Theory of Photoluminescence from the Wigner Crystal in a Strong Magnetic Field

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We develop a theory of photoluminescence from the magnetically induced two-dimensional Wigner crystal. It is found that the photoluminescence spectrum is a weighted measure of the single particle density of states of the electron system, which for an undisturbed electron lattice has the intricate structure of the Hofstadter butterfly. It is shown that the interaction of a localized hole with the electron lattice wipes out this structure, but that an itinerant hole can, in principle, detect it. Experimental implications are discussed.

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A subject of long-standing interest in condensed matter physics has been the search to observe crystalline electronic order in systems that are highly quantum mechanical. The possibility that electrons may form crystals at zero temperature which melt due to zero-point fluctuations as the density of electrons is increased was first pointed out by Wigner [1], and the system has since been known commonly as the Wigner crystal (WC). The two-dimensional electron gas (2DEG) has proven to be an excellent candidate for observing the formation of a WC, largely because such high mobility samples are available that the electrons are not necessarily dominated by disorder effects at the low densities necessary to obtain crystalline order. The possibility of forming a WC is further enhanced by application of a strong perpendicular magnetic field, since this quenches the kinetic energy that tends to melt the crystal at large electron density. Recent studies of high mobility heterojunctions in strong magnetic fields have uncovered a number of intriguing properties that in some ways are consistent with the presence of some crystalline order at the lowest available temperatures. These include rf data [2,3], transport experiments [4], cyclotron resonance [5], and photoluminescence (PL) experiments [6,7]. It is the last of these that we will discuss in this Letter.

Photoluminescence experiments on these systems have been performed in two ways. One set of experiments [6] uses a low density of Be dopants that are purposely grown into the sample approximately 250 Å away from the 2DEG. A pulse of light excites a core electron out of a Be^- acceptor, and the photoluminescence spectrum from recombination of electrons in the 2DEG with the remaining core hole is observed. More recent experiments [7] have also investigated recombination of electrons with itinerant holes in the host crystal (GaAs) valence band. Both experiments show intriguing and complicated results; among them is the observation of a pair of photoluminescence lines that appear at magnetic fields for which transport anomalies recently associated with the WC are found. At the lowest temperatures, the lower of the two lines has most of the oscillator strength; as the temperature is raised, the oscillator strength transfers to the higher of these lines, until the lower line cannot be distinguished from the background. While it is tempting to associate the lower line with a crystal phase, and the upper with a melted phase, the precise interpretation of the data is hampered by a lack of theoretical understanding of what the PL spectrum should look like when the ground state of the 2DEG really is a WC.

To address this question, we have computed the PL spectrum for the Wigner crystal using a time-dependent Hartree-Fock approximation (TDHFA) [8]. A few examples of PL spectrum obtained in our work are shown in Figs. 1 and 2. Figure 1(a) illustrates the PL for a localized hole, where we ignore the interaction of the hole with the lattice, and the filling fraction of electrons is taken to be $\nu = \frac{2}{7}$ [9]. The structure of the PL is essentially a double peak, whose origin may be understood as follows. The PL, as will be shown below, is essentially a weighted measure of the single particle density of states. Within mean-field theory (i.e., Hartree-Fock) this is determined by the energy spectrum of a single electron in a magnetic field, moving in the average potential of all the other electrons. Thus, the single particle density of states is that of an electron in a periodic potential. This spectrum has an intricate nature [10]: for rational filling fractions $\nu = p/q$ there are q subbands, and in Hartree-Fock theory p of these are filled. We therefore expect that for any filling fraction p/q, one should expect to see p lines in the PL for the ideal case of a perfect electron lattice. *An observation of this behavior in photoluminescence experiments would yield direct confirmation of the presence of a WC in the system.*

Unfortunately, the multiple peak structure is sensitive to perturbations from external potentials. For the case of an unscreened, localized core hole, the primary perturbation is from the electron-hole interaction itself; the PL spectrum when one turns on this interaction is illustrated in Fig. 1(b). Here there is a single luminescence peak, which is shifted down in energy from what was seen in

