A Many-Body Theory of the Optical Conductivity of Excitons and Trions in Two-Dimensional Materials

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The optical spectra of two dimensional (2D) materials exhibit sharp absorption peaks that are commonly identified with excitons and trions (or charged excitons). In this paper, we show that excitons and trions in doped 2D materials can be described by two coupled Schrödinger-like equations - one two-body equation for excitons and another four-body equation for trions. In electron-doped 2D materials, a bound trion state is identified with a four-body bound state of an exciton and an excited conduction band electron-hole pair. In doped 2D materials, the exciton and the trions states are not the eigenstates of the full Hamiltonian and their respective Schrödinger equations are coupled due to Coulomb interactions. The strength of this coupling increases with the doping density. Solutions of these two coupled equations can quantitatively explain all the prominent features experimentally observed in the optical absorption spectra of 2D materials including the observation of two prominent absorption peaks and the variation of their energy splittings and spectral shapes and strengths with the electron density. The optical conductivity obtained in our work satisfies the optical conductivity sum rule exactly. A superposition of exciton and trion states can be used to construct a solution of the two coupled Schrödinger equations and this solution resembles the variational exciton-polaron state, thereby establishing the relationship between our approach and Fermi polaron physics.

I. INTRODUCTION

Optical absorption and emission spectra of two-dimensional (2D) materials, most notably transition metal dichalcogenides (TMDs), exhibit distinct peaks that are attributed to neutral and charged excitons (or trions). Trions have been discussed extensively in the literature. In electron-doped materials, a trion state has been described in many different ways, i) as a bound state of two conduction band (CB) electrons and a valence band (VB) hole, or an electron bound to an exciton, ii) as a bound state of two CB electrons and a VB valence band hole, plus a CB hole, and iii) as an exciton-polaron (an exciton that is screened by CB electrons). The relationship between these different pictures is not clear. The trion state considered by Combescot et al., which consisted of a bound state of two CB electrons and a VB hole, where deemed to have negligible optical matrix element with the ground state by Sidler et al. The variational trion state used previously by the authors, which consisted of a bound state of two CB electrons and one VB hole, plus a CB hole, reproduced the measured trion optical absorption spectra in 2D transition metal dichalcogenides (TMDs) with fairly good accuracy in the low electron density limit but it could not explain the splitting of the trion and exciton absorption peaks as a function of the electron density, nor could it explain the transfer of the spectral weight in the optical absorption spectra from the exciton to the trion with the increase in the electron density. Diagrammatic perturbation theory involving summation of ladder diagrams corresponding to exciton-electron interactions as well as variational ansatz have been used to describe excitons interacting with electrons in electron-doped semiconductors. The solutions correspond to states that describe screening of the exciton by the electrons, or what are also called exciton-polarons. Exciton-polaron solutions have been successful in capturing the variations of the of the energy splittings as well as the spectral weight transfers observed in the optical absorption spectra as the electron density is varied. On the other hand, the three-body trion physics has been fairly successful in predicting the experimentally observed exciton-trion splittings (or the trion binding energies) in the limit of vanishing electron density. The connection, if any, between the exciton-polaron picture and the picture of a trion as a bound three-body or four-body state remains unclear.

FIG. 1: Bound exciton (left) and bound trion (right) states in an n-doped 2D material are depicted. A bound trion is a 4-body state and consists of two conduction band (CB) electrons, a valence band (VB) hole, and a conduction band (CB) hole. When the background electron density is non-zero, these two states are coupled due to electron-electron and electron-hole interactions.

In this paper, we use the many-body density matrix technique to describe describe excitons in electron doped two-dimensional materials. Our main results are summarized below.

Excitons and trions in doped materials can be described by two coupled Schrödinger-like equations. One is the standard two-body Schrödinger equation of a CB elec-
tron interacting with a VB hole, and the other is a four-body Schrödinger equation of two CB electrons interacting with a VB hole and a CB hole (See Fig. 1). A trimmed version of the latter, obtained by dropping the CB hole, is the standard three-body Schrödinger equation that has previously been used to describe trions. The CB hole is created when an electron is scattered out of the Fermi sea. These two equations are coupled via Coulomb matrix elements that result from electron-electron and electron-hole interactions. These coupled equations emerge naturally from the density matrix approach. Within the purview of an exciton exciting only single electron-hole pair at a time in the conduction band, the solution of these two coupled equations provide a simple but exact description of the exciton interacting with CB electrons. Solutions of these two coupled equations can quantitatively explain all the prominent features experimentally observed in the optical absorption spectra of 2D materials including the observation of two prominent absorption peaks and the variation of their energy splittings, spectral strengths, and spectral linewidths with the electron density. Interestingly, the exact solution of these two equations resembles the variational exciton-polaron solution thereby establishing the relationship between the two approaches. The solutions obtained in this work also explicitly satisfy the optical conductivity sum rule exactly at all carrier densities.

Our numerical solutions for 2D TMD materials show that the four-body Schrödinger equation has a bound state (which we will call a trion and which may be taken as a bound state of an exciton and a CB electron-hole pair) whose binding energy (defined with respect to the exciton energy given by the two-body Schrödinger equation) is larger than the Fermi energy even for electron densities approaching \( \sim 10^{13} \) cm\(^{-2}\). However, these bound states of the four-body Schrödinger equation and the bound states of the two-body Schrödinger equation are not eigenstates of the full Hamiltonian the presence of a non-zero electron density. It is energetically favorable for the exciton to release the bound CB electron-hole pair and excite another CB electron-hole pair and bind it (or scatter from it). Furthermore, this CB electron-hole pair exchange process can occur at the lowest electron densities, even when the Fermi energy is much smaller than the binding energy of the four-body state, and increases in frequency as the electron density increases. The energy thus gained is responsible for the observed near-linear increase in the exciton-trion energy difference with the Fermi energy in the optical spectra. The essential physics at non-zero electron densities is therefore that of coupling between the excitons and the trions via Coulomb interactions. The picture described here also captures the essential physics associated with the exciton-polarons. The main two peaks observed experimentally in the optical absorption spectra result from the coupling between the two-body exciton and four-body trion states and have been identified as the repulsive and attractive exciton-polarons. A complete and accurate description of the problem requires using both the bound and the unbound states of the four-body Schrödinger equation.

