Pair-excitation energetics of highly correlated many-body states

M Mootz, M Kira and S W Koch
Department of Physics, Philipps-University Marburg, Renthof 5, D-35032 Marburg, Germany
E-mail: martin.mootz@physik.uni-marburg.de

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Abstract. A microscopic approach is developed to determine the excitation energetics of highly correlated quasi-particles in optically excited semiconductors based entirely on a pair-correlation function input. For this purpose, the Wannier equation is generalized to compute the energy per excited electron–hole pair of a many-body state probed by a weak pair excitation. The scheme is verified for the degenerate Fermi gas and incoherent excitons. In a certain range of experimentally accessible parameters, a new stable quasi-particle state is predicted which consists of four to six electron–hole pairs forming a liquid droplet of fixed radius. The energetics and pair-correlation features of these ‘quantum droplets’ are analyzed.

1 Author to whom any correspondence should be addressed.

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1. Introduction

Interactions may bind matter excitations into new stable entities, quasi-particles, that typically have very different properties than the non-interacting constituents. In semiconductors, electrons in the conduction band and vacancies, i.e. holes, in the valence band attract each other via the Coulomb interaction. Therefore, the Coulomb attraction may bind different numbers of electron–hole pairs into a multitude of quasi-particle configurations. The simplest example is an exciton which consists of a Coulomb-bound electron–hole pair and exhibits many analogies to the hydrogen atom. Two excitons can bind to a molecular state known as the biexciton. Both, exciton and biexciton resonances can be routinely accessed in present-day experiments by exciting a high quality direct-gap semiconductor optically from its ground state. Even the exciton formation can directly be observed in both optical and terahertz spectroscopy and their abundance can be controlled via the intensity of the optical excitation. Also higher correlated quasi-particles can emerge in semiconductors. For instance, polyexcitons or macroscopic electron–hole droplets have been detected, especially in semiconductors with an indirect gap.

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To determine the energetics of a given quasi-particle configuration, one can apply density-functional theory based on the functional dependence of the total energy on the electron density \[ \text{[13, 14]} \]. This procedure is well established in particular for ground-state properties. However, whenever one wants to model experimental signatures of excited quasi-particle states in the excitation spectra, the applicability of density-functional theory becomes challenging, especially for highly correlated states.

In this paper, we develop a new scheme to determine the excitation energetics of highly correlated quasi-particle configurations. We start directly from the pair-correlation function, not from the density functional, and formulate a framework to compute the pair-excitation energetics. The electron–hole pair-correlation function \( g(r) \) defines the conditional probability of finding an electron at the position \( r \) when the hole is at the origin. As an example, we show in figure 1 examples of \( g(r) \) for excitons (left) and quantum droplets (right). Here, we refer to quantum droplets as a quasi-particle state where few electron–hole pairs, typically four to six, are in a liquid-like state bounded within a sphere of microscopic radius \( R \).

In general, \( g(r) \) always contains a constant electron–hole plasma contribution (gray shaded area) stemming from the mean-field aspects of the many-body states. The actual bound quasi-particles are described by the correlated part \( \Delta g(r) \) (blue shaded area) which decays for increasing electron–hole separation. For 1s excitons, \( \Delta g(r) \propto |\phi_{1s}(r)|^2 \) decreases monotonically and has the shape defined by the 1s-exciton wavefunction \( \phi_{1s}(r) \) \[15\]. Since the electrons and holes in a quantum droplet are in a liquid phase, \( \Delta g(r) \) must have the usual liquid structure where particles form a multi-ring-like pattern where the separation between the rings is defined by the average particle distance \[16–18\]. Due to the electron–hole attraction, one also observes a central peak, unlike for single-component liquids.

We derive the pair-excitation energetics for an arbitrary initial many-body state in section 2. In this connection, we first study the pair excitations of the semiconductor ground state before we extend the approach for an arbitrary initial many-body state. We then test our approach for the well-known cases of a degenerate Fermi gas and incoherent excitons in section 3. In section 4, we apply our scheme to study the energetics and structure of quantum droplets based on electron–hole correlations in a GaAs-type quantum well (QW). The effect of carrier–carrier correlations on the quantum droplet energetics is analyzed in section 5.

**Figure 1.** Schematic representation of the exciton (left) and the quantum droplet electron–hole (eh) pair-correlation function \( g(r) \). The plasma contribution (gray shaded area) is shown together with the correlation contribution (blue shaded area). The radius of the quantum droplet is indicated by the vertical dashed line and each of the rings are labeled.
Figure 2. Schematic representation of a pair excitation. The quasi-particle configuration is shown before (left) and after (right) the pair excitation. Electron (holes) are symbolized by blue (red) circles while a yellow ellipse surrounds the correlated pairs. The level of filling indicates the fraction of electrons and holes bound as correlated pairs.

2. Energy and correlations in many-body systems

For resonant excitations, the excitation properties of many direct-gap semiconductor QW systems can be modeled using a two-band Hamiltonian \[ \hat{H} = \sum_{k,\lambda} \epsilon_{\lambda}^{c} a_{\lambda,k}^{\dagger} a_{\lambda,k} + \frac{1}{2} \sum_{k,k',q,\lambda,\lambda'} V_{q} a_{\lambda',k+q}^{\dagger} a_{\lambda',k}^{\dagger} a_{\lambda,k} a_{\lambda,k'}, \] (1)

where the Fermionic operators \( a_{\nu(c),k}^{\dagger} \) and \( a_{\nu(c),k} \) create and annihilate an electron with crystal momentum \( \hbar \mathbf{k} \) in the valence (conduction) band, respectively. We consider excitations close to the \( \Gamma \) point such that the kinetic energies can be treated as parabolic

\[ \epsilon_{c}^{c} = \frac{\hbar^2 \mathbf{k}^2}{2m_{e}} + E_{g}, \quad \epsilon_{v}^{v} = -\frac{\hbar^2 \mathbf{k}^2}{2m_{h}} \] (2)

with the bandgap energy \( E_{g} \) and the effective masses for the electron \( m_{e} \) and hole \( m_{h} \). The Coulomb interaction is characterized by the matrix element \( V_{q} \) of the quantum confined system [1]. We have formally set \( V_{q=0} = 0 \) to eliminate the \( q = 0 \) contribution from the Coulomb sum, which enforces the overall charge neutrality in the system [1].

For later use, we introduce Fermion field operators without the lattice-periodic functions

\[ \hat{\Psi}_{c}(\mathbf{r}) = \frac{1}{\sqrt{S}} \sum_{k} a_{c,k} e^{i \mathbf{k} \cdot \mathbf{r}}, \quad \hat{\Psi}_{h}(\mathbf{r}) = \frac{1}{\sqrt{S}} \sum_{k} a_{v,k}^{\dagger} e^{-i \mathbf{k} \cdot \mathbf{r}} \] (3)

for electrons and holes, respectively. These can be directly used to follow e.g. electron (hole) densities \( \rho_{c(h)}(\mathbf{r}) \equiv \langle \hat{\Psi}_{c(h)}^{\dagger}(\mathbf{r}) \hat{\Psi}_{c(h)}(\mathbf{r}) \rangle \) on macroscopic length scales because the unit-cell dependence is already averaged over. The corresponding normalization area is given by \( S \).

2.1. Ground-state pair excitations

A schematic representation of a pair excitation is shown in figure 2 to illustrate the detectable energetics. The individual electrons and holes are symbolized by circles while the yellow ellipse
surrounds the correlated pairs. The level of blue (red) filling indicates the fraction of electrons (holes) bound as correlated pairs within the entire many-body system. This fraction can be changed continuously by applying, e.g. an optical field to generate pair excitations. If all pairs are bound to a single quasi-particle type, the initial energy of the system is

$$E_{\text{ini}} = N E(N),$$

where $N$ is the total number of pairs. Since $N$ is typically much larger than the number of pairs within a quasi-particle, it is meaningful to introduce $E(N)$ as the binding energy per excited electron–hole pair. For stable quasi-particle configurations, a change in $N$ does not alter $E(N)$, yielding the stability condition $\frac{\partial E(N)}{\partial N} = 0$.

We now assume that only a small number of pairs, $\delta N$, is excited from the quasi-particle into an unbound pair. An example of the excited configuration is presented in the right panel of figure 2. This state has the energy

$$E_{\text{fin}} = (N - \delta N) E(N) - \delta N E_{\text{pair}}$$

$$= N E(N) + \delta N (E_{\text{pair}} - E(N)) + \delta N \frac{\partial E(N)}{\partial N} + \mathcal{O}(\delta N^2),$$

where $E_{\text{pair}}$ is the energy of the unbound pair. After we apply the stability condition $\frac{\partial E(N)}{\partial N} = 0$, we find that the pair excitation produces an energy change $\Delta E \equiv E_{\text{fin}} - E_{\text{ini}} = \delta N (E_{\text{pair}} - E(N)) + \mathcal{O}(\delta N^2)$ such that the energy per excited particle becomes

$$\bar{E} = \lim_{\delta N \rightarrow 0} \frac{\Delta E}{\delta N} = E_{\text{pair}} - E(N).$$

This difference defines how much energy the electron–hole pair gains by forming the quasi-particle from unbound pairs.

To develop a systematic method describing the quasi-particle energetics, we start from the simplest situation where the unexcited semiconductor is probed optically, i.e. by inducing a weak pair excitation. The corresponding initial state is then the semiconductor’s ground state $|G\rangle$ where all valence bands are fully occupied while all conduction bands are empty. Following the analysis in [15], we introduce the coherent displacement-operator functional [1, 15]

$$\hat{D}[\psi] = e^{i\hat{S}[\psi]}; \quad \hat{S}[\psi] = \sum_{\mathbf{k}} \left( \psi_{\mathbf{k}} a_{c,\mathbf{k}}^\dagger a_{v,\mathbf{k}} - \psi_{\mathbf{k}}^* a_{v,\mathbf{k}}^\dagger a_{c,\mathbf{k}} \right)$$

(7)

to generate pair excitations. Here, $\varepsilon$ is an infinitesimal constant and $\psi_{\mathbf{k}}$ is a function to be determined later using a variational approach. The probed ground state has a density matrix $\hat{\rho}_G$ that determines the pair-excitation state via

$$\hat{\rho}[\psi] = \hat{D}[\psi] \hat{\rho}_G \hat{D}^\dagger[\psi].$$

(8)

We see from the definition (7) that $\hat{D}[\psi]$ generates pair excitations to the semiconductor ground state $\hat{\rho}_G$ because $\hat{S}[\psi]$ contains all elementary, direct, pair-excitation processes $a_{c,\mathbf{k}}^\dagger a_{v,\mathbf{k}} (a_{v,\mathbf{k}}^\dagger a_{c,\mathbf{k}})$ where an electron is moved from the valence (conduction) to the conduction (valence) band. The weak excitation of the probe is realized by making $\varepsilon$ infinitesimal, i.e. $\varepsilon \ll 1$.