Fig. 1(a). The structure is best interpreted in terms of the density of states. This is illustrated in Fig. 1(c), for a periodic electron system with twelve electrons and one hole per unit cell. We see that the two filled bands break up into three peaks, and by detailed examination of the single particle (mean-field) states [11], we find that each peak corresponds to a set of electron states that are successively further from the hole for increasing energy. The lowest energy electron state overwhelmingly dominates the PL spectrum, because the overlap of its wave function with that of the hole is nearly 2 orders of magnitude larger than those of the next closest electrons. In essence, the PL spectrum is dominated by a single final state of the ion–electron-gas system.

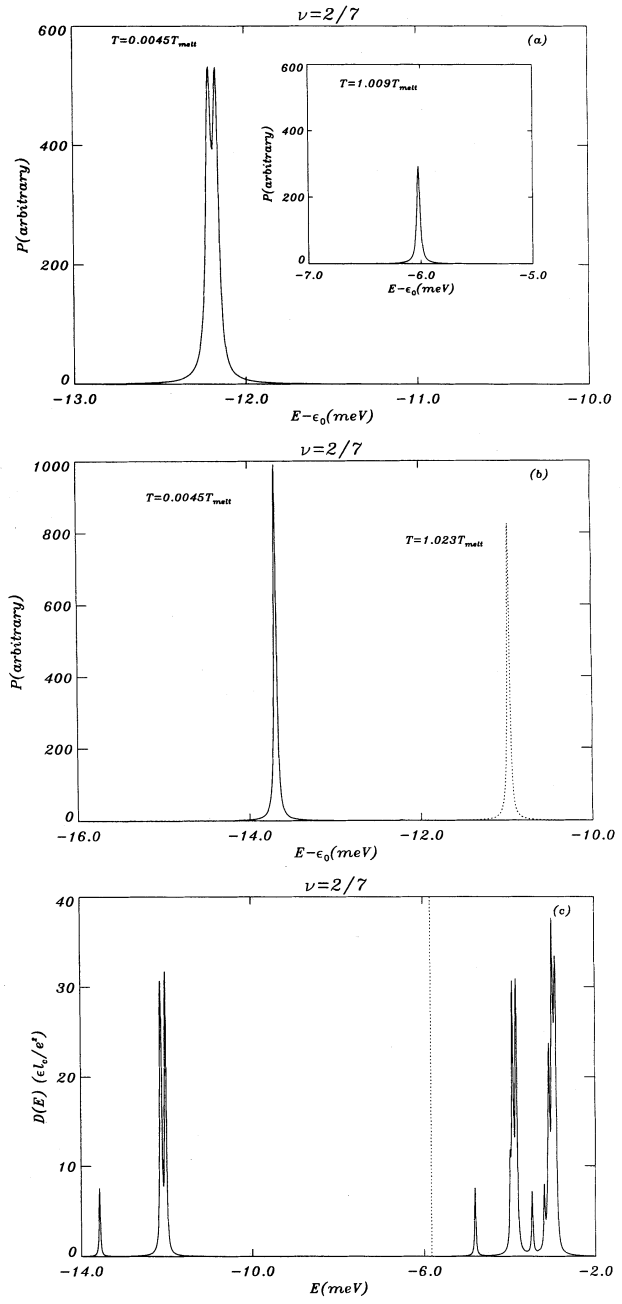

FIG. 1. (a) Photoluminescence for $\nu = \frac{2}{7}$ with no electron-hole interaction, with T below the melting temperature T_{melt}. Inset: Same for T above melting temperature. (b) Photoluminescence for $\nu = \frac{2}{7}$ with electron-hole interaction, below (solid line) and above (dotted line) melting temperature. (c) Electron density of states for $\nu = \frac{2}{7}$ in presence of hole interaction. Dotted line denotes chemical potential.

