{ex} \) is the chemical potential of excitons which is non-zero in non-equilibrium condensed exciton systems, \( E_0(N) = E_i(N) - E_0(N) \) is the excitation relative to the ground state in the \( N \)-exciton initial state and \( E_0(N - 1) = E_j(N - 1) - E_0(N - 1) \) is the excitation energy relative to ground in the \( N - 1 \) exciton final state. A similar analysis applies in the case of polariton-condensates in which the exciton system is coupled to two-dimensional cavity photon. The PL spectrum consists a segment for which \( \hbar \omega > \mu_{ex} \) due to thermal excitations in the initial state and a so-called ghost segment in which \( \hbar \omega < \mu_{ex} \) due to excitations being generated in the final state when the exciton number changes. Because they have a high energy Higgs modes are not likely to be thermally populated,
but they can be visible in the ghost mode spectrum when exciton-exciton interactions are strong. Indeed very recent work\textsuperscript{52} which appeared as this paper was under preparation has claimed that a Higgs excitation is present in the PL spectrum of a polariton condensate at energy $\hbar \omega = \mu_{ex} - E_{Higgs}$, and has made the numerical observation that $E_{Higgs}$ is close to the energy difference between the cavity-dressed $2\sigma$ and $1\sigma$ excitonic bound states. The present paper appears to explain this observation.

V. ACKNOWLEDGMENT

This work was supported by the Army Research Office under Award No. W911NF-17-1-0312 (MURI) and by the Welch Foundation under Grant No. F1473. F.X. acknowledges support under the Cooperative Research Agreement between the University of Maryland and the National Institute of Standards and Technology Physical Measurement Laboratory, Award 70NANB14H209, through the University of Maryland. F. W. is supported by the Laboratory for Physical Sciences.

Appendix A: explicit expressions for $\mathcal{E}_{k,p}(\bar{Q})$ and $\Gamma_{k,p}(\bar{Q})$

Below are explicit expressions for $\mathcal{E}_{k,p}(\bar{Q})$ and $\Gamma_{k,p}(\bar{Q})$, which appear in the energy variation $\delta E^{(2)}$ in Eq. (14).

\begin{align}
\mathcal{E}_{k,p}(\bar{Q}) &= \delta_{k,p}(\zeta \bar{Q} - \zeta + E_{k} + E_{\bar{Q}}) \\
&+ \frac{1}{A} [V(\bar{Q}) - V(\bar{k} - \bar{p})] (u_k v_p \bar{u}_k \bar{v}_p + u_k v_p \bar{u}_k \bar{v}_p + u_k v_p \bar{u}_k \bar{v}_p) \\
&- \frac{1}{A} U(\bar{Q})(u_k v_p \bar{u}_k \bar{v}_p + u_k v_p \bar{u}_k \bar{v}_p + u_k v_p \bar{u}_k \bar{v}_p) \\
&- \frac{1}{A} U(\bar{k} - \bar{p})(u_k v_p \bar{u}_k \bar{v}_p + u_k v_p \bar{u}_k \bar{v}_p + u_k v_p \bar{u}_k \bar{v}_p),
\end{align}

where $v_k$ and $u_k$ are defined in Eq. (6), and $V(\bar{Q})$ and $U(\bar{Q})$ are respectively intralayer and interlayer Coulomb interactions.

