Theory of exciton recombination from the magnetically induced Wigner crystal

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(Received 17 August 1995)

We study the theory of itinerant-hole photoluminescence of two-dimensional electron systems in the regime of the magnetically induced Wigner crystal. We show that the exciton recombination transition develops structure related to the presence of the Wigner crystal. The form of this structure depends strongly on the separation d between the photoexcited hole and the plane of the two-dimensional electron gas. When d is small compared to the magnetic length, additional peaks appear in the spectrum due to the recombination of exciton states with wave vectors equal to the reciprocal lattice vectors of the crystal. For d larger than the magnetic length, the exciton becomes strongly confined to an interstitial site of the lattice, and the structure in the spectrum reflects the short-range correlations of the Wigner crystal. We derive expressions for the energies and the radiative lifetimes of the states contributing to photoluminescence, and discuss how the results of our analysis compare with experimental observations.

I. INTRODUCTION

Recent experimental studies of high-mobility GaAs/$Al_xGa_{1-x}As$ devices using itinerant-hole photoluminescence have shown this to be a sensitive probe of the two-dimensional electron systems formed in these devices.[1] Features in the photoluminescence spectra have been found to correlate well with transport measurements of the integer and fractional quantum Hall effects,[2–4] and the insulating regime observed at very low filling fraction.[5–7] These studies are particularly interesting in this low-filling-fraction regime, where transport measurements can provide only limited information on the state of the two-dimensional electron gas. In particular, they may provide information on the transition to the Wigner crystal, which is the expected ground state under these conditions. Related experimental studies of the photoluminescence of intentionally acceptor-doped samples, in which the photoexcited hole becomes bound to an acceptor, have also found structure associated with the transition to an insulating regime.[8,9] However, the signature of the insulating state in those experiments is quite different from that in the undoped devices in which the photoexcited hole is free to move (''itinerant-hole'' photoluminescence).

In the undoped samples, the transition to the insulating regime is associated with the appearance of additional lines in the photoluminescence spectrum.[5–7] It has been suggested that the appearance of these lines does indeed indicate the formation of a magnetically induced Wigner crystal.[5,6] However, the theory underlying itinerant-hole photoluminescence in the extreme quantum limit is not well understood. Existing theories of photoluminescence in the Wigner crystal regime apply only to the limit in which the photoexcited hole is far from the electron gas compared to the typical electron-electron spacing.[10,11] This condition is not appropriate for typical devices in which itinerant-hole photoluminescence is observed: while the asymmetry of these devices does cause the hole to lie some distance from the electron gas, it is believed to be close compared to the electron-electron spacing.[12]

We present a theory for itinerant-hole photoluminescence in the Wigner crystal regime that applies to systems in which the photoexcited hole is close to the two-dimensional electron gas. We study the limit of strong magnetic field in which all the electrons and the photoexcited hole are confined to states in the lowest Landau level. To represent the asymmetry of the quantum well, the hole is assumed to lie in a plane a distance d away from the two-dimensional electron gas (which is also assumed to have zero thickness). Such a model is common in theories of photoluminescence in the fractional quantum Hall regime.[13–15] In that regime, the model presents an intrinsically strongly coupled many-body problem, which, for the most part, has required numerical investigation. For very low filling fraction and for d small compared to the typical electron-electron spacing, certain simplifications arise. Under these conditions, we expect that the low-lying energy states in the presence of the photoexcited hole involve the formation of an exciton, with one electron strongly bound to the photoexcited hole. A well-defined exciton can form provided the size of the exciton, set by the magnetic length $l \equiv \sqrt{\hbar/eB}$, is small compared to the electron-electron spacing. Moreover, since the exciton is neutral, it will couple rather weakly with the other electrons. In particular, we expect that for small d, the presence of the exciton will not significantly perturb the ground-state (Wigner crystal) configuration of the other electrons, and the exciton will behave as a rather noninvasive probe of this crystalline state. This expectation is motivated by studies of the classical ground state of a system of electrons in the presence of an ionized donor impurity located a distance d from the plane of the two-dimensional electron gas.[16] It is found that for $d < 0.29a$ (where a is the lattice constant of the Wigner crystal) the system adopts a ground-state configuration in which one electron is bound to the donor impurity, while the remaining electrons lie close to the sites of a triangular lattice.

In this paper, we discuss how the photoluminescence spectrum arising from the recombination of the exciton is affected by the presence of the crystal. We study a model in which the exciton moves in the static potential set up by a triangular lattice of electrons. Our analysis neglects the dy-

namical properties of the lattice (phonon coupling). This approximation is justified, as the structure that we will find is on a energy scale large compared to the typical magnetophonon energy, $O(l^2e^2/\epsilon a^3)$: the lattice can therefore be considered to be static on the time scale necessary to define these exciton states. In the present work we neglect exchange processes between the electron in the exciton and those forming the lattice; these will be considered in a separate paper.[17]

The behavior of our model is strongly dependent on the separation d. For small d, the coupling of the exciton to the lattice is weak; in this case, additional peaks appear to higher energy of the main exciton line arising from the recombination of exciton states with momenta equal to reciprocal lattice vectors of the crystal. The spectrum effectively carries diffraction information on the crystal and is therefore sensitive to long-range crystalline order. As d increases, the coupling becomes stronger and the low-lying exciton states become increasingly confined to the interstitial regions of the crystal. These states are less sensitive to the presence of crystalline order, but depend on the short-range correlations of the electron gas. The crossover from weak to strong coupling occurs when the (dipole) potential energy of the exciton, $O(d^2e^2/\epsilon a^3)$, becomes larger than the kinetic energy cost to confine the exciton to a size of order the lattice constant $\hbar^2/Ma^2$ (where M is the effective mass of the exciton). We show that this crossover occurs when the separation between electron and hole planes d becomes larger than the magnetic length.

We compare the results of our model with experimental observations of photoluminescence in the extreme quantum limit. We find that the energy scale predicted by our model for the splitting of the exciton transition is comparable to the energy scale of the structure observed in experiment. We argue that the observed peak splitting that has been associated with the Wigner crystal regime could arise from the ground state and first excited state of a strongly confined interstitial exciton. However, other explanations for this splitting cannot be ruled out, and it may be that the structure that we predict is not seen due to a lack of population of the higher-energy exciton states. Further experimental work is required in order to identify this structure. We argue that this could best be observed in optical absorption experiments.