It should be noted that, in most localized hole experiments [6], the dopant atom is a *neutral* acceptor in its initial state. The interaction of the core hole with the electron gas is then quite weak, leading to a negligible deformation of the WC in its initial state. However, the *final* state of the dopant is charged, which introduces a strong perturbation. The net result of this is that the PL spectrum is still dominated by a single final state, one in which a vacancy is bound to the charged ion. The PL spectrum is thus qualitatively the same as described for the case of a strong initial interaction; details will be reported elsewhere [11].

The difficulty of observing local crystalline order directly in PL for the highly localized hole is clearly related to the fact that a single electron dominates the electron-hole recombination. This problem can be alleviated in principle if the hole is not so strongly localized. We thus consider an itinerant hole in the valence band, a geometry which is only very recently being examined in the WC regime [7]. Typical results for this system are shown in Fig. 2, for filling fraction $\nu = \frac{2}{11}$. For the lowest temperatures, one can only see a single peak in the photoluminescence, essentially because at this filling the splitting between the two filled subbands is too small to resolve numerically [12]. However, an interesting effect

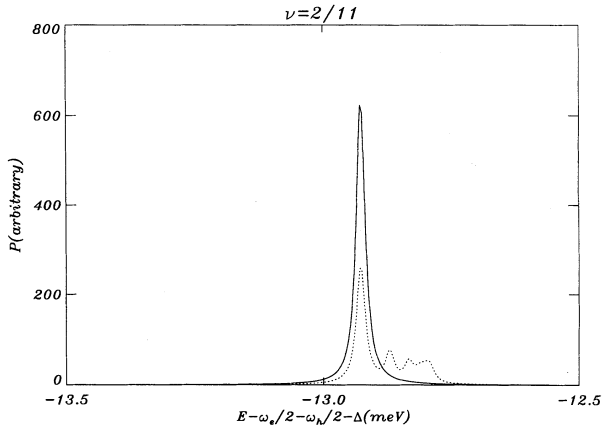

FIG. 2. Photoluminescence for itinerant hole at electron filling $\nu = \frac{2}{11}$, for $T \approx 0.005 T_{\text{melt}}$ (solid line) and $T \approx 0.05 T_{\text{melt}}$ (dotted line). Electron and hole cyclotron frequencies given here by ω_e and ω_h, respectively, and Δ is the conduction-band–valence-band gap.

occurs when the temperature is raised slightly (although not nearly enough to melt the crystal): one then finds that structure in the PL peak is introduced. This turns out to be due to the density of states for the hole. This also moves in the periodic potential of the electron lattice, and so should be expected to have eleven bands as well. Increasing the temperature moderately allows some non-negligible probability for the hole to occupy the higher bands, each adding a new line to the PL spectrum [13]. Once again, observation of this effect would constitute direct confirmation of crystalline order in the 2DEG. We believe that, with improved sample quality, itinerant hole PL experiments should offer the best opportunity to observe this type of structure, which is a direct consequence of the presence of a WC.

Our calculated PL spectrum as the temperature is raised so as to melt the WC also has very interesting behavior [14,15]. In this case, there is an upward shift in the PL peak for the case of a localized hole, as seen in Fig. 1. The increase in energy corresponds directly to the potential energy lost per electron when the carriers are no longer crystallized. What is remarkable about the shift is that it occurs almost precisely at the melting temperature; there is very little motion just above or below the transition. This is in qualitative agreement with experimental observations [7], in which two distinquishable lines are observed, with oscillator strength transferring from the lower to the upper one as the temperature is increased. One could interpret this as finite size domains of the WC with a distribution of melting temperatures, accounting for the continuous transfer of oscillator strength between the two lines. That two such lines are visible in real experiments, rather than a broad continuum PL spectrum, seems consistent with an electrostatic environment for the recombining electrons that is fairly uniform through the sample, indicating that there may be some (substantial) order in the system.