In this paper, we have focused on single electron-hole pair excitation by an exciton and have ignored multiple electron-hole pair excitations for the same reasons as discussed by Efimkin et al. Effects related to Fermi edge singularities which involve multiple electron-hole pair excitations, are therefore also ignored. Our work shows that a description based on single pair excitations can adequately explain the prominent features of the experimentally measured optical absorption spectra of 2D materials.

In the light of the introductory discussion above, the name "trion" seems like a misnomer since the involved states are either two-body or four-body states. But given the long history of the use of this terminology, in this paper we will use the term “trion” for the eigenstates of the four-body Schrödinger equation (Fig. 1). After this work had been completed, we became aware of the earlier work by Suris et al. in which mixed exciton-trion modes had been introduced to account for the coupling between the excitons and the trions resulting from Coulomb intercations. The work presented here is conceptually along the same lines, but our approach is different in many ways. The use of coupled two-body and four-body Schrödinger equations enables one to include the effects of the electron density on the wavefunctions and the binding energies. Our approach also does not require solution of Bethe-Salpeter-like equations with accompanying complexities associated with divergent integrals, artificial cut-offs of momentum integrals, and wavevector-independent Coulomb potentials, and enables us to obtain quantitative results for excitons and trions in two dimensional materials. In contrast to previous works, our approach also explicitly takes into account electron’s spin and valley degrees of freedom and exchange interactions.

II. THEORETICAL MODEL

In this Section we set up the Hamiltonian and derive the main equations. Although the focus is on electron-doped 2D TMD materials, the arguments are kept general enough to be applicable to any 2D material.

A. Hamiltonian

We consider a 2D TMD monolayer at \( z = 0 \). Light with a small in-plane momentum \( \vec{Q} \) is incident on the layer. The Hamiltonian describing electrons and holes in the TMD layer (near the \( K \) and \( K' \) points in the Brillouin zone) interacting with each other and with the optical
mode in the rotating wave approximation \[ H = \sum_{\vec{k},s} E_{c,s}(\vec{k}) c_s^\dagger(\vec{k}) c_s(\vec{k}) + \sum_{\vec{k},s} E_{v,s}(\vec{k}) b_s^\dagger(\vec{k}) b_s(\vec{k}) \\
+ \frac{1}{A} \sum_{\vec{q},\vec{k},\vec{k}',s,s'} U(q) c_s^\dagger(\vec{k} + \vec{q}) b_{s'}(\vec{k}') c_s(\vec{k}) \\
+ \frac{1}{2A} \sum_{\vec{q},\vec{k},\vec{k}',s,s'} V(q) c_s^\dagger(\vec{k} + \vec{q}) c_{s'}(\vec{k}') - \vec{q} c_{s'}(\vec{k}') c_s(\vec{k}) \\
+ \hbar \omega(\vec{Q}) a^\dagger(\vec{Q}) a(\vec{Q}) \\
+ \frac{1}{\sqrt{A}} \sum_{k,s} \left( g_s c_s^\dagger(\vec{k} + \vec{Q}) b_s(\vec{k}) a(\vec{Q}) + h.c \right) \] (1)

Here, \( E_{c,s}(\vec{k}) \) and \( E_{v,s}(\vec{k}) \) are the conduction and valence band energies, \( s, s' \) represent the spin/valley degrees of freedom in the 2D material, and we assume for simplicity that the electron and hole effective masses are independent of the spin/valley. \( U(q) \) represents Coulomb interaction between electrons in the conduction and valence bands and \( V(q) \) represents Coulomb interaction among the electrons in the conduction bands. \( \hbar \omega(\vec{Q}) \) is the energy of a photon with in-plane momentum \( \vec{Q} \), and \( g_s \) is the electron-photon coupling constant. Other than for phase factors that are not relevant in the discussion that follows, \( g_s \) for electron states near the band edges in 2D TMDs can be given by
\[ g_s = e v \sqrt{\frac{\hbar}{2(c)\omega(\vec{Q})}} \chi(z = 0) \] (2)

where, \( v \) is the interband velocity matrix element and \( \chi(z) \) describes the amplitude of the optical mode in the z-direction.

### B. Density Matrix Approach

We use a many body density matrix approach which has been fairly successful in modeling exciton physics, and it has also been previously used for trions in the limit of vanishingly small electron densities. We start from the Heisenberg equation for the photon operator, which after averaging, \[ \left[ \hbar \omega(\vec{Q}) + i\hbar \frac{\partial}{\partial t} \right] \langle a^\dagger(\vec{Q}, t) \rangle = -\frac{1}{\sqrt{A}} \sum_{k,s} g_s P_{\vec{Q}}(\vec{k}, s; t) \] (3)

The polarization \( P_{\vec{Q}}(\vec{k}, s; t) \) equals the equal-time two-body correlation \( \langle c_s^\dagger(\vec{k} + \vec{Q}, t) b_s(\vec{k}, t) \rangle \). Assuming from now onwards that in steady state all the relevant equal-time correlation functions have the time dependence \( e^{\pm i\omega t} \), the above equation becomes, \[ \left[ \hbar \omega(\vec{Q}) + i\epsilon - \hbar \omega \right] \langle a^\dagger(\vec{Q}) \rangle = -\frac{1}{\sqrt{A}} \sum_{k,s} g_s P_{\vec{Q}}(\vec{k}, s) \] (4)

where, \( P_{\vec{Q}}(\vec{k}, s; t) = P_{\vec{Q}}(\vec{k}, s) e^{i\omega t} \). The Heisenberg equation for \( P_{\vec{Q}}(\vec{k}, s) \) is,
\[ E_{c,s}(\vec{k} + \vec{Q}) - E_{v,s}(\vec{k}) + i\gamma_{ex} - \hbar \omega \] \( P_{\vec{Q}}(\vec{k}, s) = \)
\[ -\frac{1}{\sqrt{A}} g_s \langle a^\dagger(\vec{Q}) \rangle \left[ 1 - f_{c,s}(\vec{k} + \vec{Q}) \right] \]
\[ + \frac{1}{A} \sum_{\vec{q}} U(\vec{q}) P_{\vec{Q}}(\vec{k} + \vec{q}, s) \left[ 1 - f_{c,s}(\vec{k} + \vec{Q}) \right] \]
\[ -\frac{1}{A} \sum_{\vec{q},\vec{p},s'} U(\vec{q}) \]
\[ \times T_{\vec{Q}}^c(\vec{k} + (\xi + \eta)\vec{Q} - \xi\vec{p}, s; (\xi + \eta)\vec{p} - \xi\vec{Q} - \vec{q}, s'; \vec{p}, s') \]
\[ + \frac{1}{A} \sum_{\vec{q},\vec{p},s'} T_{\vec{Q}}^c(\vec{k} + (\xi + \eta)\vec{Q} - \xi\vec{p} + \vec{q}, s; (\xi + \eta)\vec{p} - \xi\vec{Q} - \vec{q}, s'; \vec{p}, s') \] (5)