As shown in [15], the pair excitation (7) generates the electron–hole distribution and polarization

$$f_{\mathbf{k},\psi} = \text{Tr} \left[ a_{c,\mathbf{k}}^\dagger a_{c,\mathbf{k}} \hat{\rho}[\psi] \right] = \text{Tr} \left[ a_{v,\mathbf{k}} a_{v,\mathbf{k}}^\dagger \hat{\rho}[\psi] \right] = \sin^2(\varepsilon |\psi_{\mathbf{k}}|),$$

$$P_{\mathbf{k},\psi} = \text{Tr} \left[ a_{v,\mathbf{k}}^\dagger a_{c,\mathbf{k}} \hat{\rho}[\psi] \right] = e^{i\varepsilon_{\mathbf{k}}} \sin(\varepsilon |\psi_{\mathbf{k}}|) \cos(\varepsilon |\psi_{\mathbf{k}}|),$$

(9)
respectively. Here, $\psi_k = |\psi_k|e^{i\varphi_k}$ has been defined in terms of a real-valued amplitude $|\psi_k|$ and phase $\varphi_k$. For the weak-excitation limit $\varepsilon \ll 1$, equation (9) reduces to

$$
E_{\text{pro}}[\psi] = E[\psi] - E_{\text{GS}} = \text{Tr} \left[ \hat{H} \hat{\rho} [\psi] \right] - \text{Tr} \left[ \hat{H} \hat{\rho}_G \right]
$$

(10)

$$
= \varepsilon^2 \left( \sum_k \frac{\hbar^2 k^2}{2\mu} |\psi_k|^2 - \sum_{k,k'} V_{k-k'} \psi_k \psi_{k'}^* \right) + O(\varepsilon^3), \quad \mu = \frac{m_em_h}{m_e + m_h},
$$

(11)

where we removed the ground-state energy $E_{\text{GS}}$ and introduced the reduced mass $\mu$.

2.2. Ordinary Wannier equation

The lowest pair-excitation energy can be found by minimizing $E_{\text{pro}}[\psi]$ with the constraint that the number of excited electron–hole pairs

$$
N_{\text{pro}} \equiv \sum_k f_{k,\psi} = \varepsilon^2 \sum_k |\psi_k|^2
$$

remains constant. This can be accounted for by the standard procedure of introducing a Lagrange multiplier $E_{\lambda}$ to the functional

$$
F[\psi] = E_{\text{pro}}[\psi] - E_{\lambda} \varepsilon^2 \sum_k |\psi_k|^2.
$$

(13)

By demanding $\delta F[\psi] = 0$ under any infinitesimal change $\psi_k \rightarrow \psi_k + \delta \psi_k$, this extremum condition produces the Wannier equation [15]

$$
\frac{\hbar^2 k^2}{2\mu} \psi_k - \sum_{k'} V_{k-k'} \psi_k = E_{\lambda} \psi_k.
$$

(14)

Fourier transform of equation (14) produces the real-space form

$$
\left[ -\frac{\hbar^2 \nabla^2}{2\mu} - V(\mathbf{r}) \right] \psi(\mathbf{r}) = E_{\lambda} \psi(\mathbf{r}),
$$

(15)

where $V(\mathbf{r})$ and $\psi(\mathbf{r})$ are the Fourier transformations of $V_k$ and $\psi_k$, respectively. Since equations (14) and (15) are the usual Wannier equations for excitons, the exciton wavefunction defines those pair excitations that produce minimal energy $E_{\lambda}$. At the same time, equation (15) is fully analogous to the Schrödinger equation of atomic hydrogen [1]. Therefore, $E_{\lambda}$ also defines the Coulombic binding energy of excitons.

For the identification of the quasi-particle energy, we use the result (6) and compute the energy per excited electron–hole pair

$$
\bar{E}_{\text{pro}} = \frac{E_{\text{pro}}}{N_{\text{pro}}}.
$$

(16)

By inserting solution (14) into equations (11) and (12), we find $\bar{E}_{\text{pro}} = E_{\lambda}$ showing that the energetics of the pair-excitations from the ground state are defined by the exciton resonances. As a result, the energy per probe-generated electron–hole pair produces a series of exciton
resonances that can be detected, e.g. in the absorption spectrum. We will show next that this variational approach can be generalized to determine the quasi-particle energetics for any desired many-body state.

2.3. Average carrier-excitation energy

Here, we start from a generic many-body system defined by the density matrix $\hat{\rho}_{\text{MB}}$ instead of the semiconductor ground state $\hat{\rho}_G$. We assume that $\hat{\rho}_{\text{MB}}$ contains spatially homogeneous excitations with equal numbers of electrons and holes, i.e.

$$N_{\text{eh}} = \sum_k f^e_k = \sum_k f^h_k, \quad \text{with} \quad f^e_k \equiv \langle a_{c,k}^\dagger a_{c,k} \rangle, \quad f^h_k \equiv 1 - \langle a_{v,k}^\dagger a_{v,k} \rangle,$$

(17)

where the electron (hole) distribution $f^e_k$ ($f^h_k$) is defined within the electron–hole picture [1]. In general, each electron–hole pair excitation increases the energy by $E_g$ because an electron is excited from the valence to the conduction band. To directly monitor the energetics of $\hat{\rho}_{\text{MB}}$, we remove the trivial $E_g N_{\text{eh}}$ contribution, yielding the average carrier energy

$$E_{\text{MB}} \equiv \langle \hat{H} \rangle - E_g N_{\text{eh}} = \text{Tr} \left[ \hat{H} \hat{\rho}_{\text{MB}} \right] - E_g N_{\text{eh}}$$

$$= \sum_k \left( \frac{\hbar^2 k^2}{2m_e} f^e_k + \frac{\hbar^2 k^2}{2m_h} f^h_k \right) - \frac{1}{2} \sum_{k,k'} V_{k-k'} \left( f^e_k f^e_{k'} + f^h_k f^h_{k'} \right) - \sum_{k,k'} V_{k-k'} P^*_{k'} P_k + \frac{1}{2} \sum_{k,k',q} \left[ V_q \left( c^q_{v,v,v,v} + c^q_{c,c,c,c} \right) - 2 V_{q+k-k} c^q_{v,c,c} \right],$$

(18)

which is an exact result for homogeneous excitation conditions. Using the cluster expansion [15], we identified the incoherent two-particle correlations

$$c^q_{v,v,v,v} \equiv \Delta \langle a_{v,k}^\dagger a_{v,k} a_{v,k+q} a_{v,k-q} \rangle, \quad c^q_{c,c,c,c} \equiv \Delta \langle a_{c,k}^\dagger a_{c,k} a_{c,k+q} a_{c,k-q} \rangle,$$

$$c^q_{c,c} \equiv \Delta \langle a_{c,k}^\dagger a_{c,k}^\dagger a_{c,k+q} a_{c,k} \rangle,$$

(19)

which represent the truly correlated parts of the respective two-particle expectation value. The first two correlations correspond to hole–hole and electron–electron correlations, respectively. Electron–hole correlations are described by $c^q_{v,c,c}$ where $\hbar q$ defines the center-of-mass momentum of the correlated electron–hole pairs.

The only coherent quantity in equation (18) is the microscopic polarization

$$P_k \equiv \langle a_{v,k}^\dagger a_{c,k} \rangle.$$

(20)

Consequently, the average carrier energy $E_{\text{MB}}$ of any $\hat{\rho}_{\text{MB}}$ is determined entirely by the single-particle expectation values $f^e_k$ and $P_k$ and the incoherent two-particle correlations $c^q_{v,v,v,v}$. In other words, the system energy is directly influenced by contributions up to second-order correlations. We will show in section 2.4 that this fundamental property allows us to determine the pair-excitation energetics of a given state when we know its singlets and doublets. In other words, we do not need to identify the properties of the higher order clusters to compute the pair-excitation energetics.

Since we are interested in long-living quasi-particles in the incoherent regime, we consider only those states $\hat{\rho}_{\text{MB}}$ which have vanishing coherences [1]. Therefore, we set $P_k$ and all coherent correlations to zero from now on. Furthermore, we assume conditions where the
electron–hole correlations \( c_{\text{eh}}^{q,k,k'} \) have a vanishing center-of-mass momentum \( \hbar \mathbf{q} = 0 \), i.e. we assume that the correlated pairs are at rest. As a result, the electron–hole correlations can be expressed in terms of

\[
\begin{align*}
  c_{\text{eh}}^{q,k,k'} &= \delta_{q,0} c_{\text{eh}}^{q,k,k} \equiv \delta_{q,0} g_{k,k'}.
\end{align*}
\]

For homogeneous and incoherent excitation conditions, the pair-correlation function can be written as

\[
g(r) \equiv \langle \hat{\psi}_e^\dagger(r) \hat{\psi}_h^\dagger(0) \hat{\psi}_h(0) \hat{\psi}_e(r) \rangle = \rho_e \rho_h + \Delta g(r),
\]

compare equation (3) [15]. The term \( \rho_e \rho_h \) describes an uncorrelated electron–hole plasma contribution, whereas the quasi-particle clusters determine the correlated part

\[
\Delta g(r) = \frac{1}{S^2} \sum_{k,k',q} c_{\text{eh}}^{q,k,k'} e^{i(k+q-k)\cdot r} = \frac{1}{S^2} \sum_{k,k'} g_{k,k'} e^{i(k-k')\cdot r}.
\]

To describe, e.g. excitons and similar quasi-particles, we use an ansatz

\[
\Delta g(r) = |g_0 \phi(r)|^2,
\]

where \( g_0 \) defines the strength of the correlation while the specific properties of the quasi-particles determine the normalized wavefunction \( \phi(r) \). In order to compute the quasi-particle energetics, we need to express \( \Delta g(r) \) in terms of the electron–hole correlation \( g_{k,k'} \). By writing \( \phi(r) = \frac{1}{2} \sum_k \phi_k e^{i k \cdot r} \), we find the unique connection

\[
g_{k,k'} = g_0^2 \phi_k^* \phi_k',
\]

where \( \phi(k) \) is the Fourier transformation of the wavefunction \( \phi(r) \).

As shown in appendix A, the electron and hole distributions \( f^e_k \) and \( f^h_k \), together with the incoherent correlations \( g_{k,k'} \), \( c_{\text{eh}}^{q,k,k'} \), and \( c_{\text{eh}}^{q,k,k} \) must satisfy the general conservation laws

\[
\left( f^e_k - \frac{1}{2} \right)^2 + g_{k,k} - \sum_{k'} c_{\text{eh}}^{q,k',k} \left( f^e_{k'} - \frac{1}{2} \right)^2 + g_{k,k'} = \frac{1}{2},
\]

\[
\left( f^h_k - \frac{1}{2} \right)^2 + g_{k,k} - \sum_{k'} c_{\text{eh}}^{q,k,k'} \left( f^h_{k'} - \frac{1}{2} \right)^2 + g_{k,k'} = \frac{1}{2}.
\]

As a consequence, we have to connect \( f^e_k \) and \( f^h_k \) with \( g_{k,k} \), \( c_{\text{eh}}^{q,k,k'} \), and \( c_{\text{eh}}^{q,k,k} \) to have a self-consistent description of the many-body state. Therefore, equation (26) has a central role when the energetics of many-body states is solved self-consistently.