We now outline how we calculate the photoluminescence in the TDHFA. (Details will be given elsewhere [11].) The photoluminescence intensity is given, for a single localized hole state, by

$$P(\omega)=\frac{I_0}{Z}\sum_n\sum_m e^{-E_n/k_BT}|\langle m,0|\hat{L}|n,h\rangle|^2\delta(\omega-E_n+E_m)\,, \tag{1}$$

where $Z=\sum_n e^{-E_n/k_BT}$, $|n,h\rangle$ is a many-body electron state with energy E_n and N electrons where there is a core hole present, $|m,0\rangle$ is a many-body electron state with $N-1$ electrons and energy E_m, ω is the luminescence frequency, and $\hat{L}=\int d^2x\,\psi(x)\psi_h(x)$ is the luminescence operator, with $\psi(x)$ the electron annihilation operator and $\psi_h(x)$ the hole annihilation operator. As written, the initial state is actually higher in energy than the final state, and we find it convenient to rework the problem in terms of absorption rather than emission. To accomplish this, we add a term $H'=-E_0c_0^\dagger c_0$ to the Hamiltonian, where $c_0^\dagger$ creates a localized hole, and take the limit $E_0\to\infty$. It is not difficult to show $P(\omega)=\lim_{E_0\to\infty}P'(\omega-E_0)/n_0(E_0)$, where P' is the absorption spectrum of the new Hamiltonian, and n_0 is the average occupation of the hole state, which just becomes 1 in the limit $E_0\to\infty$. The absorption spectrum is identical to Eq. (1), except one needs to add the energy E_0 to all the quantities E_n in the expression. After standard manipulations [16], one can show that

$$P'(\omega)=\frac{I_0}{\pi}\frac{1}{1-e^{\omega/k_BT}}\mathrm{Im}[R(\omega+i\delta)]\,.$$

The function $R(\omega+i\delta)$ is a response function, which continued to imaginary frequency has the form

$$R(i\omega)=-\int_0^\beta\langle T_\tau L(\tau)L^\dagger(0)\rangle e^{i\omega\tau}\,d\tau\,. \tag{2}$$

To compute this quantity, we consider (for the case of a localized hole state) instead of a single hole a periodic (hexagonal) lattice of them, with a unit cell that contains as many electrons as can be handled numerically. In addition, since the system is in a strong magnetic field, we project the electron part of the Hamiltonian into the lowest Landau level. Because of the symmetry of the system, $R(\omega)$ may be written in the form [17]

$$R(\omega)=\frac{n_h\Omega}{2\pi l_0^2}\sum_{\mathbf{G}}R(\mathbf{G},\omega)e^{-G^2l_0^2/4}$$

if we approximate the core-hole wave functions as delta functions, where n_h is the density of holes, Ω the volume of the system, and the vectors $\mathbf{G}$ are the reciprocal lattice vectors of the superlattice.

Our task is thus to compute the quantity $R(\mathbf{G},\omega)$. To do this, we write down the equation of motion for $R(\tau)$ in terms of its commutator with the Hamiltonian, and then apply a Hartree-Fock decomposition to the resulting expression [18]. The result may be expressed in terms of the Fourier components $R(\mathbf{G},\omega)$, which takes the form (after a very involved calculation [11])

$$\sum_{\mathbf{G}'}[(\omega+i\delta+E_0+\omega_0)\delta_{\mathbf{G},\mathbf{G}'}-B(\mathbf{G},\mathbf{G}')]R(\mathbf{G}',\omega)=\rho(\mathbf{G})\,, \tag{3}$$

where $\omega_0=-\epsilon_0-(1/2\pi l_0^2)\sum_{\mathbf{G}}\rho(\mathbf{G})V(-\mathbf{G})e^{-\mathbf{G}^2l_0^2/4}$, $V(\mathbf{G})$ is the Fourier transform of the electron-hole interaction, $\epsilon_0=\epsilon_h+\omega_e/2$, ϵ_h is the energy of the initial core hole state, ω_e is the cyclotron frequency of an electron, and

$$B(\mathbf{G},\mathbf{G}')=[W(\mathbf{G}-\mathbf{G}')\rho(\mathbf{G}-\mathbf{G}')+n_hV(\mathbf{G}-\mathbf{G}')\times e^{-(\mathbf{G}-\mathbf{G}')^2l_0^2/4}]e^{i(\mathbf{G}\times\mathbf{G}')l_0^2/2}\,,$$

where W is the sum of the direct and exchange interactions, as given in Ref. [18]. We see that the form of R is essentially that of a Green's function, and by inverting Eq. (3) one can show that it has poles at *precisely* the energies of poles in the Green's function for the system in the presence of the external interaction V due to the hole

[19], up to the constant energy shift ω_0 [20,21].