Here, \( \langle c_s^\dagger(\vec{k}) c_s(\vec{k}) \rangle = f_{c,s}(\vec{k}) \) is the electron occupation probability, \( \gamma_{ex} \) is a phenomenological decoherence rate for the polarization that includes dephasing due to all processes other than exciton-electron scattering. The energies \( E_{c,v,s}(\vec{k}) \) include renormalizations due to exchange at the Hartree-Fock level. \( \lambda_\alpha = 1 - \lambda_e = m_h/m_{ex} \) (\( m_e = m_h \)), where \( m_e \) (\( m_h \)) is the electron (hole) effective mass. Without the first term and the last two terms on the RHS, is the standard eigenvalue equation for excitons, and has eigenvalues \( E_{\vec{Q}}^e(\vec{Q}, s) \) and eigenfunctions \( \phi_{\vec{Q}}(\vec{k} + \lambda_\alpha \vec{Q}, s) \). It is not Hermitian but it can be converted into a Hermitian equation (see Appendix A). The eigenfunctions therefore form a complete set.

The last two terms in [5] on the RHS contain four-body correlations \( T_{\vec{Q}}^c \) and correspond to the diagrams shown in Fig. 2(a,b). Assuming that \( m_{tr} = 2m_e + m_h \), \( \xi = m_e/m_{tr} \), and \( \eta = m_h/m_{tr} \), we define a four-body
equal-time correlation $T_G(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2; t)$ as follows,
\begin{align}
(c_{s_1}^\dagger(\vec{k}_1, t)c_{s_2}(\vec{k}_2, t)b_{s_1}(\vec{k}_1 + \vec{k}_2 - (\vec{Q} + \vec{p}), t)c_{s_2}(\vec{p}, t))
\end{align}
(6)

The underlined vector $\vec{k}$ stands for $\vec{k} + \xi(\vec{Q} + \vec{p})$. $T_G$ describes the correlations arising from Coulomb interactions among four particles: two CB electrons, a VB hole, and a CB hole. $\vec{Q}$ is the total momentum of this 4-body state. We also define a fully connected four-body correlation (as defined in the linked cluster expansion technique\cite{22}),
\begin{align}
T^c_G(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2; t) = T^c_G(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2; t) \\
- f_{c,s_2}(\vec{p})P_G(\vec{k}_1 - \vec{Q}, s_1)\delta_{\vec{k}_2 - \vec{q}, s_2} \\
+ f_{c,s_2}(\vec{p})P_G(\vec{k}_2 - \vec{Q})\delta_{\vec{s}_1,s_2}\delta_{\vec{p},s_2}
\end{align}
(7)
The equation for the connected correlation $T^c_G(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2)$ is found to be,
\begin{align}
E_{c,s_1}(\vec{k}_1) + E_{c,s_2}(\vec{k}_2) - E_{c,s_1}(\vec{k}_1 + \vec{k}_2 - (\vec{Q} + \vec{p})) \\
- 1/2 \sum_{\vec{q}} V(\vec{q})T^c_G(\vec{k}_1 + \vec{q}, s_1; \vec{k}_2 - \vec{q}, s_2; \vec{p}, s_2) \\
\times [1 - f_{c,s_1}(\vec{k}_1) - f_{c,s_2}(\vec{k}_2)] \\
+ 1/2 \sum_{\vec{q}} U(\vec{q})T^c_G(\vec{k}_1, s_1; \vec{k}_2 - \vec{q}, s_2; \vec{p}, s_2) [1 - f_{c,s_1}(\vec{k}_1)] \\
+ 1/2 \sum_{\vec{q}} U(\vec{q})T^c_G(\vec{k}_1, s_1; \vec{k}_2 - \vec{q}, s_2; \vec{p}, s_2) [1 - f_{c,s_2}(\vec{k}_2)] \\
+ 1/2 \sum_{\vec{q}} V(\vec{q})T^c_G(\vec{k}_1 + (\xi + \eta)\vec{q}, s_1; \vec{k}_2 - \xi\vec{q}, s_2; \vec{p} + \vec{q}, s_2) \\
\times [f_{c,s_2}(\vec{p}) - f_{c,s_2}(\vec{k}_1)] \\
+ 1/2 \sum_{\vec{q}} V(\vec{q})T^c_G(\vec{k}_1 - \xi\vec{q}, s_1; \vec{k}_2 + (\xi + \eta)\vec{q}, s_2; \vec{p} + \vec{q}, s_2) \\
\times [f_{c,s_2}(\vec{p}) - f_{c,s_2}(\vec{k}_2)] \\
- 1/2 \sum_{\vec{q}} U(\vec{q})T^c_G(\vec{k}_1 - \xi\vec{q}, s_1; \vec{k}_2 - \xi\vec{q}, s_2; \vec{p} + \vec{q}, s_2) f_{c,s_2}(\vec{p}) \\
+ f_{c,s_2}(\vec{p}) \sum_{\vec{q}} U(\vec{q}) [1 - f_{c,s_1}(\vec{k}_1) - f_{c,s_2}(\vec{k}_2)] \\
\times \left[P_G(\vec{k}_1 - \vec{Q} + \vec{q}, s_1)\delta_{\vec{k}_2 - \vec{q},s} - P_G(\vec{k}_2 - \vec{Q} - \vec{q}, s_2)\delta_{\vec{k}_1 + \vec{q},s} - f_{c,s_2}(\vec{p}) \sum_{\vec{q}} U(\vec{q}) \left[P_G(\vec{k}_1 - \vec{Q} + \vec{q}, s_1)\delta_{\vec{k}_2 - \vec{q},s} [1 - f_{c,s_1}(\vec{k}_1)] \right] \\
- P_G(\vec{k}_2 - \vec{Q}, s_2)\delta_{\vec{k}_1 + \vec{q},s} f_{c,s_1}(\vec{k}_1) \right]
\end{align}