The paper is organized as follows. In the following section we review the theory of the free two-dimensional exciton in a strong magnetic field. We derive expressions for the binding energy and effective mass of the exciton for the case in which the electron and hole lie in planes separated by d. In Sec. III, we study the coupling of the exciton to the periodic potential set up by a Wigner crystal of other electrons. We derive exact expressions for the matrix elements of the periodic potential of the crystalline lattice within the basis of free exciton states, and study the resulting low-energy exciton states in the weak-coupling limit, $d \ll l$, using perturbation theory about the free exciton states. In Sec. IV we study the limit of strong coupling, $d \gtrsim l$. To do so, we develop an effective Hamiltonian for the motion of the exciton in a smooth external potential. We apply this to the case in which the potential is due to the presence of a Wigner crystal, and study the low-energy eigenstates close to an interstitial site of the electron crystal. For both the weak- and strong-coupling limits we derive expressions for the energies and relative radiative lifetimes of the low-lying energy states. In Sec. V we discuss how these results compare with existing photoluminescence measurements on $GaAs/Al_xGa_{1-x}As$ heterostructures in the extreme quantum limit. Finally, Sec. VI contains a summary of the main points of the paper.

II. FREE TWO-DIMENSIONAL EXCITON STATES IN STRONG MAGNETIC FIELD

The wave functions and the energy spectrum of a free two-dimensional exciton in a strong magnetic field have been discussed by Lerner and Lozovik.[18] They built on the work of Gor'kov and Dzyaloshinskii[19] who showed that, within the effective-mass approximation for the conduction and valence bands, the exciton states can be described in terms of a conserved two-dimensional momentum $\boldsymbol{P}$. In the limit of strong magnetic field, when the cyclotron energies of both the electron and hole become large compared to the typical electron-hole interaction energy, the exciton wave function is completely specified by the momentum $\boldsymbol{P}$ and the Landau level indices of the electron and hole. The lowest-energy exciton state, in which the electron and hole are both in the lowest Landau level, has the wave function[19]

$$\langle \boldsymbol{r}_e, \boldsymbol{r}_h | \boldsymbol{P} \rangle = \frac{1}{\sqrt{2\pi A}} e^{i\boldsymbol{P}\cdot(\boldsymbol{r}_e+\boldsymbol{r}_h)/2} e^{i\boldsymbol{r}_e\times\boldsymbol{r}_h\cdot\hat{\boldsymbol{z}}/2} e^{-(\boldsymbol{r}_e-\boldsymbol{r}_h-\boldsymbol{r}_P)^2/4}, \tag{1}$$

where $\boldsymbol{r}_e$ and $\boldsymbol{r}_h$ are the electron and hole positions, $\boldsymbol{r}_P \equiv \hat{\boldsymbol{z}}\times\boldsymbol{P}l^2/\hbar$, A is the area of the system, and the vector potential has been chosen in the symmetric gauge, $\boldsymbol{A}(\boldsymbol{r}) = \boldsymbol{B}\times\boldsymbol{r}/2$. We have chosen units in which $\hbar = l = 1$, and, in the following, we express energies in units of $e^2/4\pi\epsilon\epsilon_0 l$. To make the discussion more transparent, however, we reintroduce these units at appropriate points.

The energies of the free exciton states, relative to the zero-point kinetic energy of the electron and hole, are given by the expectation values of the electron-hole interaction potential.[18] We study a situation in which the electron and hole move in planes separated by a distance d, such that the interaction is

$$V_d^{eh}(\boldsymbol{r}) = -\frac{e^2}{4\pi\epsilon\epsilon_0}\frac{1}{\sqrt{|\boldsymbol{r}|^2+d^2}}. \tag{2}$$

The interaction energy of the exciton state $|\boldsymbol{P}\rangle$ is therefore

$$E_d(\boldsymbol{P}) \equiv \langle \boldsymbol{P}| V_d^{eh}(\boldsymbol{r}_e-\boldsymbol{r}_h)|\boldsymbol{P}\rangle = -\int_0^\infty e^{-u^2/2} e^{-ud} J_0(u|\boldsymbol{P}|)\,du, \tag{3}$$

where $J_0(z)$ is the ordinary Bessel function. We have not found a closed-form expression for this integral (in the limit $d=0$ it reduces to the expression derived in Ref. 18). However, in what follows, we are interested in exciton states with wave vectors on the scale of the reciprocal lattice vectors of the crystal, $O(1/a)$. At low filling fraction, this is much smaller than the scale on which the energy (3) varies, $O(1/l)$, so for our purposes it is sufficient to work with an expansion

$$E_d(\boldsymbol{P}) \simeq -B_d + \frac{\boldsymbol{P}^2}{2M_d} + O(P^4), \tag{4}$$

which may be interpreted in terms of a ''binding energy'' B_d and an ''effective mass'' M_d,

$$B_d = \sqrt{\pi/2}\, e^{d^2/2}\, \mathrm{erfc}(d/\sqrt{2}), \tag{5a}$$

$$B_d \sim \sqrt{\pi/2} - d + 1/2\sqrt{\pi/2}\, d^2 + O(d^3), \quad d \ll 1, \tag{5b}$$

$$B_d \sim 1/d + O(1/d^2), \quad d \gg 1, \tag{5c}$$

$$1/M_d = \sqrt{\pi/8}\, e^{d^2/2}(1+d^2)\, \mathrm{erfc}(d/\sqrt{2}) - d/2, \tag{6a}$$

$$1/M_d \sim \sqrt{\pi/8} - d + 3/2\sqrt{\pi/8}\, d^2 + O(d^3), \quad d \ll 1, \tag{6b}$$