We show in section 5 that the effect of electron–electron and hole–hole correlations can be neglected when the energetics of new quasi-particle states is analyzed. Therefore, we set \( c_{\text{eh}}^{q,k,k'} \) and \( c_{\text{eh}}^{q,k,k} \) to zero such that equation (26) reduces to

\[
\left( f^e_k - \frac{1}{2} \right)^2 + g_{k,k} = \frac{1}{4}, \quad f^e_k \equiv f^e_k = f^h_k.
\]

From this result, we see that the electron and hole distributions become identical as long as correlations are dominated by \( g_{k,k} \). A more general case with carrier–carrier correlations is studied in section 5. In the actual quasi-particle calculations, we solve equation (27)

\[
f_k = \frac{1}{2} \left( 1 + \sqrt{1 - 4 g_{k,k} \rho_{\text{MB}}} \right)
\]

that limits \( g_{k,k} \) to be below \( \frac{1}{4} \). In other words, the maximum of \( g_0 |\phi(k)|^2 \) is \( \frac{1}{2} \), based on the connection (25). The ‘+’ branch in equation (28) describes an inverted many-body system \( \hat{\rho}_{\text{MB}} \) corresponding to large electron–hole densities. Below inversion, only the ‘−’ branch contributes.
Once the self-consistent pair \((f_k, g_{k,k'})\) is found, we determine the corresponding electron–hole density via

\[
\rho_{ch} = \frac{1}{S} \sum_k f_k
\]

(29)

that becomes a functional of the electron–hole pair-correlation function due to its \(g_{k,k'}\) dependence via equation (28). In sections 3 and 4, we will use equation (27) to self-consistently determine \(f_k\) and \(g_{k,k'}\) for different quasi-particle configurations.

### 2.4. Pair-excitation energetics

To generalize the Wannier equation (14), we next analyze the pair-excitation energetics of an arbitrary homogeneous initial state \(\hat{\rho}_{MB}\). As shown in section 2.1, the simplest class of pair excitations can be generated by using the coherent displacement-operator functional (7). The pair-excitation state is then given by

\[
\hat{\rho}[\psi] = D[\psi] \hat{\rho}_{MB} D^\dagger[\psi],
\]

(30)

which is properly normalized \(\text{Tr}[\hat{\rho}[\psi]] = \text{Tr}[\hat{\rho}_{MB}] = 1\), as any density matrix should be.

As shown in appendix B, the pair excitation generates the polarization and electron–hole distribution

\[
P_{k,\psi} = (1 - f_k^e - f_k^h) \varepsilon \psi_k + \mathcal{O}(\varepsilon^3), \quad f_{k,\psi} = (1 - f_k^e - f_k^h) \varepsilon^2 \vert\psi_k\vert^2 + \mathcal{O}(\varepsilon^3),
\]

(31)

respectively where we have applied the weak excitation limit \(\varepsilon \ll 1\). For the sake of completeness, we keep the explicit dependences \(f_k^e, f_k^h\) and \(c_{q,k,k'}^{q,k',k}\) and take the limit of dominant electron–hole correlation after the central results for the pair excitations have been derived. In analogy to equation (11), pair excitations add the average carrier energy \(E_{\text{pro}}[\psi] \equiv E[\psi] - E_{MB}\) to the system. Technically, \(E[\psi]\) is obtained by replacing \(\rho_{MB}\) in equation (18) by \(\rho[\psi]\). The actual derivation is performed in appendix B, yielding again an exact relation for incoherent quasi-particles

\[
E_{\text{pro}}[\psi] = \varepsilon^2 \sum_k E_k \vert\psi_k\vert^2 - \varepsilon^2 \sum_{k,k'} V_{k,k'}^{\text{eff}} \psi_k \psi_k^*
\]

\[
+ \varepsilon^2 \sum_{k,k',q} V_q \left( c_{q,k,k'}^{q,k',k} \psi_{k-q} \psi_{k}^* + c_{q,k,k'}^{q,k,k'} \psi_{k} \psi_{k-q}^* - \text{Re}[c_{q,k,k'}^{q,k,k'}] \varepsilon^2 \vert\psi_k\vert^2 \right)
\]

\[
+ \mathcal{O}(\varepsilon^3),
\]

(32)

where we identified the renormalized kinetic electron–hole pair energy

\[
E_k \equiv \left[ \frac{\hbar^2 k^2}{2\mu} - \sum_{k'} V_{k-k'} \left( f_k^e + f_k^h \right) \right] \left( 1 - f_k^e - f_k^h \right) + 2 \sum_{k'} V_{k-k'} g_{k,k'}.
\]

(33)

The unscreened Coulomb interaction \(V_{k-k'}\) is modified through the presence of electron–hole densities and correlations via

\[
V_{k,k'}^{\text{eff}} \equiv \left( 1 - f_k^e - f_k^h \right) V_{k-k'} \left( 1 - f_k^e - f_k^h \right) + 2 g_{k,k'} V_{k-k'}.
\]

(34)

Since the phase-space filling factor \(1 - f_k^e - f_k^h\) becomes negative once inversion is reached, the excitation level changes the nature of the effective electron–hole Coulomb interaction.
from attractive to repulsive. At the same time, $g_{k,k'}$ can either enhance or decrease the Coulomb interaction depending on the nature of the pair correlation. The exact generalization of equation (32) for coherent quasi-particles is presented in appendix C.

2.5. Generalized Wannier equation

As in section 2.2, we minimize the functional $E_{\text{pro}}[\psi]$ with the constraint that the excitation $\epsilon^2 \sum_k |\psi_k|^2$ remains constant. Following the same variational steps as those producing equation (14), we obtain the generalized Wannier equation for incoherent quasi-particles:

$$E_k \psi_k - \sum_{k'} V_{k,k'}^{\text{eff}} \psi_{k'} + \sum_{k',q} V_q \left( c_{q,k',k+q} \psi_{k+q} + c_{q,k,k'} \psi_{k-q} \right)$$

$$- \sum_{k',q} V_q \text{Re} \left[ c_{q,k',k} + c_{q,k,k'} \right] \psi_{k'} = E_{\lambda} \psi_{k}. \quad (35)$$

For vanishing electron–hole densities and correlations, equation (35) reduces to the ordinary exciton Wannier equation (14). Since the presence of two-particle correlations and densities modifies the effective Coulomb interaction, it is possible that new quasi-particles emerge. The generalized Wannier equation with all coherent and incoherent contributions is presented in appendix C.

For the identification of the quasi-particle energy, we compute the energy per excited electron–hole pair (16). The number of excited electron–hole pairs of the probed many-body system is

$$N_{\text{pro}} \equiv \sum_k f_{k,\psi} = \epsilon^2 \sum_k \left( 1 - f_{k}^e - f_{k}^h \right) |\psi_k|^2 \quad (36)$$

according to equation (31). By inserting equation (35) into equation (32) and using the definitions (16) and (36), the energy per excited electron–hole pairs follows from

$$\bar{E}_{\text{pro}} = E_{\lambda} \frac{\sum_k |\psi_k|^2}{\sum_k |\psi_k|^2 \left( 1 - f_{k}^e - f_{k}^h \right)} \quad (37)$$

that defines the quasi-particle energy, based on the discussion in section 2.1.

3. Pair-excitation spectrum of the degenerate Fermi gas and of incoherent excitons

For all our numerical evaluations, we use the parameters of a typical 10 nm GaAs-QW system. Here, the reduced mass is $\mu = 0.0581 m_0$, where $m_0$ is the free-electron mass and the 1s-exciton binding energy is $E_0 = 9.5$ meV. This is obtained by using the dielectric constant $\varepsilon_r = 13.74$ of GaAs in the Coulomb interaction.

To compute the quasi-particle energetics for a given electron–hole density $\rho_{\text{eh}}$, we always start from the conservation law (27) to generate a self-consistent many-body state $\rho_{\text{MB}}$. We then use the found self-consistent pair ($f_k$, $g_{k,k'}$) as an input to the generalized Wannier equation (35) and numerically solve the pair excitation $\psi_{k}$ and $E_{\lambda}$. As shown in section 5, the effect of electron–electron and hole–hole correlations on the quasi-particle energetics is negligible such that we set $c_{q,k',k}$ and $c_{q,k,k'}$ to zero in equation (35).

The variational computations rigorously determine only the lowest energy $E_0$. However, it is useful to analyze also the characteristics of the excited states $E_{\lambda}$ to gain additional information.
about the energetics of the pair excitation acting upon $\hat{\rho}_{MB}$. To deduce the quasi-particle energetics, we normalize the energy $E_\lambda$ via equation (37). The resulting energy per excited electron–hole pair $\bar{E}_{pro}$ defines then the detectable energy resonances.

3.1. Degenerate Fermi gas

The simplest form of $\hat{\rho}_{MB}$ for an excited state is provided by the degenerate Fermi gas [20–23]

$$f_k = \theta(k - k_F), \quad g_{k,k'} = 0$$

because the two-particle correlations vanish. It is straightforward to show that the pair $(f_k, g_{k,k'})$ satisfies the conservation law (27) even though the system is inverted for all $k$ below the Fermi wave vector $k_F = \sqrt{4\pi \rho_{eh}}$. Due to this inversion, the degenerate Fermi gas provides a simple model to study quasi-particle excitations under optical gain conditions.

Figure 3(a) presents the electron–hole distribution $f_k$ as function of $k$ for the electron–hole density $\rho_{eh} = 2.5 \times 10^{10} \text{ cm}^{-2}$. The distribution has a Fermi edge at $k_F = 0.56 \times 10^8 \text{ m}^{-1}$ while $g_{k,k}$ is zero for all $k$ values (not shown). The numerically computed ground-state wavefunction
$\psi_k$ is plotted in figure 3(b) as solid line. We have applied the normalization $\sum_k |\psi_k|^2 = 1$. As a comparison, we also show the corresponding zero-density result ($f_k = 0$, $g_{k,k'} = 0$) as shaded area. While the zero-density wavefunction decays monotonically from the value 1.47, the degenerate Fermi gas has a $\psi_k$ that is negative-valued up to the Fermi edge $k_F$. Exactly at $k = k_F$, $\psi_k$ abruptly jumps from the value $-0.74$ to 1.89. Above roughly $k = 1.3 \times 10^8 \text{m}^{-1}$, both wavefunctions show a similar decay. The energetics of the related pair excitations is discussed later in section 3.3.