In deriving the above equation, all six-body correlations are reduced to four-body correlations using the cluster expansion\cite{22}. By ignoring higher order correlations we are ignoring the generation of multiple particle-hole pairs in the CB. Here, $\gamma_t$ is a phenomenological decoherence rate. If \( \vec{r}_{e_1}, \vec{r}_{e_2}, \vec{r}_{h_1} \) are \( \vec{r}_{h_2} \) the coordinates of the two electrons, the VB hole, and the CB hole, respectively, then $\vec{k}_1$, $\vec{k}_2$, $\vec{Q}$, and $\vec{p}$ are the momenta associated with the coordinates $\vec{r}_{e_1} - \vec{r}_{h_1}$, $\vec{r}_{e_2} - \vec{r}_{h_1}$, $\vec{R} = \xi(\vec{r}_{e_1} + \vec{r}_{e_2}) + \eta\vec{r}_{h_1}$, and $\vec{R} - \vec{r}_{h_2}$, respectively. Here, $\vec{R}$ is the center of mass coordinate of the two electrons and the VB hole. Ignoring the last two terms on the RHS in 6 that involve $P_G$, Fourier transform of the remaining terms will result in a four-body Schrödinger equation. Each term on the RHS in the above equation (except the last two) describes Coulomb interaction between two of the four particles. The last two terms involving $P_G$ capture the generation of four-body correlation from two-body correlation, or the creation of an CB electron-hole pair by an exciton, and correspond to the diagrams shown in Figs. 3(c, d). A similar equation was obtained by Esser et al.\cite{23}, but in that work the connected nature of $T^c_G$ was overlooked, the terms containing interactions with the CB hole were ignored, the phase-space restricting factors were ignored too, and the terms containing the polarization $P_G$ were also missed. 3(c) and 3(d) are a closed system of coupled Schrödinger equations for two-body and four-body systems.

C. Trion States

The trion states are defined here as the eigenstates of the four-body Schrödinger equation given in 6 and written as $\phi_{m,G}^{tr}(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2)$. The corresponding eigenenergies are $E_{m,G}^{tr}(\vec{Q}, s_1, s_2)$. 8 is not Hermitian but it can be converted into a Hermitian equation (see Appendix B), with a few suitable approximations, and the eigenfunctions therefore form a complete set. The eigenstates include bound four-body states, unbound exciton-electron scattering states, and completely unbound four-body states. The eigenfunctions $\phi_{m,G}^{tr}(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2)$ are either symmetric or antisymmetric in $\vec{k}_1$ and $\vec{k}_2$ depending on the values of $s_1$ and $s_2$ and on the spin state of the two electrons (singlet or triplet).

D. The Fate of the Conduction Band Hole

The 4th, 5th, and 6th terms on the RHS in the four-body Schrödinger equation include interactions involving the CB hole that is generated when an electron is scattered out of the Fermi sea by the exciton. This CB hole can have a minimum radius of the order of $1/k_F$ in real space, which can be much larger than the exciton and (8) trion radii at low electron densities. For this reason, in-
teractions involving the CB hole have been ignored in previous works. Here we argue that the CB hole needs to be taken into account in bound trion states. The LHS of (5) has the energies of the photoexcited electron, the VB hole, and the initial and final energies of the electron scattered out of the Fermi sea. All energies include renormalization due to exchange. An electron within the Fermi sea has a much larger energy renormalization than an electron well outside the Fermi sea. Consequently, when an electron is scattered out of the Fermi sea to bind to an exciton and form a trion state, the difference in its energy exciton state and the higher energy exciton states in 2D materials can be in the hundreds of milli electron volts range.

The exciton self-energy can be expressed as,

$$\Sigma_{n,s}(Q,\omega) = \sum_{m,n} \left[ 1 + \delta_{s,s'} \right] M_{m,n}(Q, s, s')^2$$

(12)

where $M_{m,n}(Q, s, s')$ equals,

$$\frac{1}{A^3} \sum_{k,p,q} \phi^{r*}_{m,n,Q}(k - \xi(p + Q), s; (\xi + \eta)p - \xi Q + q, s'; p', s)$$

$$\times \sqrt{1 - f_{c,s}(\vec{k})} \left[ 1 - f_{c,s}(\vec{p}) \right] \sqrt{1 - f_{c,s}(\vec{p} + q)}$$

$$\times \left\{ \begin{array}{l}
V(q) P_Q(\vec{k}) - U(\vec{q}) P_Q(\vec{k} - \vec{Q}, s) - U(\vec{q}) \phi^{ex}_{n,Q}(k - \lambda_e \vec{Q}, s) \\
\end{array} \right\}$$

(13)

It is evident from the above expression that the coupling term $M_{m,n}(Q, s, s')$ increases with the electron density. The self-energy expression in (12) assumes that an exciton state does not couple to a different exciton state due to Coulomb interactions via an intermediate trion state. Given that the energy separation between the lowest energy exciton state and the higher energy exciton states in 2D materials can be in the hundreds of milli electron volts range, the approximation made in this assumption is expected to be very good for the lowest energy exciton state.

The expressions in (11) and (12) constitute the main results of this work. The optical conductivity in (11) corresponds to the diagrams and processes shown in Fig. 5. Only fully connected diagrams contribute to the optical conductivity. Diagrams with multiple particle-hole excitations at the same time are not included in this work.
Here, \( m_T = m_{ex}m_e/(m_{ex} + m_e) \), is the reduced mass of the exciton and the electron. Use of the state in (14) to evaluate exciton-electron scattering contributions to the self-energy in (12) is equivalent to the use of the Born approximation in exciton-electron scattering.\(^{23}\) Note that electrons with both

G. Optical Conductivity Sum Rule

This optical conductivity sum rule for 2D TMDs can be derived from the restricted Thomas-Reiche-Kuhn optical conductivity sum rule\(^{25}\) and, assuming a full valence band, can be expressed as\(^{25}\)

\[
\int_0^\infty \omega \text{Re}\{\sigma_\omega(\omega)\} \, d\omega = \frac{e^2 v^2}{2h} \sum_s \int \frac{d^2 k}{(2\pi)^2} \left( 1 - f_{c,s}(\mathbf{k}) \right)
\]

Here, \( v \) is the interband velocity matrix element between the valence and conduction band Bloch states (see Sec. II A). Band filling is incorporated into the above sum rule. The completeness of the exciton eigenfunctions \( \phi_{n,Q}^{ex}(\mathbf{k}, s) \) can be used to see that the derived optically conductivity in (11) satisfies the above sum rule exactly.