$$1/M_d \sim 1/d^3 + O(1/d^4), \quad d \gg 1. \tag{6c}$$

In many ways the exciton behaves as a simple free particle with a mass M_d. In particular, the velocity of an exciton in the state $|\boldsymbol{P}\rangle$ is $\partial E_d(\boldsymbol{P})/\partial \boldsymbol{P}$.[19] In view of this, it is convenient to think of the contribution to $E_d(\boldsymbol{P})$ that is momentum dependent as the ''kinetic energy'' of the exciton, even though this energy originates from the electron-hole interaction. The exciton is overall charge neutral, but it does carry a dipole moment and therefore couples weakly to an external electrostatic potential. The dipole moment is of size $-ed\hat{\boldsymbol{z}}$ perpendicular to the plane of the interface, and of size $-e\boldsymbol{r}_P$ parallel to this plane. It is the dipole moment of the exciton that will cause it to be scattered by the Wigner crystal of other electrons. The strength of the coupling of the exciton to a Wigner crystal with a lattice constant a is determined by the competition between the typical dipole energy of the exciton, $d^2e^2/\epsilon a^3$, which is minimized at an interstitial site of the lattice, and the kinetic energy cost to confine the exciton to such a site, $1/(M_d a^2)$. Using the asymptotic expression for the mass (6c), it is found that these two energies become equal when $d/l \simeq 1/\nu^{1/10}$, where ν is the filling fraction of the two-dimensional electron gas. This condition is so weakly dependent on the filling fraction that it is accurate to say that one expects the exciton states to cross over from being weakly coupled to the lattice when $d \lesssim l$ to strongly confined to interstitial regions when $d \gtrsim l$.

In order to understand the photoluminescence spectrum it is essential to know both the energies of the exciton states and the rate at which they decay to emit radiation. Within the effective-mass approximation for the electron and hole bands, the radiative lifetime of an exciton state depends on two factors: the band-to-band dipole matrix element of the host semiconductor, and a matrix element between the electron and hole envelope functions. The first contribution is constant for all transitions. We study only the envelope term, which is sufficient to describe the relative recombination rates of different exciton states. The operator that describes electron-hole annihilation may be written in second quantized notation as

$$\hat{L} = \sqrt{\frac{2\pi l^2}{A}} \int d^2\boldsymbol{r}\ \hat{\psi}_e(\boldsymbol{r})\hat{\psi}_h(\boldsymbol{r}), \tag{7}$$

where $\hat{\psi}_h$ and $\hat{\psi}_e$ are the field annihilation operators for holes and electrons, and the wave vector of the emitted photon has been assumed to be zero. We have chosen a convenient normalization, which is arbitrary since we consider only relative radiative decay rates. We define the ''luminescence strength'' of a transition between any pair of (many-body) states $|i\rangle \to |f\rangle$ by

$$L_{if} = |\langle f|\hat{L}|i\rangle|^2. \tag{8}$$

For the free exciton states discussed above (1) the only available final state is the vacuum and we find

$$L_{\boldsymbol{P}} = |\langle \mathrm{vac}|\hat{L}|\boldsymbol{P}\rangle|^2 = \delta_{\boldsymbol{P},0}. \tag{9}$$

Thus, due to momentum conservation, only the $\boldsymbol{P}=0$ exciton state can emit a long-wavelength photon. Since the free exciton states form a complete set of states for the exciton in the lowest Landau level, the luminescence strength of a transition in which an exciton in a general state $|\psi\rangle$ decays to emit radiation may be found from Eq. (9) to be given by

$$L_\psi = |\langle \boldsymbol{P}=0|\psi\rangle|^2. \tag{10}$$

We will use this expression in the following discussions, which concern the exciton states in an external potential, for which the momentum is not a good quantum number.

The relative intensity of a photoluminescence transition is proportional to its luminescence strength multiplied by the probability for a photoexcited hole to be in the initial state of the transition. These probabilities are difficult to quantify: the lifetime of the photoexcited hole can be shorter than its equilibration time, so one may observe recombination from high-energy nonequilibrium states. Although we are primarily interested in photoluminescence, we note here that the luminescence strength L_{if} also characterizes the strength of the transition, as observed in optical absorption, from the state $|f\rangle$ to the state $|i\rangle$. In this case, uncertainties related to nonequilibrium populations do not arise as the lifetime of the hole does not limit the time available for the initial states to equilibrate.

III. EXCITON STATES IN THE PRESENCE OF A WIGNER CRYSTAL: WEAK COUPLING LIMIT

We will now study the scattering of the exciton states (1) by the electrons forming the Wigner crystal. As described in the Introduction, we neglect the exchange interaction between the electron in the exciton and those forming the crystal. We anticipate that this is a good approximation at low filling fraction, when the fraction of basis states excluded from the exciton wave function by the Pauli exclusion principle is small; this is confirmed by numerical studies in which the effects of exchange are included.[17] The model we discuss also correctly represents a situation in which the spin or subband index of the electron in the exciton is different from the corresponding index of the electrons in the two-dimensional electron gas.

Neglecting exchange interactions, the exciton is scattered only by the charge density

$$\rho(\boldsymbol{r}) = \sum_\alpha \left(\frac{-e}{2\pi l^2} e^{-(\boldsymbol{r}-\boldsymbol{R}_\alpha)^2/2l^2} + \overline{\rho} \right) \delta(z), \tag{11}$$

which represents a system of electrons in lowest Landau level orbitals centered at the sites $\{\boldsymbol{R}_\alpha\}$ of a triangular lattice with a lattice constant a [the filling fraction is therefore $\nu=(4\pi/\sqrt{3})l^2/a^2$]. The magnitude of $\overline{\rho}$ is chosen to provide a uniform neutralizing positive background. The resulting external potential energies of the electron and hole are

$$V^e(\boldsymbol{r}_e)+V^h(\boldsymbol{r}_h)=\int \rho(\boldsymbol{r})\frac{e}{4\pi\epsilon\epsilon_0}\left[\frac{-1}{|\boldsymbol{r}_e-\boldsymbol{r}|}+\frac{1}{\sqrt{(\boldsymbol{r}_h-\boldsymbol{r})^2+d^2}}\right]d^2\boldsymbol{r}. \tag{12}$$