The case of the itinerant hole is treated similarly to the case outlined above, except there is an important simplification: Since the hole density is low at all points in space, it is safe to ignore any deformation of the electron lattice due to the hole. For this situation, we find that Eq. (1) may be written directly in terms of the Green's functions for the electron lattice. Because there are many hole states close in energy on the scale of temperature for the itinerant hole, we also take a thermal average of Eq. (1) over the different hole states that the electrons may decay into. The hole wave functions may be generated by numerically computing its Green's function. Details will be given elsewhere [11].

In summary, we have developed a theory of photoluminescence for the WC in a strong magnetic field. We find that one can use PL to unambiguously demonstrate the presence of a WC, by observing a gap structure associated with the unique energy spectrum of an electron in a periodic potential and a magnetic field. We show that electron-hole interactions tend to close these gaps, and argue that the best situation for finding this structure would be in an itinerant hole experiment.

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[8] A mean-field approach cannot account for correlation effects associated with the Fermi-edge singularity encountered in PL in metals. However, since the WC has no Fermi surface, this is not a problem for our present purpose.

[9] The filling $\nu=\frac{2}{7}$ is higher than the fillings at which the WC is believed to be the ground state of the system. However, our results should be qualitatively the same as for lower fillings, and the use of a larger ν allows us to obtain better numerical accuracy. Results for an experimentally relevant filling ($\nu=\frac{2}{11}$) for the itinerant hole case are given in Fig. 2.

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[13] The gap in the hole density of states is large enough to be resolved, even though the corresponding one in the electron density of states is not. This is due to the exchange interaction between electrons (which has no analog in the single hole problem we need to solve), which tends to close the gap between occupied subbands.

[14] The Hartree-Fock approach to melting cannot account for fluctuation effects that might lead, for example, to a Kosterlitz-Thouless transition. However, since the PL for a core hole is a probe of *local* electron structure, we believe our calculation gives the correct qualitative behavior.

[15] Excited phonon modes at finite temperatures strongly renormalize the electron density modulations, leading to lowered melting temperatures than those calculated in the TDHFA, as well as a broadening of the peak structures in all our figures. However, we do not expect this to affect any of our qualitative conclusions.

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[17] More explicitly,

$$R(\mathbf{G},\omega)=\frac{1}{g}\sum_X e^{-iG_xX+iG_xG_yl_0^2/2}R_{ii}(X,X-G_yl_0^2;\omega)\,,$$

where g is the Landau level degeneracy, X is the guiding center quantum number, and

$$R_{ii}(X_1,X_2;\omega)=-\int_0^\beta\langle T_\tau a_{X_1}(\tau)c_i(\tau)c_i^\dagger a_{X_2}^\dagger\rangle e^{i\omega\tau}\,d\tau\,,$$

with $a_X^\dagger$ creating an electron in state X and $c_i^\dagger$ creating a hole in state i.

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[20] However, the residues of the poles are not the same as for the Green's function. The residues are determined by the overlaps of the mean-field single particle electron wave functions with the hole wave function. It is thus appropriate to think of R as a weighted Green's function. Since $P'(\omega)$ involves the imaginary part of $R(\omega)$, our final result takes the form of a weighted density of states.

[21] Because we have taken the limit $E_0\rightarrow\infty$, we always consider situations in which the initial state of the hole is occupied with probability 1; we thus do not consider excitonic correlations between the electrons and the hole, which would enter here as a nonzero expectation value $\langle\psi(0)\psi_h(0)\rangle$, for a hole tightly bound to a site at the origin. This should be valid so long as the hole is not too close to the electron gas on the scale of the magnetic length, as is the case in most experiments. See A. H. MacDonald, E. H. Rezayi, and D. Keller, Phys. Rev. Lett. **68**, 1939 (1992).