III. VARIATIONAL EIGENSTATES AND CONNECTION WITH EXCITON-POLARON STATES

Variational states for exciton-polarons have been constructed in previous works\(^{21,22}\). These variational states resemble the Fermi polaron states of an impurity atom in a cold Fermi gas\(^{27,28}\). Here we show that variational states can be constructed using the eigenstates of the two-body and four-body Schrödinger equations in (3) and (8), respectively, and which give results for the eigenenergies in exact agreement with the exciton self-energy given in (12). The eigenenergies can be obtained from the poles of the exciton Green’s function and these energies are the roots of the equation,

\[
\hbar \omega - E_{ex}^{\text{ex}}(\mathbf{Q}, s) + i \gamma_{ex} - \sum_{n,s} \phi_{n,Q}^{ex}(\mathbf{Q}, s) = 0
\]

where the exciton self-energy is as given in (12). Since the two-body and four-body Schrödinger equations are coupled via the Coulomb matrix elements, one can construct approximate eigenstates of the Hamiltonian (within the purview of single CB electron-hole pair excitations) by a simple superposition as follows,

\[
|\psi_{n,s}(\mathbf{Q})\rangle = \frac{\alpha_n}{\sqrt{A}} \sum_k \phi_{n,Q}^{ex}(\mathbf{Q}, s) \frac{\mathbf{k}}{N_{ex}}
\]

\[
\times c_s^{\dagger}(\mathbf{k} + \lambda_{c}\mathbf{Q}) b_s(\mathbf{k} - \lambda_{b}\mathbf{Q})(GS)
\]

\[
+ \sum_{m,s'} \frac{\beta_m}{\sqrt{A^3}} \sum_{k_{1,2,\mathbf{p}}} \phi_{m,Q}^{ex}(\mathbf{k}_{1,2}, s; \mathbf{p}, s')
\]

\[
\times c_{s'}^{\dagger}(\mathbf{k}_{1,2} - \lambda_{c}\mathbf{Q}) b_{s'}(\mathbf{k}_{1,2} + \lambda_{b}\mathbf{Q})(GS)
\]

\[
\sum_{k_{1,2,\mathbf{p}}}
\]

\[
N_{ir}
\]

\[
\frac{\mathbf{k}_{1,2,\mathbf{p}}}{N_{ir}}
\]

\[
\frac{\mathbf{k}_{1,2,\mathbf{p}}}{N_{ir}}
\]

\[
\frac{\mathbf{k}_{1,2,\mathbf{p}}}{N_{ir}}
\]
\[
\times c_s^\dagger(\vec{k}_1)c_s^\dagger(\vec{k}_2)b_s(\vec{k}_1 + \vec{k}_2 - (\vec{Q} + \vec{p}))c_{s'}(\vec{p})|GS\rangle
\]

(18)

where \(|GS\rangle\) is the ground state of the electron doped material. The above state resembles a Fermi-polaron variational state\(^{10,20,28}\). The normalization terms are,

\[
N_{ex} = \sqrt{1 - f_{c,s}(\vec{k} + \vec{Q})}
\]

\[
N_{tr} = \sqrt{(1 + \delta_{s,s'})f_{c,s'}(\vec{p}) \left[ 1 - f_{c,s}(\vec{k}_1) \right] \left[ 1 - f_{c,s'}(\vec{k}_2) \right]}
\]

(19)

The underlined vectors, \(\vec{k}_1\) and \(\vec{k}_2\), are defined as earlier in Section\(^{11}\). The states in the superposition are properly normalized and are orthogonal. The restrictions \(\vec{k}_1, \vec{k}_2 \neq \vec{p}\) in the summations in the second term follow from the fact that the states of the four-body Schrödinger equation correspond to fully connected diagrams and should have no direct optical matrix element with the ground state \(|GS\rangle\). This restriction also keeps the superposed trion states in the variational state orthogonal to the exciton states. When the variational state given above is used to minimize energy with respect to the Hamiltonian given earlier in \(1\), the trion states are found to be coupled to the exciton states via the Coulomb matrix elements \(M_{m,n}(\vec{Q}, s, s')\) given earlier (see \(13\)), and the energy eigenvalues \(\hbar\omega\) are found to obey \(17\) provided \(\gamma_{ex}\) and \(\gamma_{tr}\) are set to zero. Therefore, the same physics is captured by our coupled two-body and four-body Schrödinger equations and the exciton-polaron formalism. Furthermore, the optical conductivity calculated using the above variational state also matches the one found earlier in \(11\). The formalism presented here shows that the variational polaron state can be written in terms of the eigenstates of the two-body and the four-body Schrödinger equations (i.e. in terms of the exciton and trion eigenstates) and that all bound and unbound trion states must be included in the variational polaron state. Signatures of the resulting quantum coherence between the exciton and trion states have been observed experimentally\(^{31}\).

IV. NUMERICAL SIMULATION RESULTS AND DISCUSSION

For simulations, we consider a monolayer of 2D MoSe\(_2\) on a SiO\(_2\) substrate. In monolayer MoSe\(_2\), spin-splitting of the conduction bands is large (~35 meV\(^{20}\)) and the lowest conduction band in each of the \(K\) and \(K'\) valleys is optical coupled to the topmost valence band\(^{20}\). We use effective mass values of 0.7\(m_e\) for both \(m_e\) and \(m_h\) which agree with the recently measured value of 0.35\(m_e\) for the exciton reduced mass\(^{12}\). We use a wavevector-dependent dielectric constant \(\epsilon(q)\) appropriate for 2D materials, as described in our earlier work\(^{12}\) to screen the Coulomb potentials. We should emphasize here that besides \(\gamma_{ex}\) and \(\gamma_{tr}\), and unlike in previous works\(^{8,10}\), there are no other free parameters in our theoretical model and no artificial upper cut-offs of momenta integrals to avoid divergences.