The motion of the exciton in this potential is fully described by the matrix elements of all interactions within the basis of free exciton states (1):

$$H_{\boldsymbol{P}',\boldsymbol{P}}\equiv\langle\boldsymbol{P}'|V^{eh}(\boldsymbol{r}_e-\boldsymbol{r}_h)+V^e(\boldsymbol{r}_e)+V^h(\boldsymbol{r}_h)|\boldsymbol{P}\rangle. \tag{13}$$

The first term is diagonal in this basis and gives rise to the free exciton energy (3), (4). The last two terms describe the scattering. Due to the discrete translational symmetry of the crystal, the only matrix elements that are nonzero are those between states that differ by a reciprocal lattice vector of the crystal, $\{\boldsymbol{K}\}$. Since we are ultimately interested in states that can emit radiation, and which therefore from Eq. (10) must contain some component of the zero-momentum free exciton state, we need only study the basis states $|\boldsymbol{K}\rangle$ in which the wave vector of the exciton is a reciprocal lattice vector. After a lengthy but straightforward process we find

$$H_{\boldsymbol{K}',\boldsymbol{K}}=\delta_{\boldsymbol{K}',\boldsymbol{K}}E_d(\boldsymbol{K})+(1-\delta_{\boldsymbol{K}',\boldsymbol{K}})\nu\frac{e^{-3(\boldsymbol{K}-\boldsymbol{K}')^2/4}}{|\boldsymbol{K}-\boldsymbol{K}'|}\times[e^{i\boldsymbol{K}\times\boldsymbol{K}'\cdot\hat{\boldsymbol{z}}/2}-e^{-|\boldsymbol{K}-\boldsymbol{K}'|d}e^{-i\boldsymbol{K}\times\boldsymbol{K}'\cdot\hat{\boldsymbol{z}}/2}]. \tag{14}$$

While the above expressions for the matrix elements are exact and therefore fully describe the behavior of our model, in their present form they are not useful for analytic purposes. To obtain some insight into the properties of this system we therefore introduce some approximations. In what follows, we develop a perturbation theory in d/l, based on the free exciton states, which is valid in the limit of low filling fraction.

Consider the case $d=0$ in which the hole moves in the same plane as the electrons. In this limit, the zero-momentum exciton state is not scattered by the potential, and is therefore an exact energy eigenstate

$$H_{\boldsymbol{K}',\boldsymbol{K}=0}=E_{d=0}(0)\,\delta_{\boldsymbol{K}',0}. \tag{15}$$

All other energy eigenstates must be orthogonal to this state, so from Eq. (10) they must have zero luminescence strength. There is therefore only a single line in the photoluminescence spectrum. Moreover, this line appears at the same energy as the exciton line in the absence of the two-dimensional electron gas. This recovers a restricted version of the general result that when electrons and holes are confined to the lowest Landau level and move in the same plane, photoluminescence contains no spectroscopic information on the state of the two-dimensional electron gas.[20,13]

Although the free exciton state with zero momentum is an exact energy eigenstate of the system at $d=0$, other free exciton states are not. There do remain nonzero off-diagonal matrix elements coupling these states. However, in the limit of small filling fraction, this coupling may be neglected for the lowest-energy states: states with small momenta $|\boldsymbol{K}|\sim 1/a$, are only coupled by off-diagonal terms of order $(l/a)^3$, whereas the energy spacing between these states is of order $\hbar^2/(M_{d=0}a^2)\sim(l/a)^2$. We choose to work in an expansion in small filling fraction, and neglect terms of order $(l/a)^3$ and higher. Within this approximation, the free exciton states are eigenstates of the Hamiltonian at $d=0$.

We now study the deviations from $d=0$ within a perturbation expansion in the separation d. Expanding the Hamiltonian in this parameter, we find

$$H_{\boldsymbol{K}',\boldsymbol{K}}\simeq\delta_{\boldsymbol{K}',\boldsymbol{K}}\left[-B_d+\frac{\boldsymbol{K}^2}{2}\left(\sqrt{\frac{\pi}{8}}-d+\frac{3}{2}\sqrt{\frac{\pi}{8}}d^2\right)\right]+(1-\delta_{\boldsymbol{K}',\boldsymbol{K}})\nu d+O(1/a^3)+O(d^3/a^2). \tag{16}$$

To lowest order in d all states are coupled by a matrix element $(1-\delta_{\boldsymbol{K}',\boldsymbol{K}})\nu d$. This leads to a mixing of the radiative state, $|\boldsymbol{K}=0\rangle$, with all other exciton states of reciprocal lattice vector; if these states become populated on photoexcitation, each will contribute an additional line to the photoluminescence spectrum. The strongest new line arises from the six lowest-lying energy states, at $|\boldsymbol{K}|=4\pi/(\sqrt{3}a)$. Within degenerate perturbation theory, these states are split into a fivefold degenerate level shifted in energy by $-\nu d$, and a single state shifted by $+5\nu d$. We now apply first-order perturbation theory to calculate the mixing of $|\boldsymbol{K}=0\rangle$ into these states. This amplitude is zero for the five degenerate states, and equal to $\sqrt{6}\nu d$ for the single state split off from these five. The luminescence spectrum therefore consists of a strong spectral line from what was the free zero-momentum exciton state, at an energy (relative to the band-gap plus zero point kinetic energy) and with a luminescence strength

$$E_0=-B_d, \tag{17}$$

$$L_0=1-O(d^2), \tag{18}$$

and an additional line arising from the exciton states with the smallest nonzero reciprocal lattice vector, with an energy and luminescence strength

$$E_1=-B_d+\frac{(4\pi/\sqrt{3}a)^2}{2}\left[\sqrt{\frac{\pi}{8}}-d+\frac{3}{2}\sqrt{\frac{\pi}{8}}d^2\right]+5\nu d, \tag{19}$$

$$L_1=\left|\frac{\sqrt{6}\nu d}{(1/2)\sqrt{\pi/8}(4\pi/\sqrt{3}a)^2}\right|^2=\frac{36}{\pi^3}d^2. \tag{20}$$

The next line to higher energy arises from the six states at $|\boldsymbol{K}|=4\pi/a$, and has a luminescence strength that is smaller than this by a factor of $[(4\pi/\sqrt{3})/(4\pi)]^4=1/9$.