3.2. Incoherent excitons

According to the ansatz (25), the exciton state is determined by the electron–hole pair-correlation function

$$g_{k,k'} = \phi_{1s,k} \phi_{1s,k'}$$

with the 1s-exciton wavefunction $\phi_{1s,k}$ defining the initial many-body state $\hat{\rho}_{\text{MB}}$, not the pair-excitation state. Here, we have included the strength of the electron–hole correlation $g_0$ into the 1s-exciton wavefunction to simplify the notation. To compute $\phi_{1s,k}$, we have to solve the ordinary density-dependent Wannier equation [1, 15]

$$\tilde{E}_k \phi_{1s,k} - (1 - 2 f_k) \sum_{k'} V_{k-k'} \phi_{1s,k'} = E_{1s} \phi_{1s,k},$$

$$\tilde{E}_k = \frac{\hbar^2 k^2}{2\mu} - 2 \sum_{k'} V_{k-k'} f_{k'},$$

with the constraint imposed by the conservation law (27). In practice, we solve equations (27) and (40) iteratively. Since the specific choice $E_{1s}$ defines the electron–hole density (29) uniquely, we can directly identify the self-consistent pair $(f_k, g_{k,k})$ as function of $\rho_{eh}$. The explicit steps of the iteration cycle are presented in appendix D.

Figure 4(a) shows the resulting normalized electron–hole pair-correlation function $\Delta \tilde{g}(r) = \Delta g(r) / \rho_{eh}^2$ for an electron–hole density of $\rho_{eh} = 2.5 \times 10^{10} \text{cm}^{-2}$. For the incoherent excitons, $\Delta \tilde{g}(r)$ is a monotonically decaying function. The corresponding iteratively solved $f_k$ (black line) and $g_{k,k}$ (red line) are plotted in figure 4(b). The pair correlation $g_{k,k}$ decays monotonically from the value 0.21. Also the electron–hole distribution $f_k$ function decreases monotonically, peaking at 0.30. This implies that the phase-space filling already reduces the strength of the effective Coulomb potential (34) for small momentum states which typically dominate the majority of ground-state configurations.

The corresponding normalized ground-state wavefunction $\psi_k$ of the pair excitation is shown in figure 4(c) (solid line) together with the zero-density result (shaded area). Both functions show a similar decay for $k$ values larger than $2 \times 10^8 \text{m}^{-1}$. In contrast to the zero-density result, we observe that $\psi_k$ has a peak at $k = 0.59 \times 10^8 \text{m}^{-1}$. Interestingly, the maximum of $\psi_k$ is close to $k_F$ of the degenerate Fermi gas analyzed in figure 3 because both cases have the same density giving rise to sufficiently strong phase-space filling effects.

3.3. Energetics of pair excitations

We next analyze the influence of the electron–hole density $\rho_{eh}$ on the pair-excitation energetics for the degenerate Fermi gas and for incoherent excitons. The result for the degenerate Fermi gas is presented in figure 5(a) where the ground-state energy $E_0$ (solid line), the continuum (shaded area) and the ground-state energy per excited electron–hole pair $\tilde{E}_{\text{pro}}$ (dashed line)

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Figure 4. Solutions of the generalized Wannier equation for incoherent excitons. (a) The normalized electron–hole pair-correlation function $\Delta \tilde{g}(r)$ is shown for $\rho_{\text{eh}} = 2.5 \times 10^{10} \text{ cm}^{-2}$. (b) The corresponding electron–hole distribution $f_k$ (black line) and the correlation $g_{k,k}$ (red line) as function of $k$. (c) Normalized ground-state wavefunction for vanishing electron–hole density (shaded area) and $\rho_{\text{eh}} = 2.5 \times 10^{10} \text{ cm}^{-2}$ (solid line).
In contrast to the degenerate Fermi gas, the ground-state energy blue shifts. This blue shift remains present in $E_{\text{pro}}$ (dashed line) and is consistent with the blue shift of the excitonic absorption when excitons are present in the system, as detected in several measurements \cite{6, 8, 28, 29}. The nearly linear dependence of $E_{\text{pro}}$ on the electron–hole density is consistent with earlier theoretical studies \cite{30–32}. In particular, $E_0$ blue shifts faster than the continuum does. If we interpret the energy difference of $E_0$ and continuum as the exciton-binding energy, we find that the exciton-binding energy decreases from 9.5 to 8.0 meV as the density is increased to $\rho_{\text{eh}} = 3.6 \times 10^{10} \text{ cm}^{-2}$, which shows that excitons remain bound even at elevated densities. For later reference, the density $2.5 \times 10^{10} \text{ cm}^{-2}$ produces $E_{\text{pro}} = -7.1 \text{ meV}$ energy per excited electron–hole pair.

4. Pair-excitation spectrum of quantum droplets

To define a quantum droplet state, we assume that the electron–hole pairs form a liquid confined within a small droplet with a radius $R$ as discussed in connection with figure 1. Since the QW is two dimensional, the droplet is confined inside a circular disc with radius $R$. We assume that the droplet has a hard shell created by the Fermi pressure of the plasma acting upon the droplet. As a result, the solutions correspond to standing waves. Therefore, we define the quantum droplet state via the standing-wave ansatz

$$
\phi(r) = J_0 \left( x_n \frac{r}{R} \right) e^{-\kappa r} \theta(R - r) \tag{41}
$$

to be used in equation (24). Here, $x_n$ is the $n$th zero of the Bessel function $J_0(x)$. The Heaviside function $\theta(x)$ confines the droplet inside a circular disc with radius $R$. The additional decay constant $\kappa$ is used for adjusting the electron–hole density \cite{29} when the quantum droplet has radius $R$ and $n$ rings.
Figure 6. Solutions of the generalized Wannier equation for quantum droplets. (a) The normalized electron–hole pair-correlation function \( \Delta \bar{g}(r) \) is shown for \( g_{\text{max}} = 0.24 \) (shaded area) and \( g_{\text{max}} = \frac{1}{4} \) (dashed line). The quantum droplet has \( n = 4 \) rings, \( R = 90.8 \text{ nm} \) (vertical line) and \( \rho_{\text{eh}} = 2.5 \times 10^{10} \text{ cm}^{-2} \). (b) The corresponding electron–hole distribution \( f_k \) (black lines) and correlation \( g_{k,k} \) (red lines) as function of \( k \) for \( g_{\text{max}} = 0.24 \) (solid lines) and \( g_{\text{max}} = \frac{1}{4} \) (dashed lines). (c) The resulting normalized ground-state wavefunctions \( \psi_k \).

For a given quantum droplet radius \( R \), ring number \( n \) and electron–hole density \( \rho_{\text{eh}} \), we fix the peak amplitude of \( g_{k,k} \) to \( g_{\text{max}} = \max[g_{k,k}] \) which defines the strength of the electron–hole correlations. This settles \( g_0 \) for any given \((R, n, \rho_{\text{eh}})\) combination. Based on the discussion following equation (28), the largest possible peak amplitude of \( g_{k,k} \) is \( \frac{1}{4} \) which yields vanishing \((1 - 2f_k)\) at the corresponding momentum.

Once \( g_0 \) produces a fixed \( g_{\text{max}} \), we only need to find which \( \kappa \) value produces the correct density for a given \((R, n)\) combination. In other words, \( \kappa \) alters \( \rho_{\text{eh}} \) because it changes the width of \( g_0 \phi_k \) whose peak amplitude is already fixed. Since we want to solve \( \tilde{E}_{\text{pers}} \) for a given \((R, n, \rho_{\text{eh}})\) combination, we solve the specific \( \kappa \) value iteratively. In more detail, we construct \( f_k \) by using \( g_0 \phi_k \) as input to equation (28) for a fixed \((R, n)\) as function of \( \kappa \). We then find iteratively which \( \kappa \) satisfies the density condition (29).

Figure 6(a) presents the normalized electron–hole pair-correlation function \( \Delta \bar{g}(r) \) for an electron–hole correlation strength of \( g_{\text{max}} = 0.24 \) (shaded area) and \( g_{\text{max}} = \frac{1}{4} \) (dashed line).
The quantum droplet has $n = 4$ rings and a radius of $R = 90.8 \text{ nm}$ indicated by a vertical line. We assume that the electron–hole density is $\rho_{\text{ch}} = 2.5 \times 10^{10} \text{ cm}^{-2}$ such that the iteration yields $\kappa = 1.1 \times 10^7 \text{ m}^{-1}$ ($\kappa = 1.7 \times 10^6 \text{ m}^{-1}$) for $g_{\text{max}} = 0.24$ ($g_{\text{max}} = \frac{1}{4}$), which settles the consistent quantum droplet configuration. We observe that $\Delta \tilde{g}(r)$ has four rings including the half oscillation close to the origin which appears due to the Coulomb attraction between electrons and holes. Additionally, the electron–hole pair-correlation function is only non-zero up to the hard shell at $r = R$, according to equation (41). By comparing the results of $g_{\text{max}} = 0.24$ and $\frac{1}{4}$, we note that the oscillation amplitude decreases slower as function of $r$ with increasing $g_{\text{max}}$ because the decay parameter $\kappa$ is smaller for elevated $g_{\text{max}}$.

The corresponding self-consistently computed electron–hole distribution $f_k$ and correlation $g_{k,k}$ are plotted in figure 6(b) as black and red lines, respectively for $g_{\text{max}} = 0.24$ (solid lines) and $g_{\text{max}} = \frac{1}{4}$ (dashed lines). The electron–hole distribution $f_k$ peaks to 0.4 (0.5) at $k = 1.3 \times 10^8 \text{ m}^{-1}$ for $g_{\text{max}} = 0.24$ ($g_{\text{max}} = \frac{1}{4}$). We see that the peak of $f_k$ sharpens as $g_{\text{max}}$ is increased. Interestingly, $f_k$ and $g_{k,k}$ show small oscillations indicated by vertical lines whose amplitude becomes larger with increasing electron–hole correlation strength.

As we compare the $f_k$ of the quantum droplets with that of the excitons (figure 4(b)), we note that quantum droplets exhibit a significant reduction of the Pauli blocking, i.e. $(1 - 2f_k)$, at small momenta. As a result, quantum droplets produce a stronger electron–hole attraction than excitons for low $k$, which makes the formation of these quasi-particle states possible once the carrier density becomes large enough. Figure 6(c) presents the corresponding normalized ground-state wavefunctions $\psi_k$. The wavefunction $\psi_k$ is qualitatively different from the state obtained for both, the degenerate Fermi gas and excitons, presented in figures 3(b) and 4(c), respectively. In particular, the quantum droplet produces a $\psi_k$ that has small oscillations for small $k$ (vertical lines) which are synchronized with the oscillations of $f_k$. Additionally, $f_k$ shows a strong dip close to the inversion $k = 1.3 \times 10^8 \text{ m}^{-1}$. The dip becomes more pronounced as $g_{\text{max}}$ is increased.