A. Trion and Exciton Radii and Energies

As discussed in previous works\(^{8,10}\), exchange correlations favor only singlet trion bound states in MoSe\(_2\), in which the exciton belongs to one valley and the bound electron-hole pair belongs to the other valley. We write

\[
\gamma_{tr}, \text{ and unlike in previous works}^{8,10} \text{ there are no other free parameters in our theoretical model and no artificial upper cut-offs of momenta integrals to avoid divergences.}
\]

\[
\text{A. Trion and Exciton Radii and Energies}
\]

\[
\text{As discussed in previous works}^{8,10} \text{, exchange correlations favor only singlet trion bound states in MoSe}_2, \text{ in which the exciton belongs to one valley and the bound electron-hole pair belongs to the other valley. We write}
\]
a product variational wavefunction for the lowest energy four-body bound singlet trion eigenstate with \( Q = 0 \), as follows,

\[
\phi^{tr}_{m=0,Q=0}(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2) = \chi(\vec{k}_1, s_1; \vec{k}_2, s_2) \varphi(\vec{p}, s_2)
\]

(20)

The form of the CB hole wavefunction \( \varphi(\vec{p}, s_2) \) is determined from exchange energy considerations, as discussed in Sec. [II]D. Since the trion radii are expected to be much smaller than the size of the CB hole, which cannot be smaller than \( \sim 1/k_F \), the simplest and the easiest way to get the smallest CB hole is to assume that \( \varphi(\vec{p}, s_2) = \sqrt{f_{c,s_2}(\vec{p})}/n_{s_2} \), where proper wavefunction normalization requires \( n_{s_2} = A^{-1} \sum_a f_{c,a}(\vec{p}) \). We assume the following symmetric variational wavefunction for \( \chi(\vec{r}_1, s_1; \vec{r}_2, s_2) \) (assuming \( s_1 \neq s_2 \))

\[
\chi(\vec{r}_1, s_1; \vec{r}_2, s_2) \propto \exp(-|\vec{r}_1|-a_{tr})/|\vec{r}_2|/a_{tr} + \{\vec{r}_1 \leftrightarrow \vec{r}_2\}
\]

(21)

For the lowest energy \( Q = 0 \) bound exciton state we use the variational wavefunction

\[
\phi^{ex}_{n=0,Q=0}(\vec{r}, s) \propto \exp(-|\vec{r}|/a_{ex})
\]

(22)

Using the radii, \( a_{tr} \), \( b_{tr} \), and \( a_{ex} \), as variational parameters, we find the eigenenergies, \( E^*_{n=0}(Q = 0, s_1, s_2) \) and \( E^*_{n=0}(Q = 0, s_1) \), as a function of the electron density. The results are shown in Fig. 4 which plots these energies with respect to the material bandgap \( E_g \). The corresponding trion and exciton radii are plotted in Fig. 5. The trion binding energy \( E^*_b \), defined as \( E^*_b = E^*_{n=0}(Q = 0, s_1) - E^*_{n=0}(Q = 0, s_1, s_2) \), is also plotted. The exciton binding energy decreases with the electron density due to phase space filling. The trion binding energy first increases with the electron density and then it decreases. The initial increase is due to two reasons,

- Suppose an exciton with center of mass momentum \( \vec{Q} = 0 \) grabs an electron with momentum \( \vec{p} \) within the Fermi sea to form a four-body bound state with momentum \( \vec{Q} = 0 \). The center of mass kinetic energy of the four-body state would be \( \frac{\hbar^2}{2m_e} - \frac{\hbar^2}{2m_c} \). The first term is the center of mass kinetic energy of the two CB electrons and one VB hole in the four-body bound state. The second term represents the kinetic energy of the CB hole in the four-body bound state. These energies are included in the terms on the LHS of (2). Averaging this energy with respect to the CB hole wavefunction \( \varphi(\vec{p}, s) \) contributes a factor \( E_F/3 \) to the trion binding energy.
- At small electron densities, phase space filling restricts electron-electron Coulomb repulsion more than electron-hole Coulomb attraction.

As the electron density increases further, the reduced phase space diminishes electron-hole Coulomb attraction as well and the binding energy of the trion decreases rapidly. It remains an open question if the trion binding energy eventually goes to zero or not at high enough electron densities. The approximations made in this work do not permit us to generate reliable results for electron densities higher than \( \sim 2 \times 10^{13} \) cm\(^{-2} \). Interestingly, the Fermi energy remains smaller than the trion binding energy for electron densities smaller than \( \sim 10^{13} \) 1/cm\(^3 \). Note that the binding energies of the exact eigenenergies are expected to be larger than our variational solutions. In this work, no bound trions states with an anti-symmetric wavefunction were found even for vanishingly small electron densities.

Fig. 5 shows that the exciton and the larger trion radii decrease with the electron density because phase space blocking inside the Fermi surface causes the wavefunctions to spread out more in the momentum space. For electron densities higher than \( \sim 5 \times 10^{12} \) cm\(^{-2} \), the two trion radii are almost identical and approximately equal to 0.9 nm.

B. Optical Conductivity Spectra

Fig. 6 shows the calculated real part of the optical conductivity, \( \sigma_{\omega}(\omega) \), is plotted for different electron densities for monolayer 2D MoSe\(_2\) on SiO\(_2\). The spectra are all normalized to peak optical conductivity value at zero electron density. T = 5K. The frequency axis is offset by the exciton eigenenergy \( E^{ex}_0(Q, s) \) of the two-body Schrödinger equation.

![FIG. 6: Calculated real part of the optical conductivity, \( \sigma_{\omega}(\omega) \), is plotted for different electron densities for monolayer 2D MoSe\(_2\) on SiO\(_2\). The spectra are all normalized to peak optical conductivity value at zero electron density. T = 5K. The frequency axis is offset by the exciton eigenenergy \( E^{ex}_0(Q, s) \) of the two-body Schrödinger equation.](image)
FIG. 7: Calculated energy difference between the two dominant peaks in the optical absorption spectra in Fig. 6 is plotted as a function of the Fermi energy $E_F$ for monolayer 2D MoSe$_2$ on SiO$_2$. The dashed line has unit slope and shows that the calculated energy difference varies as $E_F$ at high electron densities.