Thus, for small d the presence of the crystal causes peaks to appear in the photoluminescence spectrum to higher energy than the main exciton line. The energy spacing between these peaks is primarily determined by the effective mass of the exciton and the reciprocal lattice vectors of the Wigner

crystal, with a small correction due to the coupling to the lattice potential. The luminescence strengths of these higher-energy exciton states are small compared to the lowest-energy exciton transition, and will therefore give rise to much weaker spectral features. The luminescence strengths of these features grow as d increases. However, at the same time, the exciton becomes more strongly coupled to the lattice potential, so the energies of these states deviate from the energies of the reciprocal lattice vector excitons. Once the coupling is larger than the spacing between the free exciton states, $\nu d \gtrsim 1/M_d a^2$, the corrections to these energies become large, and the above perturbation theory breaks down. This condition is equivalent to the strong-coupling condition, $d \gtrsim l$, that we derived earlier. In the strong-coupling limit, a quite different approach is required to describe the low-energy exciton states.

IV. EXCITON STATES IN THE STRONG-COUPLING LIMIT: THE INTERSTITIAL EXCITON

We now turn to the limit of strong exciton-lattice coupling, in which the kinetic energy of the exciton is small compared to its dipole potential energy in the lattice. If we were to neglect the kinetic energy completely, then the ground state of the system would be the same as that of the classical ground state of electrons in the presence of an ionized donor impurity:[16] the exciton, with its weak dipole moment, would position itself at an interstitial site of the lattice, and, for $d<0.29a$, the remaining electrons would form an essentially undeformed Wigner crystal. The exciton does, however, have some residual dynamics. In this section we develop an effective Hamiltonian that describes the motion of the center of the exciton in a smooth external potential. We then apply this to the potential close to an interstitial site of the Wigner crystal (12), and study the energies and luminescence strengths of the low-energy states. Those who do not care to follow the derivation of the effective theory may proceed to Sec. IV B without loss of continuity.

A. Effective Hamiltonian

In order to derive an effective Hamiltonian for the motion of an exciton in a smooth external potential, we extend the approach of Gor'kov and Dzyaloshinskii[19] to include the potential energies $V^e(\boldsymbol{r}_e)$ and $V^h(\boldsymbol{r}_h)$ felt by the electron and hole. We start from the full Hamiltonian for the interacting electron-hole pair in a uniform magnetic field:

$$H=\frac{(\boldsymbol{p}_e+e\boldsymbol{A}_e)^2}{2m_e}+\frac{(\boldsymbol{p}_h-e\boldsymbol{A}_h)^2}{2m_h}+V^{eh}(\boldsymbol{r}_e-\boldsymbol{r}_h)+V^e(\boldsymbol{r}_e)+V^h(\boldsymbol{r}_h), \tag{21}$$

and work in the symmetric gauge. We transform to a new set of coordinates, the first of which is the momentum defined in Ref. 19; the remaining coordinates are the position of the center of mass and a momentum and position describing the internal motion

$$\boldsymbol{P}\equiv\boldsymbol{p}_e+\boldsymbol{p}_h-\frac{e}{2}\boldsymbol{B}\times(\boldsymbol{r}_e-\boldsymbol{r}_h), \tag{22}$$

$$\boldsymbol{R}_c\equiv\frac{m_e\boldsymbol{r}_e+m_h\boldsymbol{r}_h}{m_e+m_h}, \tag{23}$$

$$\boldsymbol{p}\equiv\frac{m_h\boldsymbol{p}_e-m_e\boldsymbol{p}_h}{m_e+m_h}+\frac{e}{2}\boldsymbol{B}\times\left(\frac{m_e\boldsymbol{r}_e+m_h\boldsymbol{r}_h}{m_e+m_h}\right), \tag{24}$$

$$\boldsymbol{r}\equiv\boldsymbol{r}_e-\boldsymbol{r}_h. \tag{25}$$

The center of mass and the internal coordinates behave as independent, canonically conjugate pairs. In terms of these new coordinates, the Hamiltonian is

$$H=\frac{(\boldsymbol{P}+e\boldsymbol{B}\times\boldsymbol{r})^2}{2M}+\frac{(\boldsymbol{p}+\gamma e/2\boldsymbol{B}\times\boldsymbol{r})^2}{2\mu}+V^{eh}(\boldsymbol{r})+V^e(\boldsymbol{R}_c+\eta_h\boldsymbol{r})+V^h(\boldsymbol{R}_c-\eta_e\boldsymbol{r}), \tag{26}$$

where $M\equiv m_e+m_h$, $\mu\equiv m_e m_h/(m_e+m_h)$, $\gamma\equiv(m_h-m_e)/(m_e+m_h)$, and $\eta_\alpha\equiv m_\alpha/(m_e+m_h)$.

First consider the free exciton, $V^e=V^h=0$. The Hamiltonian is independent of $\boldsymbol{R}_c$, so the momentum $\boldsymbol{P}$ is conserved and can be replaced by its eigenvalue to leave a Hamiltonian for the internal coordinates, $\boldsymbol{r}$ and $\boldsymbol{p}$. If the interaction potential $V^{eh}(\boldsymbol{r})$ is neglected in relation to the kinetic energy, the internal motion consists of Landau level orbits centered on $\boldsymbol{r}_P\equiv\hat{\boldsymbol{z}}\times\boldsymbol{P}l^2/\hbar$. This is the neglect of Landau level coupling, and is the simplification used in Refs. 18,19; in the lowest Landau level, it leads to the states that we have discussed above (1).