As discussed above, the largest possible peak amplitude of $g_{k,k}$ is $\frac{1}{4}$. By approaching $g_{\text{max}} = \frac{1}{4}$, the energy per excited electron–hole pair $\tilde{E}_{\text{pro}}$ decreases slightly from $\tilde{E}_{\text{pro}} = -10.12$ to $-10.14 \text{ meV}$ as $g_{\text{max}}$ is changed from 0.24 to $\frac{1}{4}$. In general, for a fixed quantum-droplet radius $R$, ring number $n$ and electron–hole density $\rho_{\text{ch}}$, we find that $\tilde{E}_{\text{pro}}$ is minimized when the amplitude of $g_{k,k}$ is maximized. Consequently, we use $g_{\text{max}} = \frac{1}{4}$ in our calculations to study the energetics of quantum droplets. For this particular case, the quantum droplet’s ground state is 3.0 meV below the exciton energy, based on the analysis in section 3.2. Therefore, the quantum droplets are quasi-particles where electron–hole pairs are stronger bound than in excitons, as concluded above.

4.1. Density dependence

The quantum droplet ansatz (41) is based on a postulated radius $R$ for the correlation bubble. Even though we find the self-consistent configuration $(f_k, g_{k,k})$ for each $R$, we still need to determine the stable quantum droplet configurations. As the main condition, the quantum droplet’s pair-excitation energy must be lower than that of the excitons and the biexcitons.

In the formation scheme of macroscopic electron–hole droplets, these droplets emerge only after a critical density is exceeded [11]. In addition, stable droplets grow in size as the overall particle density is increased. Therefore, it is reasonable to assume that also quantum droplets share these properties. We use the simplest form where the area of the quantum droplet scales
Figure 7. Energetics of quantum droplets. (a) The ground-state energy $E_0$ (black solid line), the first excited state $E_1$ (red solid line), the continuum (shaded area) and the energy per excited electron–hole pair (black dashed line) are presented as function of $\rho_{eh}$. The quantum droplet has $n = 4$ rings and $R = 90.8$ nm. The density-dependent exciton (dashed blue line) and biexciton-binding energy (dotted red line) are also plotted. (b) The corresponding result for quantum droplets with the density-dependent $R$ defined in equation (42).

Energy linearly with density. This condition connects the radius and density via

$$R = R_0 \sqrt{\frac{\rho_{eh}}{\rho_0}},$$

(42)

where $R_0$ is the radius at reference density $\rho_0$. To determine the effect of the droplet’s $\rho_{eh}$-dependent size, we also compute the quantum droplet properties for a fixed $R = R_0$. In the actual calculations, we use $R_0 = 90.8$ nm and $\rho_0 = 2.5 \times 10^{10}$ cm$^{-2}$.

In both cases, we find the fully consistent pair $(f_k, g_{k,k})$ as described in section 4 and compute the pair-excitation energy for different $\rho_{eh}$. Figure 7(a) shows the ground-state energy $E_0$ (solid black line), the first excited state $E_1$ (solid red line), the continuum (shaded area) and the energy per excited electron–hole pair (black dashed line) as function of $\rho_{eh}$ when a constant-$R$ quantum droplet has $n = 4$ rings. The corresponding result for the density-dependent $R$, defined by equation (42), is shown in figure 7(b). In both frames, the position of the density-dependent exciton (dashed blue line) and biexciton energy (dotted red line) are indicated, based on the calculation shown in figure 5 and the experimentally deduced biexciton binding energy $2.2$ meV in [29].

For both $R$ models, the quantum droplet’s pair-excitation energy $\bar{E}_{pro}$ (black dashed line) is significantly lower than both the exciton and the biexciton energy, which makes the $(n = 4)$-ring quantum droplet energetically stable for densities exceeding $\rho_{eh} = 2.5 \times 10^{10}$ cm$^{-2}$. We also see that all excited states of the quantum droplets have a higher energy than the exciton. Therefore, only the quantum droplet’s ground state is energetically stable enough to exist permanently. However, the quantum droplet state with $n = 4$ rings does not exist for an electron–hole density below $\rho_{eh} = 2.47 \times 10^{10}$ cm$^{-2}$ (vertical line) because this case corresponds to the smallest possible $\kappa = 0$. In other words, one cannot lower $\kappa$ to make $f_k$ narrower in order to produce $\rho_{eh}$ smaller than $2.47 \times 10^{10}$ cm$^{-2}$. More generally, one can compute the threshold $\rho_{eh}$ of a quantum
droplet with $n$ rings by setting $\kappa$ to zero in equation (41) and by generating the corresponding $\phi_k$, $g_{k,k}$ and $f_k$ via equation (27). Since $\phi_k$ and $f_k$ peak at $k$ that is proportional to $x_n$, it is clear that $\rho_{eh} \propto \int_0^\infty dk k f_k$ increases monotonically as function of $n$. Therefore, one finds quantum droplets with a higher ring number only at elevated densities.

4.2. Ground-state energy

To determine the quantum droplet’s binding energy, we define

$$E_{\text{bind}} \equiv \tilde{E}_{\text{pro}}(1s) - \tilde{E}_{\text{pro}}(\text{droplet}),$$

where $\tilde{E}_{\text{pro}}(1s)$ and $\tilde{E}_{\text{pro}}(\text{droplet})$ are the ground-state energies of the exciton and the quantum droplet, respectively. Figure 8 presents $E_{\text{bind}}$ for all possible ring numbers for both constant $R$ (dashed line) and $\rho_{eh}$-dependent $R$ (solid line), as function of $\rho_{eh}$. Here, we follow the lowest $E_{\text{bind}}$ among all $n$-ring states as the ground state of the quantum droplet. As explained in section 4.1, each $n$-ring state appears as an individual threshold density is crossed. The horizontal line indicates the binding energy of the biexciton. We see that both droplet-radius configurations produce discrete energy bands. As the electron–hole density is increased, new energy levels appear as sharp transitions. Each transition increases the ring number $n$ by one such that the ring number directly defines the quantum number for the discrete energy levels. We see that only quantum droplets with more or equal than four rings have a larger binding than biexcitons do, making one-, two- and three-ring quantum droplets instable. The constant $R$ and the density-dependent $R$ produce a qualitatively similar energy structure. As main differences, the constant $R$ produces ring-to-ring transitions at higher densities and the energy bands spread to a wider energy range. For example, the energy range of the $n = 4$ energy band is $[3.0, 3.8]$ meV for constant $R$ while it is $[3.0, 3.2]$ meV for the density-dependent $R$. In general, the actual stable droplet configuration has to be determined by experiments. Since the

Figure 8. Ground-state energy of quantum droplets. The ground-state energy is presented as function of $\rho_{eh}$ for a constant (dashed line) and density-dependent $R$ (solid line). The biexciton-binding energy is indicated by the horizontal line.

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Figure 9. Properties of quantum droplets. (a) The ground-state energy (solid line) is presented as function of $\rho_{\text{eh}}$ for the density-dependent $R$. The dashed lines denote excited states and the biexciton-binding energy is marked by the horizontal line. (b) Number of correlated electron–hole pairs within the $j$th ring as function $\rho_{\text{eh}}$ from the first ($j=1$) up to the fifth ($j=5$) ring. (c) The electron–hole pair-correlation function $r\Delta g(r)$ is shown before (shaded area) and after (solid line) the four-to-five-ring droplet transition. These cases are indicated by circles in frame (a).

density-dependent droplet radius is consistent with the properties of macroscopic electron–hole droplets, we use equation (42) to study the properties of quantum droplets.

Figure 9(a) shows again the ground-state energy of the quantum droplet as function of electron–hole density $\rho_{\text{eh}}$ for the density-dependent $R$. The dashed lines continue the energy levels after the next higher quantum droplet state becomes the ground state. The biexciton-binding energy is indicated by a horizontal line. We see that the binding energy of the unstable ($n=3$)-liquid state remains smaller than the biexciton-binding energy even at elevated $\rho_{\text{eh}}$ making it instable at all densities. In contrast to that, $E_{\text{bind}}$ of the ($n=4$)- and ($n=5$)-liquid
state is stronger than the biexciton value while it remains relatively stable as the electron–hole density is increased.

4.3. Ring structure of quantum droplets

We also can analyze the number of correlated electron–hole pairs within the jth ring of the quantum droplet. Since \( S_{\text{drop}} \int d^2r \Delta g(r) = S_{\text{drop}} 2\pi \int dr r \Delta g(r) \) defines the total number of correlated pairs [15]:

\[
\Delta N_j = S_{\text{drop}} 2\pi \int_{x_{j-1}}^{x_j} dr r \Delta g(r) \tag{44}
\]

is the number of correlated pairs within the jth ring when \( S_{\text{drop}} = \pi R^2 \) is the area of the quantum droplet. Figure 9(b) shows \( \Delta N_j \) as function of \( \rho_{\text{eh}} \) from the first up to the fifth ring. We see that the number of electron–hole pairs within the innermost rings becomes larger, while it decreases within the outermost rings, as \( \rho_{\text{eh}} \) is made larger. Interestingly, each ring has roughly the same number of electron–hole pairs after the n-ring droplet has become the ground state via a sharp transition, compare with figure 9(a). More precisely, \( \Delta N_j \) is close to one such that the nth quantum droplet state has about n electron–hole pairs after the transition. Consequently, the n-ring quantum droplet has roughly n electron–hole pairs. Therefore, already the first stable quantum droplet with \( n = 4 \) rings has four correlated electrons and holes showing that it is a highly correlated quasi-particle. As derived in appendix E, one can solve analytically that for ring numbers up to \( n = 3 \) the nth quantum droplet state has very close n correlated electron–hole pairs while the ratio \( \Delta N/n \) converges toward 1.2 for a very large ring number.

Figure 9(c) presents examples for the electron–hole pair-correlation function \( r \Delta \bar{g}(r) \) before (shaded area) and after (solid line) the four-to-five-ring droplet transition. The corresponding binding energies and electron–hole densities are indicated with circles in figure 9(a). Before the transition, the oscillation amplitude of \( r \Delta \bar{g}(r) \) decreases as function of \( r \) while after the transition the oscillation amplitude stays almost constant indicating that the decay parameter \( \kappa \) is close to zero, just after the transition. This is consistent with our earlier observation that a n-ring quantum droplet emerges only above a threshold density matching the density of the \( \kappa = 0 \) state.