FIG. 8: The maximum optical conductivity (real part) values of the two absorption peaks in Fig. 6, normalized to the maximum optical conductivity value at zero electron density, are plotted as a function of the electron density for monolayer 2D MoSe$_2$ on SiO$_2$.

The values of $\gamma_{ex}$ and $\gamma_{tr}$ were both chosen to be 4 meV. The spectra shows two distinct peaks. At small electron densities, the higher energy peak dominates and carries all the spectral weight. As the electron density increases, the spectral weight shifts from the higher energy peak to the lower energy peak. The higher and lower energy peaks have also been called repulsive and attractive exciton-polaron peaks, respectively. Very often in the literature they are just referred to as the exciton peaks and the trion peak, respectively. These two peaks arise from the Coulomb coupling of the excitons and the trions, as discussed earlier in this paper. For very small electron densities, their energies coincide with those of excitons and trions as obtained from the two-body and the four-body Schrödinger equations. The coupling between the excitons and the trions, described by the matrix elements $M_{m,n}(Q, s, s')$ in (13), increases with the electron density and, therefore, the energy difference between the two peaks in the absorption spectra also increases with the electron density. Fig. 7 plots this energy difference as a function of the Fermi energy $E_F$ for monolayer 2D MoSe$_2$ on SiO$_2$. The dashed line has unit slope and shows that the calculated energy difference varies approximately as $E_F$ at high electron densities. Fig. 6 shows that as the electron density increases, the lower energy peak shifts down to lower energies much more than the upward motion of the higher energy peak. This happens because the continuum of exciton-electron scattering states lies just above the higher energy peak and prevent the higher energy peak from moving upwards too much. Fig. 8 plots the peak optical conductivity (real part) of the two absorption peaks, normalized to the peak optical conductivity at zero electron density, as a function of the electron density. The lower energy peak begins to dominate when the electron density exceeds $6 \times 10^{12}$ cm$^{-2}$. As the electron density increases, the higher energy peak also loses spectral weight to the broad continuum of single electron-hole pair excitations from exciton-electron scattering. This results not only in the broadening of the higher energy peak but also in the appearance of a broad pedestal around the base of the peak that is more prominent on its higher energy side. The lower energy peak, on the other hand, does not broaden as the electron density increases. The integrated area under the plotted conductivity spectra in Fig. 6 is almost conserved (but not exactly conserved) as the electron density increases, in agreement with the sum rule in (16). These observations are all in good agreement with the experimental results reported for two dimensional materials$^{1,5}$, and with previous theoretical works$^{8,9,13}$.

C. The Importance of Including Exciton-Electron Scattering States (Unbound Trion States)

An important point that needs to be emphasized here is that without including exciton-electron scattering, that takes spectral weight away from the higher energy peak as shown in Fig. 6, the peak optical conductivity of the lower energy peak can never exceed the peak conductivity of the higher energy peak. This follows from the basic physics of two coupled systems and can be seen as follows. Assuming $\gamma_{ex} = \gamma_{tr} = \gamma$ for simplicity, $\vec{Q} = 0$, and ignoring exciton-electron scattering states, the poles of the exciton’s Green’s function will be at energies given by (17) and are found to be,

$$\hbar\omega = \frac{E_{ex}^0 + E_{tr}^0}{2} - i\gamma \pm \sqrt{\left(\frac{E_{ex}^0 - E_{tr}^0}{2}\right)^2 + |M_{0,0}|^2}$$

(23)

The corresponding spectral weights for the lower and the higher energy peaks in the optical absorption spectra
where,

\[ W = \frac{\sqrt{\left( E_{0}^{ex} - E_{0}^{te} \right) \frac{2}{2} + |M_{0,0}|^2} - \left( E_{0}^{ex} - E_{0}^{te} \right) \frac{2}{2} + |M_{0,0}|^2}}{2\sqrt{\left( E_{0}^{ex} - E_{0}^{te} \right) \frac{2}{2} + |M_{0,0}|^2}} \]  \hspace{1cm} (24)

When the electron density is zero, \( M_{0,0} \) is zero, and the spectral weight all lies in the higher energy exciton peak in the optical absorption spectrum. As the electron density increases, the spectral weight begins to shift to the lower energy peak. But even when the electron density, and therefore \( M_{0,0} \), are very large, the value of \( W \) never exceeds 1/2. Including the contribution of exciton-electron scattering states (or unbound trion states) is therefore necessary in producing the results shown in Fig. [3] (where the peak optical conductivity of the lower energy peak is shown to become much larger than the peak optical conductivity of the higher energy peak at high electron densities) and accurately reproducing the experimental observations.

V. CONCLUSION

In this paper, we presented a theoretical model that explains the behavior of trions and excitons in doped 2D materials. Coulomb scattering couples the exciton and trion states in doped materials. This coupling is well described by two coupled Schrödinger equations for excitons and trions that we derived using the many body density matrix technique. The calculated optical conductivity was shown to explain the prominent features of the experimentally measured optical absorption spectra and also satisfy the optical conductivity sum rule exactly. The eigensolution of the coupled Schrödinger equations, constructed using a superposition of exciton and trion states, had the same form as a Fermi polaron state and revealed the connection between our approach and polaron physics. The work presented here will help to clarify the physics associated with excitons and trions in doped 2D materials.

There are still several questions that remain open in this area. For example, it is not clear if the trion eigenequation has bound states at very high electron densities (much larger than \( \sim 10^{13} \) cm\(^{-2}\)). If not, then how does the conductivity spectra evolve at very high electron densities? At high enough electron densities, multiple electron-hole pair excitations, ignored in this work, are also expected to become important. Their inclusion is expected to broaden the lower energy absorption peak as well and steal spectral weight from it. Indeed, multiple pair excitations have already been shown to play an important role in Fermi polaron physics in atomic systems. Finally, the role of Fermi edge affects, which involves multiple electron-hole pair excitations, in this context is not clear but they are also expected to become increasingly important at high electron densities. Exploring answers to these questions will be the subject of future work.