1. G. Aad, T. Abajyan, B. Abbott, J. Abdallah, S. A. Khalek, A. Abdelalim, O. Abidinov, R. Aben, B. Abi, M. Abolins, et al., Physics Letters B \textbf{716}, 1 (2012).
2. S. Chatrchyan, V. Khachatryan, A. M. Sirunyan, A. Tumasyan, W. Adam, E. Aguilo, T. Bergauer, M. Dragicevic, J. Erö, C. Fabjan, et al., Physics Letters B \textbf{716}, 30 (2012).
3. D. Pekker and C. Varma, Annu. Rev. Condens. Matter Phys. \textbf{6}, 269 (2015).
4. P. W. Anderson, Phys. Rev. \textbf{110}, 827 (1958).
5. P. W. Anderson, Phys. Rev. \textbf{130}, 439 (1963).
6. F. Englert and R. Brout, Phys. Rev. Lett. \textbf{13}, 321 (1964).
7. P. W. Higgs, Phys. Rev. Lett. \textbf{13}, 508 (1964).
8. G. S. Guralnik, C. R. Hagen, and T. W. B. Kibble, Phys. Rev. Lett. \textbf{13}, 585 (1964).
9. R. Sooryakumar and M. V. Klein, Phys. Rev. Lett. \textbf{45}, 660 (1980).
10. P. B. Littlewood and C. M. Varma, Phys. Rev. B \textbf{26}, 4883 (1982).
11. D. Podolsky, A. Auerbach, and D. P. Arovas, Phys. Rev. B \textbf{84}, 174522 (2011).
12. Y. Barlas and C. M. Varma, Phys. Rev. B \textbf{87}, 054503 (2013).
13. R. Matsunaga, Y. I. Hamada, K. Makise, Y. Uzawa, H. Terai, Z. Wang, and R. Shimano, Phys. Rev. Lett. \textbf{111}, 057002 (2013).
14. R. Matsunaga, N. Tsuji, H. Fujita, A. Sugio, K. Makise, Y. Uzawa, H. Terai, Z. Wang, H. Aoki, and R. Shimano, Science \textbf{345}, 1145 (2014).
15. http://science.sciencemag.org/content/345/6201/1145.full.pdf
M.-A. Méasson, Y. Gallais, M. Cazayous, B. Clair, P. Rodière, L. Cario, and A. Sacuto, Phys. Rev. B 89, 060503 (2014).

G. E. Volovik and M. A. Zubkov, Journal of Low Temperature Physics 175, 486 (2014).

D. Sherman, U. S. Pracht, B. Gorshunov, S. Poran, J. Jesudasan, M. Chand, P. Raychaudhuri, M. Swanson, N. Trivedi, A. Auerbach, et al., Nat. Phys. 11, 188 (2015).

C. Rüegg, B. Normand, M. Matsumoto, A. Furrer, D. F. McMorrow, K. W. Krämer, H. U. Güdel, S. N. Gvasaliya, H. Mutka, and M. Boehm, Phys. Rev. Lett. 100, 205701 (2008).

M. Endres, T. Fukuhara, D. Pekker, M. Cheneau, P. Schauß, C. Gross, E. Demler, S. Kuhr, and I. Bloch, Nature 487, 454 (2012).

P. Merchant, B. Normand, K. Krämer, M. Boehm, D. McMorrow, and C. Rüegg, Nature physics 10, 188 (2015).

C. Rüegg, B. Normand, M. Matsumoto, A. Furrer, D. F. McMorrow, K. W. Krämer, H. U. Güdel, S. N. Gvasaliya, H. Mutka, and M. Boehm, Phys. Rev. Lett. 100, 205701 (2008).

M. Greiner, C. A. Regal, and D. S. Jin, Nature 426, 537 (2003).

C. A. Regal, M. Greiner, and D. S. Jin, Phys. Rev. Lett. 92, 040403 (2004).

J. Léonard, A. Morales, P. Zupancic, T. Donner, and T. Esslinger, Science 358, 1415 (2017), http://science.sciencemag.org/content/358/6369/1415.full.pdf.

L. V. Keldysh and Y. V. Kopaev, Sov. Phys. Solid State 6, 2219 (1965).

Y. E. Lozovik and V. I. Yudson, Sov. Phys. JETP 44, 389 (1976).

C. Comte and P. Nozieres, J. Phys. (Paris) 43, 1069 (1982).

X. Zhu, P. B. Littlewood, M. S. Hybertsen, and T. M. Rice, Phys. Rev. Lett. 74, 1633 (1995).

J. P. Eisenstein and A. H. MacDonald, Nature 432, 691 (2004).

D. Nandi, A. Finck, J. Eisenstein, L. Pfeiffer, and K. West, Nature 488, 481 (2012).

X. Liu, K. Watanabe, T. Taniguchi, B. I. Halperin, and P. Kim, Nat. Phys. 13, 746 (2017).

J. Li, T. Taniguchi, K. Watanabe, J. Hone, and C. Dean, Nat. Phys. 13, 751 (2017).

G. W. Burg, N. Prasad, K. Kim, T. Taniguchi, K. Watanabe, A. H. MacDonald, L. F. Register, and E. Tutuc, Phys. Rev. Lett. 120, 177702 (2018).

Z. Wang, D. A. Rhodes, K. Watanabe, T. Taniguchi, J. C. Hone, J. Shan, and K. F. Mak, Nature 574, 76 (2019).

J.-J. Su and A. H. MacDonald, Nature Phys. 4, 799 (2008).