5. Influence of electron–electron and hole–hole correlations

So far, we have analyzed the properties of quantum droplets without electron–electron and hole–hole correlations based on the assumption that electron–hole correlations dominate the energetics. We will next show that this scenario is plausible also in dense interacting electron–hole systems. We start by reorganizing the carrier–carrier correlations \( c_{\lambda,\lambda;\lambda,\lambda}^{q,k,k} \), defined in equation (19), into \( \Delta(a_{\lambda,\lambda;\lambda,\lambda}^{\uparrow} a_{\lambda,\lambda;\lambda,\lambda}^{\uparrow} a_{\lambda,-\lambda;\lambda,-\lambda} a_{\lambda,-\lambda;\lambda,-\lambda}) \) using \( k = K + p \), \( k' = K - p \) and \( q = p - p' \). In this form, we see that two annihilation (or creation) operators assign a correlated carrier pair that has a center-of-mass momentum of \( 2\hbar K \). Like for electron–hole correlations, we concentrate on the case where the center-of-mass momentum of the correlated pairs vanishes

\[
\Delta(a_{\lambda,\lambda;\lambda,\lambda}^{\uparrow} a_{\lambda,\lambda;\lambda,\lambda}^{\uparrow} a_{\lambda,-\lambda;\lambda,-\lambda} a_{\lambda,-\lambda;\lambda,-\lambda}) \equiv -\delta_{K,0} F_{p,p}^{\lambda} \quad \Leftrightarrow \quad c_{\lambda,\lambda;\lambda,\lambda}^{q,k,k} = -\delta_{k,-k} F_{k,k-q}^{\lambda} \tag{45}
\]

that follows from a straightforward substitution \( K = \frac{1}{2}(k + k') \), \( p = \frac{1}{2}(k - k') \) and \( p' = \frac{1}{2}(k - k') - q \). Since the transformations \( p \rightarrow -p \) and \( p' \rightarrow -p' \) correspond to exchanging creation
and annihilation operators in $c_{\lambda,\lambda',\lambda',\lambda}$, respectively, the $F_{\lambda,p,p'}$ function must change its sign with these transformations due to the Fermionic antisymmetry. In other words, $F_{\lambda,p,p'}$ must satisfy
\[ F_{-\lambda,p,-p'} = F_{\lambda,-p,-p'} = -F_{\lambda,p,p'} = -F_{-\lambda,-p,-p'}, \quad (46) \]
when the sign of the momentum is changed.

Like for electron–hole correlations, carrier–carrier effects can be described through the corresponding pair-correlation function
\[ g_{\lambda}(r) \equiv \langle \Psi_{\lambda}^\dagger(r) \Psi_{\lambda}^\dagger(0) \Psi_{\lambda}(r) \Psi_{\lambda}(0) \rangle = \rho_{\lambda}^2 - f_{\lambda}^2(r) + \Delta g_{\lambda}(r), \quad (47) \]
\[ f_{\lambda}(r) \equiv \frac{1}{S} \sum_k f_k^\lambda e^{-ik \cdot r} \quad \text{with} \quad \lambda = e, h, \quad (48) \]
where we have applied homogeneous conditions, used the definition (3), and introduced $f_{\lambda}(r)$ as the Fourier transformation of $f_{\lambda}^k$. The first term describes again a plasma contribution analogously to the first part in the electron–hole pair-correlation function (22). The correlated contribution is defined by
\[ \Delta g_{\lambda}(r) \equiv \frac{1}{S^2} \sum_{K,p,p'} \Delta \langle a_{\lambda,K+p}^\dagger a_{\lambda,K-p}^\dagger a_{\lambda,K-p'} a_{\lambda,K+p'} \rangle e^{-i(p-p') \cdot r} = - \frac{1}{S^2} \sum_{p,p'} F_{\lambda,p,p'}^\lambda e^{-i(p-p') \cdot r}, \quad (49) \]
where we have applied the condition (45). We note that $\Delta g_{\lambda}(r)$ vanishes at $r = 0$ due to the Pauli-exclusion principle among Fermions, enforced by equation (46).

Due to the conservation law (26), the electron and hole distributions $f_k^e$ and $f_k^h$ become different only when the electron–electron and hole–hole correlations are different. To study how the carrier–carrier correlations modify the overall energetics, we assume identical electron–electron and hole–hole correlations $F_{\lambda,p,p'}^e = F_{\lambda,p,p'}^h$ to simplify the book-keeping. With this choice, equations (26) and (45) imply identical distributions that satisfy
\[ (f_k - \frac{1}{2})^2 + g_{k,k} + F_{k,k} = \frac{1}{4}, \quad F_{k,k} \equiv F_{k,k}^e = F_{k,k}^h. \quad (50) \]
We see that also carrier–carrier correlations modify $f_k$ via a diagonal $F_{k,k}$, just like $g_{k,k}$.

In the same way, the generalized Wannier equation (35) is modified through the presence of carrier–carrier correlations in the form of equation (46). By inserting equations (45) and (50) into equation (35), the original $E_k$ and $V_{k,k'}^{\text{eff}}$ can simply be replaced by
\[ E_k = \left[ \frac{\hbar^2 k^2}{2\mu} - 2 \sum_{k'} V_{k-k'} f_k \right] (1 - 2f_k) + 2 \sum_{k'} V_{k-k'} \left( g_{k,k'} + F_{k,k} \right), \quad (51) \]
\[ V_{k,k'}^{\text{eff}} \equiv (1 - 2f_k) V_{k-k'} (1 - 2f_{k'}) + 2 \left( g_{k,k'} + F_{k,k} \right) V_{k-k'}, \quad (52) \]
to fully account for the carrier–carrier contributions.

As a general property, the repulsive Coulomb interaction tends to extend the $r$-range where the presence of multiple carriers is Pauli blocked. In other words, carrier–carrier correlations build up to form a correlation hole to $g_{\lambda}(r)$. To describe this principle effect, we use an ansatz
\[ F_{k,k'} \equiv F_0 \cos(\theta_k - \theta_{k'}) e^{-\xi(|k|-|k'|)} \quad (53) \]
that satisfies the antisymmetry relations (46). The strength of the correlation is determined by \( F_0 \) and \( l_c \) corresponds to a correlation length. As equation (53) is inserted to equation (49), a straightforward integration yields

\[
\Delta g_s(r) = -\frac{F_0^2}{(2\pi)^2} \frac{r^2}{(l_c^2 + r^2)^3},
\]

which is rotational symmetric and vanishes at \( r = 0 \), as it should for homogeneous Fermions.

To compute the quasi-particle energetics with carrier–carrier correlations, we use the same quantum droplet state (41) as computed for vanishing carrier–carrier correlations in section 4, i.e. we keep the quantum droplet radius \( R \), ring number \( n \) and decay parameter \( \kappa \) unchanged. For a given combination \((F_0, l_c)\), we then adjust the strength of the electron–hole correlations \( g_0 \) such that \( g_{k,k} + F_{k,k} \) is maximized, i.e. \( \max[g_{k,k} + F_{k,k}] = \frac{1}{4} \), according to equation (50). In analogy to section 4, this yields a vanishing \((1 - 2F_k)\) at one momentum state. Since \( F_{k,k} \) is positive, the presence of carrier–carrier correlations must be compensated by reducing the magnitude of the electron–hole correlations \( g_{k,k} \). Additionally, equation (50) modifies the electron–hole distribution \( f_k \) and the electron–hole density in comparison to the case with vanishing \( F_{k,k} \).

Figure 10(a) shows the normalized electron–hole pair-correlation function \( \Delta \tilde{g}(r) \) for vanishing carrier–carrier correlations \((F_0 = 0, \text{black line})\). The vertical lines indicate the maxima of \( \Delta \tilde{g}(r) \) identifying the centers of the liquid-state rings. The quantum droplet state has a radius of \( R = 90.8 \) nm, \( n = 4 \) rings and an electron–hole density of \( \rho_{eh} = 2.5 \times 10^{10} \text{cm}^{-2} \). The corresponding result for non-vanishing carrier–carrier correlations with \( F_0 = 0.3 \) and \( l_c = 12.5 \) nm is plotted as red line. The presence of carrier–carrier correlations increases the electron–hole density to \( \rho_{eh} = 2.7 \times 10^{10} \text{cm}^{-2} \) due to the normalization procedure described above. We see that the presence of carrier–carrier correlations reduces the amplitude of the ring-state oscillations in \( \Delta \tilde{g}(r) \) only slightly. This suggests that carrier–carrier correlations play a minor role in the build up of electron–hole correlations in quantum droplets.

The corresponding normalized carrier–carrier pair-correlation function \( \tilde{g}_s(r) \equiv g_s(r)/\rho_{eh}^2 \) is presented in figure 10(b) without \((F_0 = 0, \text{black line})\) and with \((F_0 = 0.3, \text{red line})\) carrier–carrier correlations. Additionally, the pure correlated contribution \(- \Delta \tilde{g}_s(r) \equiv -\Delta g_s(r)/\rho_{eh}^2 \) for \( F_0 = 0.3 \) is shown as a shaded area. Even without carrier–carrier correlations, \( \tilde{g}_s(r) \) shows a range of Pauli blocked carriers at short distances followed by the Friedel oscillations [33]. Interestingly, \( \tilde{g}_s(r) \) dips at exactly the same positions where \( \Delta \tilde{g}(r) \) peaks indicated by vertical lines in figure 10. Consequently, the carriers try to avoid each other within the rings of the quantum droplets, which is clearly related to the Fermion character of electrons. We observe that the presence of \( \Delta \tilde{g}_s(r) \) increases the range of Pauli-blocked carriers. To show the range of Pauli blocking, the inset of figure 10(b) plots the same data up to the first Friedel oscillation \( r = 26.6 \) nm. To quantify Pauli blocking, we determine the half-width value where \( g_s(r_{1/2}) = \frac{1}{2} \rho_{eh}^2 \). We find that \( r_{1/2} \) increases from 8.8 nm for \( F_0 = 0 \) to 11.2 nm for \( F_0 = 0.3 \), i.e. the correlation hole increases the range of Pauli blocking by roughly 27% which is significant.

In the next step, we compute the ground-state energy of particle excitations from the generalized Wannier equation (35) with the \( f_k, g_{k,k} \) and \( F_{k,k} \) entries (51) and (52). The actual energy per excited particle follows from equation (37) and this is compared against the exciton binding deduced as in section 3.2. The results produce a quantum droplet energy that grows from 2.99 to 3.08 meV as the carrier–carrier correlations are included. The small increase shows that the correlated arrangement of the carriers saves energy. However, carrier–carrier correlations
change the quantum droplet binding only by 3.3%, for the studied case. In other words, even a large correlation hole \( \Delta g_\lambda(r) \) cannot affect much the energetics of the quantum droplet, which justifies the assumption of neglecting carrier–carrier correlations for quantum droplets.

6. Discussion

We have developed a systematic method to compute the pair-excitation energetics of many-body states based on the correlation-function formulation of quasi-particles. In particular, we have generalized the Wannier equation to compute the energy per excited electron–hole pair of a many-body state probed by a weak pair excitation of a quasi-particle. As an unconventional aspect, we determine the many-body state via the pair-correlation function \( g(r) \) and work out the lower-order expectation values self-consistently, based on \( g(r) \), not the other way around. As a major benefit, \( g(r) \) characterizes the many-body state and its energetics, which allows us to identify the properties of different quasi-particles directly.

We have applied the scheme to study especially the energetics and properties of quantum droplets as a new quasi-particle. Our computations show that the pair-excitation energetics of quantum droplets has discrete bands that appear as sharp transitions. Additionally, each ring
contains roughly one electron–hole pair and only quantum droplets with more than four rings, i.e. electron–hole pairs are stable. We also show that the energy structure of quantum droplets originates dominantly from electron–hole correlations because the carrier–carrier correlations increase the binding energy of quantum droplets only slightly.