VI. ACKNOWLEDGMENTS

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Appendix A: Appendix: Hermitian Two-Body Exciton Schrödinger Equation

Equation \( (30) \), without the first term and the last two terms on the RHS, is the standard eigenvalue equation for excitons\(^{20-23} \). However, the equation is not Hermitian. It can be converted into a Hermitian equation. We define \( \mathcal{P}_{Q}(\vec{k}, \vec{s}) \) as \( \mathcal{P}_{\bar{Q}}(\vec{k}, \vec{s})/\sqrt{1 - f_{c,s}(\vec{k} + \bar{Q})} \). We also include \( \left[ 1 - f_{c,s}(\vec{k} + Q) \right] \) in the last term on the RHS in \( (30) \) containing the Coulomb potential \( V(\vec{q}) \). This added factor does not show up at this level in the density matrix technique but its inclusion ensures the Hermiticity of the set of coupled two-body and four-body Schrödinger equations. Physically, it restricts the phase space for electron scattering just like the first and the second terms on the RHS. With these definitions and changes, we obtain,

\[
\begin{align*}
\mathbf{E}_{c,s}(\vec{k} + \vec{Q}) - \mathbf{E}_{c,s}(\vec{k}) & \rightleftharpoons \mathcal{P}_{\bar{Q}}(\vec{k}, \vec{s}) \\
- \frac{1}{A} \sum_{\bar{Q}_s} U(\vec{q}) & \sqrt{1 - f_{c,s}(\vec{k} + \bar{Q})} \\
+ \sqrt{1 - f_{c,s}(\vec{k} + \bar{Q})} & \sum_{\bar{Q}_s} U(\vec{q}) \\
\times \mathcal{P}_{\bar{Q}}(\vec{k} + \vec{q}, \vec{s}) & \sqrt{1 - f_{c,s}(\vec{k} + \bar{Q} + \vec{q})} \\
- \frac{1}{A} & \sum_{\bar{Q}_s} V(\vec{q}) \sqrt{1 - f_{c,s}(\vec{k} + \bar{Q})} \\
\times T_{\bar{Q}}(\vec{k} + (\xi + \eta)\vec{p} - \xi\vec{Q} - \vec{q} - \vec{s}; \vec{p}, \vec{s}') & \sqrt{1 - f_{c,s}(\vec{k} + \bar{Q})} \\
\times T_{\bar{Q}}(\vec{k} + (\xi + \eta)\vec{p} - \xi\vec{Q} + \vec{q} + \vec{s}; \vec{p}, \vec{s}') & \sqrt{1 - f_{c,s}(\vec{k} + \bar{Q})} \\
\end{align*}
\]

(A1)

The homogeneous part of the above equation is now a Hermitian eigenvalue equation. It has a complete set
of orthonormal eigenfunctions $\phi^{ex}_{\vec{q},s}(\vec{k} + \lambda_0 \vec{Q}, s)$. In the limit of very low electron density, when phase space filling effects can be ignored, and assuming $Q << k_F$, the eigenenergies of the bound exciton states can be expressed as,

$$E_n^{ex}(\vec{Q}, s) = E_n^{ex}(\vec{Q} = 0, s) + \frac{\hbar^2 Q^2}{2m_{ex}} \quad (A2)$$

Appendix B: Appendix: Hermitian Four-Body Trion Schrödinger Equation

Equation (5), without the last two terms on the RHS, is a four-body eigenvalue equation for trions. The equation is not Hermitian. It can be converted into a Hermitian eigenvalue equation with a few approximations. The term \[1 - f_{c,s_1}(\vec{k}_1) - f_{c,s_2}(\vec{k}_2)\] on the RHS can be replaced by \[\left[1 - f_{c,s_1}(\vec{k}_1) \right] \left[1 - f_{c,s_2}(\vec{k}_2)\right]\]. The difference between the two, \(-f_{c,s_1}(\vec{k}_1)f_{c,s_2}(\vec{k}_2)\), stems from the fact that the four-body correlation function $T^{ex}_Q$ can be non-zero if the correlations are between electrons outside the Fermi sea or if they are between holes inside the Fermi sea. For the former, the Coulomb interaction is repulsive. But for the latter, the Coulomb interaction is attractive because the CB hole mass is negative [12].

In this work, correlation between holes in the Fermi sea may be ignored since the trion radii are smaller than the inverse Fermi momentum for electron densities smaller than $2 \times 10^{11}$ cm$^{-2}$. Similarly, one can replace the terms \[f_{c,s_2}(\vec{p}) - f_{c,s_1/2}(\vec{k}_1/2)\] on the RHS by \[f_{c,s_2}(\vec{p}) \left[1 - f_{c,s_1/2}(\vec{k}_1/2)\right]\]. We then define $T^{ex}_Q(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2)$ as,

$$T^{ex}_Q(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2) = \frac{T^{ex}_Q(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2)}{\sqrt{f_{c,s_2}(\vec{p}) \left[1 - f_{c,s_1}(\vec{k}_1)\right] \left[1 - f_{c,s_2}(\vec{k}_2)\right]}} \quad (B1)$$

With the above approximations and definitions, we obtain,

$$\left[E_{E,s_1}(\vec{k}_1) + E_{E,s_1}(\vec{k}_2) - E_{E,s_1}(\vec{k}_1 + \vec{k}_2) - (\vec{Q} + \vec{p})\right] - E_{E,s_2}(\vec{p}) + v_{\tau tr} - \hbar\omega) T^{ex}_Q(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2) = \sqrt{\frac{1 - f_{c,s_1}(\vec{k}_1)}{A}} \left[1 - f_{c,s_2}(\vec{k}_2)\right] \sum_q V(q) \times$$

$$T^{ex}_Q(\vec{k}_1 + \vec{q}, s_1; \vec{k}_2 - \vec{q}, s_2; \vec{p}, s_2) \times \sqrt{\frac{1 - f_{c,s_1}(\vec{k}_1 + \vec{q})}{A}} \left[1 - f_{c,s_2}(\vec{k}_2 - \vec{q})\right] \sum_q U(q)$$

The homogeneous part of the above equation is now a Hermitian eigenvalue equation. It has a complete set of orthonormal eigenfunctions $\phi^{ex}_{m_0,\vec{q}}(\vec{k}_1, s_1; \vec{k}_2, s_2; \vec{p}, s_2)$. In the limit of very low electron density, when phase space...
filling effects can be ignored, and assuming $Q \ll k_F$, the eigenenergies of the bound trion states can be expressed as,

$$E_{tr}^m(\vec{Q}, s_1, s_2) = E_{tr}^m(\vec{Q} = 0, s_1, s_2) + \frac{\hbar^2 Q^2}{2m_t}$$  \quad (B3)