We now introduce the potentials, V^e and V^h, and follow a similar approximation. The momentum is no longer conserved. However, provided the potential is sufficiently weak, the momentum only changes slowly compared to the rapid cyclotron motion. The motion can then be treated adiabatically, with the fast internal motion fixed in the lowest Landau level and adjusting to follow the slowly changing momentum. This procedure is analogous to the Born-Oppenheimer approximation in the theory of molecular dynamics,[21] where the fast electronic degrees of freedom are eliminated to provide an effective theory for the slow atomic coordinates. In the same way, we obtain an *effective Hamiltonian* for the operators $\boldsymbol{P}$ and $\boldsymbol{R}_c$. Before we write down this Hamiltonian, we make one last change of variables. It is convenient to work is terms of the coordinate $\boldsymbol{R}\equiv(\boldsymbol{r}_e+\boldsymbol{r}_h)/2$ rather than the center of mass. This avoids any irrelevant mass dependence in our analysis (''irrelevant'' since both electron and hole are restricted to the lowest Landau level). This transformation does not affect the commutation relation of the position and momentum coordinates. The effective Hamiltonian in terms of these coordinates is found to be

$$H=\hbar\omega_\mu/2+E_d(\boldsymbol{P})+\int\{V^e[\boldsymbol{R}+(\boldsymbol{r}'+\boldsymbol{r}_P)/2]+V^h[\boldsymbol{R}-(\boldsymbol{r}'+\boldsymbol{r}_P)/2]\}\frac{e^{-\boldsymbol{r}'^2/2l^2}}{2\pi l^2}d^2\boldsymbol{r}', \tag{27}$$

where $\hbar\omega_\mu/2=\hbar eB/2\mu$ is the zero-point kinetic energy of the electron and hole, and $E_d(\boldsymbol{P})$ is the dispersion relation (3) arising from their mutual attraction.

To convert this expression into a more convenient form, we make use of the simplifications available in the problem. As we are interested in small momenta, $O(1/a) \ll 1/l$, we expand the dispersion relation to quadratic order. Also, provided the exciton is not very close to a lattice site, the potentials V^e and V^h are smooth on the length scale of the magnetic length, and it is a good approximation to expand these terms in the integral of Eq. (27) to first order in $(\boldsymbol{r}' + \boldsymbol{r}_P)$. We find

$$H \simeq \hbar\omega_\mu/2 - B_d + \frac{\boldsymbol{P}^2}{2M_d} + V^e(\boldsymbol{R}) + V^h(\boldsymbol{R}) - \frac{1}{2}\boldsymbol{P}\cdot[\nabla V^e(\boldsymbol{R}) - \nabla V^h(\boldsymbol{R})]\times\hat{\boldsymbol{z}}. \quad (28)$$

This is a general expression for the motion of the exciton in smooth external potentials, and in a strong magnetic field. An expression for the mass M_d was calculated earlier (6). Since the position and momentum operators are canonically conjugate coordinates, the energy eigenstates of the exciton follow from the solutions of the Schrödinger equation with the Hamiltonian (28); it is to be understood that the final term of this expression is symmetrized in the position and momentum operators, such that the Hamiltonian is Hermitian. Two approximations were used to derive this expression. Firstly, the rapid cyclotron motion was assumed to adiabatically follow the changing momentum; this is valid provided the spacing between the energy levels arising from the center-of-mass motion is small compared to the cyclotron energy $\hbar\omega_\mu$ (no Landau level coupling). Secondly, we expanded the external potential to first order in $(\boldsymbol{r}' + \boldsymbol{r}_P)$, neglecting terms of order $l^2\nabla^2 V$.

B. Motion in the Wigner crystal: harmonic approximation

We now use the formalism developed in the previous subsection to study the motion of the exciton in a Wigner crystal. We therefore introduce the electrostatic interactions (12) as the external potentials in the effective Hamiltonian (28). This expression for the effective Hamiltonian requires the external potential to vary slowly on the scale of the exciton size l. We therefore expect this formalism to provide an appropriate description of the low-energy states when $d \gtrsim l$, in which the exciton remains far from the interstitial sites. However, we still assume that $d/a \ll 1$, and expand the effective Hamiltonian in this parameter to find

$$H = \frac{\boldsymbol{P}^2}{2M_d} + \frac{e^2}{8\pi\epsilon\epsilon_0}\sum_\alpha \frac{d^2}{|\boldsymbol{R}-\boldsymbol{R}_\alpha|^3} + \frac{e^2}{4\pi\epsilon\epsilon_0}\frac{l^2\boldsymbol{P}}{\hbar}\cdot\sum_\alpha \hat{\boldsymbol{z}} \times \frac{\boldsymbol{R}-\boldsymbol{R}_\alpha}{|\boldsymbol{R}-\boldsymbol{R}_\alpha|^3}, \quad (29)$$

where the sums run over the triangular lattice sites $\{\boldsymbol{R}_\alpha\}$, and the final term should be interpreted as the symmetrized product of the momentum-dependent and position-dependent factors. We have dropped the constant binding energy $-B_d$ and the zero-point kinetic energy $\hbar\omega_\mu/2$ and have also neglected a constant energy $-\nu d$, which arises from the dipole moment of the exciton in the electric field of the neutralizing positive background. Each of the terms appearing in this effective Hamiltonian has a simple intuitive interpretation. The first two terms simply represent the kinetic energy of the exciton and the dipole energy for the exciton to be centered at the position $\boldsymbol{R}$. The final term is less familiar, but also arises from a dipole energy: in this case, due to the in-plane dipole moment of the exciton, which is of a size $-e\boldsymbol{r}_P$ for an exciton with momentum $\boldsymbol{P}$. The resulting contribution to the Hamiltonian resembles the first-order coupling of a charged particle to a vector potential.

We have argued that for large d/l the exciton is confined at the interstitial sites of the triangular lattice. We now study the low-lying states of this effective Hamiltonian within the simplest approximation, in which the potential is expanded to harmonic order about an interstitial site. Keeping only the contributions to the confinement potential arising from the nearest three lattice electrons, we find

$$H = \frac{\boldsymbol{P}^2}{2M_d} + \frac{1}{2}\frac{243\sqrt{3}}{4}\frac{d^2}{a^5}\boldsymbol{R}^2 - \frac{9\sqrt{3}}{2}\frac{1}{a^3}(\boldsymbol{R}\times\boldsymbol{P})\cdot\hat{\boldsymbol{z}}, \quad (30)$$