We also have checked that having a soft wall for the quantum droplet changes the energetics only slightly. More specifically, we allowed the pair-correlation function to tunnel into the wall region where it decayed exponentially, as is characteristic for a soft wall. For tunneling depths of 5 nm, the binding energy of the electron–hole droplet decreases only by 1%. Therefore, the explicit nature of the electron–hole confinement does not play a significant role for quantum droplets.

The developed method can be used more generally to determine the characteristic quasi-particle energies based on the correlation function. As further examples, we successfully analyze the energetics of the degenerate Fermi gas and high-density excitons. We also have extended the method to analyze coherent quasi-particles. As possible new directions, one can study different pair-excitation schemes to analyze the role of, e.g. spin. In this connection, one expects to detect bonding and antibonding branches for quasi-particles such as biexcitons. In general, the approach is limited only by the user’s knowledge of the pair-correlation function. It also might be interesting to develop the approach to the direction where quasi-particles are identified via \( N \)-particle correlations to systematically analyze how the details of highly correlated states affect the excitation energetics and the response in general.

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**Appendix A. Connection of correlations and expectation values**

We first analyze a normally ordered \((N+1)\)-particle expectation value

\[
\langle \hat{N} + 1 \rangle \equiv \langle a_{\lambda_1, k_1}^{\dagger} \ldots a_{\lambda_N, k_N}^{\dagger} \hat{N}_{\text{tot}} a_{\lambda_N, k_N} \ldots a_{\lambda_1, k_1} \rangle
\]  

that contains the total number operator \( \hat{N}_{\text{tot}} \equiv \sum_{k, \lambda} a_{\lambda, k}^{\dagger} a_{\lambda, k} \). Since \( \hat{N}_{\text{tot}} \) contains all electronic states, it produces

\[
\hat{N}_{\text{tot}} \hat{\rho}_N = N \hat{\rho}_N
\]

for all states \( \hat{\rho}_N \) containing \( N \) carriers within all bands of the system. Since we may consider only cases where the total number of carriers is conserved, we may limit the analysis to the states \( \hat{\rho}_N \) from here on.

By applying the commutator relation \([\hat{N}_{\text{tot}}, a_{\lambda, k}]_− = −a_{\lambda, k} \) \( N \) times, equation (A.1) becomes

\[
\langle \hat{N} + 1 \rangle = −N \langle \hat{O}_N \rangle + \langle \hat{O}_N \hat{N}_{\text{tot}} \rangle \quad \text{with} \quad \hat{O}_N \equiv a_{\lambda_1, k_1}^{\dagger} \ldots a_{\lambda_N, k_N}^{\dagger} a_{\lambda_N, k_N} \ldots a_{\lambda_1, k_1}.
\]

Using the property (A.2), we find

\[
\langle \hat{O}_N \hat{N}_{\text{tot}} \rangle = \text{Tr}[\hat{O}_N \hat{N}_{\text{tot}} \hat{\rho}_N] = \text{Tr}[\hat{O}_N \hat{N} \hat{\rho}_N] = N \langle \hat{O}_N \rangle.
\]
By combining the result (A.4) with (A.1) and (A.3), we obtain a general reduction formula [34]

\[ \sum_{k',k}^\prime \langle a_{\lambda, k_1}^\dagger \ldots a_{\lambda', k_n}^\dagger a_{\lambda, k_1} a_{\lambda', k_2} \ldots a_{\lambda, k_1} \rangle = (N - N) \langle \hat{O}_N \rangle \] (A.5)

that directly connects \( N \) and \( (N + 1) \)-particle expectation values.

For \( N = 1 \), equation (A.5) becomes

\[ \sum_{k',k}^\prime \langle a_{\lambda, k}^\dagger a_{\lambda', k}^\dagger a_{\lambda, k} a_{\lambda', k} \rangle = (N - 1) \langle a_{\lambda, k}^\dagger a_{\lambda, k} \rangle . \] (A.6)

We then express the two-particle contribution exactly in terms of the Hartree–Fock factorization [1] and the two-particle correlations (19) and assume homogeneous conditions where all coherences vanish. By using a two-band model, equation (A.6) yields then

\[ \left( f_k^e - \frac{1}{2} \right)^2 + \sum_{k'} (c_{\text{eh}}^{k,k',k} - c_{\text{cc}}^{0,k',k}) = \frac{1}{4}, \] (A.7)

\[ \left( f_k^h - \frac{1}{2} \right)^2 + \sum_{k'} (c_{\text{eh}}^{k,k,k'} - c_{\text{vv}}^{0,k',k}) = \frac{1}{4} \]

for electrons (\( \lambda = c \)) and holes (\( \lambda = v \)), respectively. With the help of equation (21), equation (A.7) casts into the form

\[ \left( f_k^e - \frac{1}{2} \right)^2 + g_{k,k} - \sum_{k'} c_{\text{cc}}^{0,k',k} = \frac{1}{4}, \quad \left( f_k^h - \frac{1}{2} \right)^2 + g_{k,k} - \sum_{k'} c_{\text{vv}}^{0,k',k} = \frac{1}{4} \] (A.8)

that connects the density distributions with the pair-wise correlations.

### Appendix B. Probe-induced quantities

To compute the probe-induced electron–hole density and polarization, we use the following general properties of the displacement operator (7) [1, 15]:

\[ D^\dagger [\psi] a_{\nu, k} D[\psi] = \cos(\epsilon \psi_k) a_{\nu, k} - e^{-i\psi_k} \sin(\epsilon \psi_k) a_{\nu, k}, \] (B.1)

\[ D^\dagger [\psi] a_{\nu, k} D[\psi] = \cos(\epsilon \psi_k) a_{\nu, k} + e^{i\psi_k} \sin(\epsilon \psi_k) a_{\nu, k}. \]

Transformation (B.1) allows us to construct the density- and polarization-induced pair excitations exactly. More specifically, we start from the expectation value

\[ \langle a_{\lambda, k}^\dagger a_{\lambda', k} \rangle_\psi \equiv \text{Tr} \left[ a_{\lambda, k}^\dagger a_{\lambda', k} \hat{D} [\psi] \hat{\rho}_{\text{MB}} \hat{D}^\dagger [\psi] \right] = \text{Tr} \left[ \hat{D}^\dagger [\psi] a_{\lambda, k}^\dagger \hat{D} [\psi] \hat{D}^\dagger [\psi] a_{\lambda', k} \hat{D} [\psi] \hat{\rho}_{\text{MB}} \right], \] (B.2)

where we have utilized cyclic permutations under the trace and the unitary of the displacement operator (7).
To compute the pair-excitation energy, we have to compute how all those single-particle expectation values and two-particle correlations that appear in equation (18) are modified by the pair excitation. By inserting transformation (B.1) into equation (B.2), we can express any modified single-particle expectation value in terms of \( \varepsilon, \psi_k \) and \( \vec{f}_k \). The change in density and polarization becomes then

\[
f_k,\psi = \langle a_{\kappa,k}^\dagger a_{\kappa,k} \rangle_\psi - f_k^e + f_k^h = 2 \sin^2(\varepsilon |\psi_k|) (1 - f_k^e - f_k^h),
\]

\[
P_k,\psi = \langle a_{\kappa,k}^\dagger a_{\kappa,k} \rangle_\psi = e^{\imath \varepsilon k} \sin(\varepsilon |\psi_k|) \cos(\varepsilon |\psi_k|) (1 - f_k^e - f_k^h),
\]

respectively. Since the many-body state \( \hat{\rho}_{\text{MB}} \) is probed by a weak laser pulse, we apply the weak-excitation limit \( \varepsilon \ll 1 \), producing

\[
P_k,\psi = (1 - f_k^e - f_k^h) \varepsilon |\psi_k|^2 + \mathcal{O}(\varepsilon^3), \quad f_k,\psi = (1 - f_k^e - f_k^h) \varepsilon^2 |\psi_k|^2 + \mathcal{O}(\varepsilon^3)
\]

to the leading order.

Following the same derivation steps as above, we find that the pair excitations change the electron–hole correlation by

\[
c_{e_{\chi},\psi}^{q,k,k'} = \Delta (a_{\kappa,k}^\dagger a_{\kappa,k}^{\dagger} a_{q,k'} q a_{q,k}-q) \psi - c_{e_{\chi}}
\]

\[
= \varepsilon \left[ c_{v,v,c}^{q,k,k'} \psi_k^* + \left( c_{v,v,c}^{q-k,q-k} \right)^* \psi_{k+q}^* - c_{v,v,c,c}^{q,k,k'} \psi_{k-q}^* - \left( c_{v,v,c,c}^{q-k-k',q-k+q} \right)^* \psi_k^* \right]
\]

\[
+ \varepsilon^2 \left[ c_{e_{\chi}}^{q-k-k',q-k,k} \psi_{k+q}^* \psi_{k-q}^* + \left( c_{e_{\chi}}^{q-k+k',q-k+q} \right)^* \psi_k^* \psi_k^* \right]
\]

\[
- \frac{1}{2} c_{v,v,c}^{q,k,k'} (|\psi_k|^2 + |\psi_k|^2 + |\psi_{k+q}|^2 + |\psi_{k-q}|^2) + c_{v,v,c,c}^{q,k,k',q-k+q} \psi_{k+q}^* \psi_k^* + c_{v,v,v,v}^{q,k,k'} \psi_{k+q}^* \psi_k^* \psi_k^* \psi_{k-q}^*
\]

\[
- c_{v,v,c,c}^{q,k,k'} \psi_k^* \psi_{k+q}^* - \left( c_{v,v,c,c}^{q-k-k',q-k+q} \right)^* \psi_k^* \psi_{k+q}^* \right] + \mathcal{O}(\varepsilon^3)
\]

(B.5)

out of the initial many-body correlation \( c_{e_{\chi},\psi}^{q,k,k'} \). Besides the correlations (19), equation (B.5) contains also coherent two-particle correlations:

\[
c_{v,v,c,c}^{q,k,k'} \equiv \Delta (a_{\kappa,k}^\dagger a_{\kappa,k}^{\dagger} a_{q,k'} q a_{q,k}-q), \quad c_{v,v,c,c}^{q,k,k'} \equiv \Delta (a_{\kappa,k}^\dagger a_{\kappa,k}^{\dagger} a_{q,k'} q a_{q,k}-q),
\]

\[
c_{v,v,c,c}^{q,k,k'} \equiv \Delta (a_{\kappa,k}^\dagger a_{\kappa,k}^{\dagger} a_{q,k'} q a_{q,k}-q).
\]

(B.6)

From these, \( c_{v,v,c,c}^{q,k,k'} \) and \( c_{v,v,v,v}^{q,k,k'} \) describe correlations between polarization and density while \( c_{v,v,c,c}^{q,k,k'} \) corresponds to the coherent biexciton amplitude. Therefore, also the coherent two-particle correlations (B.6) contribute to the pair-excitation spectroscopy even though they do not influence the initial many-body energy (18). The remaining \( c_{v,v,c,c}^{q,k,k'} \) and \( c_{v,v,v,v}^{q,k,k'} \) transform analogously. With the help of equations (B.4)–(B.5) we can then construct exactly the energy change (32) induced by the pair-wise excitations.
Appendix C. Generalized Wannier equation with coherences

As the exact relations (B.4) and (B.5) are inserted to the system energy (32), we obtain the pair-excitation energy exactly

\[ E_{\text{pro}}[\psi] = E_{\text{pro}}^{\text{coh}}[\psi] + E_{\text{pro}}^{\text{inc}}[\psi] + O(\varepsilon^3), \]

\[ E_{\text{pro}}^{\text{coh}} = 2\varepsilon \sum_{k} \left[ \bar{E}_k \text{Re}[P_k \psi_k^*] - \sum_{k'} V_{k-k'} \left( 1 - f_k^c - f_{k'}^h \right) \text{Re}[P_k \psi_{k'}^*] + \text{Re}[\Gamma_k \psi_k^*] \right] \]

\[ -2\varepsilon^2 \sum_{k,k'} V_{k-k'} \left( \text{Re}[P_k P_{k'} \psi_k^* \psi_{k'}^*] - \text{Re}[P_k P_{k'}^*] \right) |\psi_k|^2 + \text{Re}[P_k P_{k'}^* \psi_k^* \psi_{k'}^*] \]

\[ + \varepsilon^2 \sum_{k,k',q} V_q \text{Re} \left[ \left( c_{q,k',q} + c_{q,k',k}^* - 2c_{q,k',k} \right) \psi_k^* \psi_{k'}^* \right], \]

\[ E_{\text{pro}}^{\text{inc}} = \varepsilon^2 \sum_{k} \bar{E}_k |\psi_k|^2 - \varepsilon^2 \sum_{k,k'} V_{\text{eff}}^{k,k'} \psi_k \psi_{k'}^* + \varepsilon^2 \sum_{k,k',q} V_q \left( c_{q,k',k}^* \psi_k \psi_{k'}^* \psi_{k-q}^* + c_{q,k',k} \psi_{k-q} \psi_{k'} \psi_k^* \right), \]

where we have divided \( E_{\text{pro}}[\psi] \) into coherent (coh) and incoherent (inc) contributions. The coherent contribution \( E_{\text{pro}}^{\text{coh}}[\psi] \) includes

\[ \Gamma_k \equiv \sum_{k',q,\lambda} V_q \left[ c_{q,k',k}^* \left( c_{\lambda,\lambda,\lambda} - c_{\lambda,\lambda,\lambda}^* \right) \right] \]

that is exactly the same as the microscopically described Coulomb scattering term in the semiconductor Bloch equations [15]. The incoherent part \( E_{\text{pro}}^{\text{inc}}[\psi] \) and the coherent energy contain different renormalized kinetic energies

\[ \bar{E}_k \equiv \bar{E}_k \left( 1 - f_k^c - f_k^h \right) + \sum_{k',q} V_q \text{Re} \left[ c_{q,k',k}^* + c_{q,k',k} \right] + \sum_{k',q} V_{k'+q-k} \text{Re} \left[ c_{\lambda,\lambda,\lambda}^* \right] + \text{Re} \left[ c_{\lambda,\lambda,\lambda} \right], \]

\[ \bar{E}_k = \frac{\hbar^2 k^2}{2\mu} - \sum_{k'} V_{k-k'} \left( f_k^c + f_k^h \right), \]

respectively. We also have identified the effective Coulomb matrix element

\[ \tilde{V}_{k,k'}^{\text{eff}} = \left( 1 - f_k^c - f_k^h \right) V_{k-k'} \left( 1 - f_{k'}^c - f_{k'}^h \right) - \sum_{k',q} V_{k-k'} \left( c_{\lambda,\lambda,\lambda} - c_{\lambda,\lambda,\lambda}^* \right) \]

\[ - \sum_{k',q} V_q \left( c_{\lambda,\lambda,\lambda}^* - c_{\lambda,\lambda,\lambda} - c_{\lambda,\lambda,\lambda}^* - c_{\lambda,\lambda,\lambda} \right), \]

that contains the unscreened Coulomb interaction together with the phase-space filling contribution \( 1 - f_k^c - f_k^h \) and electron–hole correlations \( c_{\lambda,\lambda,\lambda} \).

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We then minimize the energy functional (C.1) as described in section 2 to find a condition for the ground-state excitations. As a result, we obtain

\[ s_{\text{coh}} + \varepsilon E_{\text{coh}}[\psi] + \varepsilon E_{\text{inc}}[\psi] = \varepsilon E_{\lambda}\psi_k, \]

\[ s_{\text{coh}} \equiv \tilde{E}_k P_k - (1 - f_k^c - f_k^h) \sum_{k'} V_{k-k'} P'_{k'} + \Gamma_k, \]

\[ E_{\text{coh}}[\psi] = 2 \sum_{k'} V_{k-k'} \left( P_k P_k \psi_k^* + \text{Re}[P_k P_k^*] \psi_k \right) - 2 \sum_{k'} V_{k-k'} \left( P_k P_k \psi_k^* + P_k P_k^* \psi_k \right) \]

\[ + \sum_{k',q} V_q \left( c_{q,k-k+q} - c_{q,k-k-q} \right) \left( \psi_k^* - \psi_{k+q}^* \right), \]

\[ E_{\text{inc}}[\psi] \equiv \tilde{E}_k \psi_k - \sum_{k'} \tilde{V}_{k,k'} \psi_k + \sum_{k',q} V_q \left( c_{q,k-k+q} \psi_k + c_{q,k-k-q} \psi_k \right). \]

We see that the presence of coherences generates the coherent source term \( s_{\text{coh}} \) to the generalized Wannier equation which is the dominant contribution in equation (C.5). However, since \( s_{\text{coh}} \) corresponds exactly to the homogeneous part of the semiconductor Bloch equations \([15]\), it vanishes for stationary \( P_k \). Therefore, the ground state of excitation must satisfy the generalized Wannier equation

\[ E_{\text{coh}}[\psi] + E_{\text{inc}}[\psi] = E_{\lambda}\psi_k. \]

In the main part, we analyze the pair excitations of incoherent many-body systems such that \( E_{\text{coh}}[\psi] \) is not present.

**Appendix D. Self-consistent exciton solver**

To find the wavefunction \( \phi_{1s,k} \) and the electron–hole distribution \( f_k \) that satisfy the ordinary density-dependent Wannier equation (40) and the conservation law (27), we define a gap equation as in \([35]\)

\[ \Delta_k \equiv \sum_{k'} V_{k-k'} \phi_{1s,k'}, \quad \epsilon_k \equiv \frac{1}{2} \left( \tilde{E}_k - E_{1s} \right), \quad \Omega_k = \sqrt{\epsilon_k^2 + \Delta_k^2}. \]  

(D.1)

As a result, we obtain the integral equations

\[ \phi_{1s,k} = \frac{1}{2} \frac{\Delta_k}{\Omega_k}, \quad f_k = \frac{1}{2} \left( 1 - \frac{\epsilon_k}{\Omega_k} \right) \]

(D.2)

which simultaneously satisfy the ordinary density-dependent Wannier equation (40) and the conservation law (27). Equations (D.1) and (D.2) are solved numerically by using the iteration steps

\[ \Delta_k^{(n+1)} = \sum_{k'} V_{k-k'} \phi_{1s,k'}^{(n)}, \quad \epsilon_k^{(n+1)} = \frac{1}{2} \left( \frac{\hbar^2 k^2}{2\mu} - E_{1s} \right), \]

\[ \Omega_k^{(n+1)} = \sqrt{(\epsilon_k^{(n+1)})^2 + (\Delta_k^{(n+1)})^2}, \]

\[ \phi_{1s,k}^{(n+1)} = \frac{1}{2} \frac{\Delta_k^{(n+1)}}{\Omega_k^{(n+1)}}, \quad f_k^{(n+1)} = \frac{1}{2} \left( 1 - \frac{\epsilon_k^{(n+1)}}{\Omega_k^{(n+1)}} \right). \]

(D.3)

One typically needs 40 iteration steps to reach convergence.
Appendix E. Number of correlated electron–hole pairs within droplet

To compute the number of correlated pairs within the droplet close to the transition, we start from the quantum droplet pair-correlation function defined by (41). Since the decay constant $\kappa$ is negligible small after each transition, see section 4.2, we set $\kappa = 0$ in equation (41), yielding

$$\phi(\mathbf{r}) = J_0\left(x_n \frac{R}{r}\right) \theta(R - r). \quad (E.1)$$

The correlated electron–hole density is then given by

$$\Delta n \equiv \iint d^2r \Delta g(\mathbf{r}) = 2\pi g_0^2 \int_0^R dr \ r J_0(x_n \frac{R}{r})^2 = \pi g_0^2 R^2 [J_1(x_n)]^2, \quad (E.2)$$

where we have introduced polar coordinates and used the properties of the Bessel functions in the last step.

To determine the parameter $g_0$ as function of the ring number $n$ and the droplet radius $R$, we compute the Fourier transformation of $g_0 \phi(\mathbf{r})$, producing

$$g_0 \phi_k = g_0 \int d^2r \ \phi(\mathbf{r}) e^{-i k \cdot r} = 2\pi g_0 \int_0^R dr \ r J_0(kr) J_0\left(x_n \frac{R}{r}\right), \quad (E.3)$$

where we have again introduced polar coordinates and identified $J_0(kr) = 2\pi \int_0^{2\pi} d\theta \ e^{i k r \cos \theta}$ [36]. For a maximally excited quantum droplet state, the maximum of $g_0 \phi_k$ is $\max[g_0 \phi_k] = \frac{1}{2}$, based on the discussion in section 4. At the same time, the integral in equation (E.3) is maximized for $k = x_n / R$. By applying the orthogonality of Bessel functions, we obtain

$$\max[g_0 \phi_k] = \pi g_0 R^2 [J_1(x_n)]^2 = \frac{1}{2}, \quad (E.4)$$

such that $g_0$ can be written as

$$g_0 = \left[2\pi R^2 [J_1(x_n)]^2\right]^{-1}. \quad (E.5)$$

By inserting equation (E.5) into equation (E.2) and multiplication of $\Delta n$ with the droplet area $S_{\text{drop}} \equiv \pi R^2$, the number of correlated pairs within the droplet close to the transition becomes

$$\Delta N \equiv \pi R^2 \Delta n = \frac{1}{4[J_1(x_n)]^2}. \quad (E.6)$$

This formula predicts that quantum droplets contain $\Delta N = 3.4, 4.6$ and $5.9$ correlated electron–hole pairs for $n = 3, 4$ and $5$ rings, respectively. For ring numbers larger than $n = 10$, $\Delta N$ approaches $1.2 \ n$.

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