plus a constant energy of $11.57d^2/a^3$ due to the dipole energy at the interstitial site of the triangular lattice.[16] We have again set $e^2/4\pi\epsilon\epsilon_0 l = \hbar = l = 1$. Due to the rotational invariance of this Hamiltonian, its eigenstates may be classified by their angular momenta, $\boldsymbol{R}\times\boldsymbol{P}\cdot\hat{\boldsymbol{z}}$. We restrict attention to states with *zero* angular momentum, since only these states can emit long-wavelength radiation. The zero angular momentum states of the two-dimensional harmonic oscillator have the wave functions and energies

$$\psi_n^{\rm sho}(\boldsymbol{R}) = \frac{1}{\sqrt{\pi R_0^2}} e^{-\boldsymbol{R}^2/2R_0^2}\mathscr{L}_n(\boldsymbol{R}^2/R_0^2), \quad (31)$$

$$E_n = \hbar\Omega(2n+1), \quad (32)$$

where

$$\hbar\Omega = \sqrt{\frac{243\sqrt{3}}{4}\frac{d^2}{a^5}\frac{e^2}{4\pi\epsilon\epsilon_0}\frac{\hbar^2}{M_d}} \quad (33a)$$

$$\overset{d\gg l}{\to} \sqrt{\frac{243\sqrt{3}}{4}\frac{l^6}{a^5 d}\frac{e^2}{4\pi\epsilon\epsilon_0 l}}, \quad (33b)$$

$$R_0 = \left(\frac{4}{243\sqrt{3}}\frac{4\pi\epsilon\epsilon_0}{e^2}\frac{\hbar^2 a^5}{d^2 M_d}\right)^{1/4} \quad (34a)$$

$$\overset{d\gg l}{\to} \left(\frac{4}{243\sqrt{3}}\frac{a^5}{d^5}\right)^{1/4} l, \quad (34b)$$

and $\mathscr{L}_n(z)$ are the Laguerre polynomials. The expressions for $d \gg l$ follow from the asymptotic expansion of the effective mass (6c). Note that the two-dimensional harmonic oscillator states with zero angular momentum are spaced in energy by *twice* the oscillator quantum, $\hbar\Omega$.

Radiative recombination occurs from each of these states. For consistency with the definition of the luminescence strength in the weak-coupling limit, we construct states consisting of a superposition of harmonic oscillator states, placed at each interstitial site of a crystal with an area A. The only such linear combination with nonzero luminescence

strength is the state in which all states are combined with the same amplitude and phase. From Eq. (9) we find that the luminescence strength of such a state constructed from $\psi_n^{\rm sho}$ is

$$L_n \equiv \frac{4}{\sqrt{3}a^2}\left|\int d^2\boldsymbol{R}\,\psi_n^{\rm sho}(\boldsymbol{R})\right|^2 = \frac{16\pi R_0^2}{\sqrt{3}a^2}. \tag{35}$$

Note that this is independent of the index n, so the ground state and all excited states have the same luminescence strengths.[22] Such behavior contrasts with the weak-coupling limit for which the lowest-energy exciton state is much more strongly coupled to radiation than the higher-energy states. As the spacing d is increased from zero, so that the system evolves from the weak-coupling to the strong-coupling limit, the higher-energy exciton transitions grow in luminescence strength from zero (optically inactive) to eventually attain the same strength as the lowest-energy exciton state.

In summary, within the harmonic approximation for the potential at an interstitial site of the lattice, the exciton contribution to the photoluminescence spectrum consists of a series of lines that are uniformly spaced and have relative intensities given by the relative populations of the states. This approximation is valid provided the typical size of the exciton state is small compared to the lattice constant. From Eq. (34a), one finds that this depends only very weakly on the filling fraction, and occurs when $d \gtrsim l$. This is the same as the strong-coupling condition under which the perturbation expansion of the previous section failed.

V. EXPERIMENTAL COMPARISONS

Several groups have reported studies of the itinerant-hole photoluminescence spectra of the two-dimensional electron systems formed in high-mobility GaAs/Al_xGa_{1-x}As devices.[1,2,5–7] Measurements on single heterojunctions and single quantum wells show qualitatively the same behavior in photoluminescence: in the fractional quantum Hall regime two spectral lines are observed; as the filling fraction is reduced below about $\nu = 1/6$, the higher-energy peak is found to split to form a doublet. The filling fraction below which this doublet structure appears correlates well with the filling fraction at which transport measurements show a transition to an insulating state.[5] This has motivated claims that the doublet is associated with the formation of a magnetically induced Wigner crystal.[5,6] However, the origin of this doublet is still not well understood. In this section, we discuss whether this doublet can be accounted for in terms of the structure that we predict. We will compare the energy splitting of the doublet with the energy difference between the two lowest-lying exciton states of our model that are optically active.

For numerical comparisons of our theory with experiment, we will focus on the results of Goldys *et al.* presented in Ref. 5. This paper reports studies of the photoluminescence spectrum of a high-mobility GaAs/Al_xGa_{1-x}As heterostructure with density $3.2\times10^{10}\,\mathrm{cm}^{-2}$. Specifically we will concentrate on a field of $B = 12.5$ T, which is appropriate for Fig. 4(a) of Ref. 5 for which the doublet structure, with a splitting of 0.5 meV, is well developed. Under these conditions, the filling fraction is $\nu = 0.1$, corresponding to a magnetic length of $l = 72$ Å and a lattice constant of $a = 600$ Å for the triangular Wigner crystal.

The most uncertain parameter that enters our model is the spacing d. This distance is expected to depend on the details of the band bending in the vicinity of the interface, and may even vary with magnetic field.[12] We will assume values of $d = 50$ and 100 Å as small and large estimates for this quantity. These are consistent with recent numerical studies of the binding of an exciton to the interface of a single heterojunction.[12] Note that these values are also consistent with the condition $d < 0.29a$, which is required by our theory in order that the exciton does not strongly deform the crystal.

The smaller of these two separations, $d = 50$ Å, is less than the magnetic length, so we expect our weak-coupling formulas to be appropriate. In the weak-coupling limit, the difference in energy between the main exciton line and the lowest-energy optically active state is primarily determined by the free exciton energy at a wave vector equal to the smallest nonzero reciprocal lattice vector $|\boldsymbol{K}| = 4\pi/(\sqrt{3}a)$. Taking the dielectric constant of GaAs to be $\epsilon = 12.53$, we find from Eq. (3) that the energy spacing is 1.3 meV.

The larger spacing, $d = 100$ Å, satisfies $d > l$, and we should therefore use our strong-coupling formulas, which state that the energy spacing between adjacent optically active exciton states is $2\hbar\Omega$ where $\hbar\Omega$ is given by Eq. (33a). Using the appropriate value for the effective mass of the exciton (6a), this evaluates to an energy spacing of 0.7 meV at $d = 100$ Å. Under these conditions, the characteristic size of the ground-state wave function is $R_0 = 150$ Å, which is smaller than, but comparable to, the distance to the closest saddle point of the crystal potential, $a/\sqrt{12} = 170$ Å. This indicates that, under these conditions, the harmonic approximation is at the limit of its validity.

An exact calculation of the energy spacings predicted by our model for these two cases is likely to lead to energies that are slightly smaller than the above estimates:[17] deviations from the weak-coupling limit will tend to push the energy spacing towards the (smaller) energy spacing one would obtain by using the strong-coupling theory, away from the strong-coupling limit, the use of the harmonic approximation will overestimate the strength of the confinement potential and hence also overestimate the energy spacing. We also expect a small reduction of both energies due to the finite thicknesses of the electron and hole subband wave functions in a real device. Even allowing for these reductions, the resulting energy spacings remain larger than the expected zone-boundary magnetophonon energy, 0.2 meV.[23] Our neglect of the dynamical properties of the Wigner crystal is therefore a consistent assumption.

The energy separation between the lowest two optically active states in our model is on the same scale as the observed energy splitting of the doublet, 0.5 meV.[5] It is therefore possible that this structure is due to the splitting of the exciton peak as described by our model. Note that, if it is indeed correct to attribute this structure to the predictions of our model, one must assume that the experimental system is in the strong-coupling limit, in which the exciton is strongly confined to an interstitial site. There are two important reasons for this, in addition to the fact that the experimental energy spacing (0.5 meV) compares more favorably with a spacing of $d = 100$ Å than with smaller values. Firstly, it is

observed that the intensities and the radiative lifetimes of the two peaks forming the doublet are similar.[5] The luminescence strengths of the two transitions must therefore be comparable. In the weak-coupling limit, all higher-energy exciton states have a much smaller luminescence strength than the lowest-energy exciton line; one must go to the strong-coupling limit before these become equal [see Eq. (35) and the following discussion]. Secondly, the observed energies of the two transitions forming the doublet are independent of the temperature of the substrate over the range $0.1-3$ K.[5] Since the Wigner crystal is expected to melt at a temperature of 0.3 K at these densities,[24] one cannot attribute this structure to a nearly free exciton state: the energy of this state would be sensitive to the changing long-range correlations at the melting transition. Instead, it is appropriate to attribute this structure to a strongly confined interstitial exciton, which is only sensitive to the short-range correlations of the electron gas.

Although it is possible that the doublet structure is due to the splitting of the exciton transition, as described by our model, there are certain problems with this interpretation. Firstly, it is not clear that one should even observe any measurable signal from the higher-energy exciton states: these states lie more than 0.5 meV above the lowest-energy exciton transition, so, if the exciton were to be in thermal equilibrium, the expected populations of these states would be vanishingly small. Secondly, assuming that there is some non-equilibrium population of the higher-energy exciton states, one would perhaps expect to see more than one additional line (recall that for a strongly confined interstitial exciton all excited states have the same luminescence strength). Furthermore, this is not the only possible explanation for this structure. For instance, the recombination from a negatively charged exciton, which has been resolved in low-density GaAs wells at weak field[25,26] and in CdTe quantum wells in strong magnetic field,[27] also may be expected to appear in these systems, and has not as yet been identified. We are therefore cautious to ascribe the observed doublet to the line splitting predicted by our model, which may not appear in these experiments due to a lack of population of the higher-energy exciton states. Further experiments are required in order to identify this structure. In particular, a study of the optical absorption spectrum would be very valuable in this respect. The exciton transitions that we have discussed will present much stronger spectral features in this experiment than in photoluminescence since the population factors, which reduce the observed intensities of high-energy exciton states in photoluminescence, are removed.

VI. SUMMARY

We have presented a model of photoluminescence in the presence of a magnetically induced Wigner crystal, for systems in which the photoexcited hole lies close to the interface. This model predicts a splitting of the exciton transition due to the scattering of the exciton by the electrons forming the Wigner crystal. We studied the behavior of our model as a function of the separation d. For $d<l$, the exciton is spread over many unit cells of the crystal. Additional peaks with small luminescence strength can appear in the photoluminescence spectrum, arising from higher-energy states in which the exciton has a wave vector equal to a reciprocal lattice vector of the crystal. The spectrum carries diffraction information and is therefore sensitive to long-range crystalline order. For $d>l$, the exciton becomes strongly confined to an interstitial region. We studied this limit by developing an effective theory for the center-of-mass motion of the exciton. Within the harmonic approximation for the potential at an interstitial site, the spectrum consists of an equally spaced set of peaks with relative intensities determined by the relative populations of the various states.

We discussed the photoluminescence experiments of Ref. 5 and showed that the energy splitting of the doublet, which has been associated with the presence of a Wigner crystal, is comparable to the energy splitting we expect from our model for $d\simeq 100$ Å. We argued that this could be due to the recombination from a interstitial exciton. However, other explanations are also possible, and further experimental investigation is required to identify the structure our model predicts. In particular, this could best be observed in optical absorption spectroscopy.

ACKNOWLEDGMENTS

While preparing this work for publication, I learned of work by J. R. Chapman, N. F. Johnson, and V. N. Nicopoulos, proposing an interpretation of the photoluminescence experiments that is somewhat different from that presented here.[28] I am grateful to R.G. Clark, A. Turberfield, and D. Heiman for stimulating my interest in this problem and for helpful subsequent discussions. I would also like to thank Dr. John Chalker for much help with the development and presentation of this work, and Professor Bertrand Halperin and the Physics Department at Harvard University for their hospitality and for many useful discussions on this and related topics. This work was supported in part by DENI and in part by the NATO Science Fellowship Programme.

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