

# Long-lived charged multiple-exciton complexes in strong magnetic fields

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We consider the charged exciton complexes of an ideal two-dimensional electron-hole system in the limit of strong magnetic fields. A series of charged multiple-exciton states is identified and variational and finite-size exact-diagonalization calculations are used to estimate their binding energies. We find that, because of a hidden symmetry, bound states of excitons and an additional electron cannot be created by direct optical absorption and, once created, have an infinite optical recombination lifetime. We also estimate the optical recombination rates when electron and hole layers are displaced and the hidden symmetry is violated. [S0163-1829(96)51928-4]

Two-electron atoms are among the simplest systems in which electronic exchange and correlation play an important role. Studies of these systems<sup>1</sup> have played a vital role in the development of practical techniques for accurate calculations in many-electron atoms and molecules. Among two-electron atomic systems the  $H^-$  ion, which is barely bound, is the most difficult to describe accurately. The semiconductor analogs<sup>2</sup> of the  $H^-$  ion are the charged exciton states,  $X^-$  and  $X^+$ , for which, respectively, two electrons are bound to a single hole and two holes are bound to a single electron. Recently there has been considerable experimental interest<sup>3-6</sup> in the charged exciton states of two-dimensional (2D) electron systems, in part because the reduced dimensionality leads to relatively larger binding energies. At zero magnetic field, the charged exciton has a single spin-singlet bound state and the binding energy seen in 2D systems in recent experiments is in reasonable agreement with theory.<sup>7</sup> Experiments have also demonstrated that for 2D charged excitons an additional spin-triplet bound state becomes stable in sufficiently strong external magnetic fields. In this paper we address the strong magnetic field limit for 2D charged multiple-exciton complexes.<sup>8</sup> We find that in addition to the spin-triplet charged single-exciton states there exist a series of charged bound multiple-exciton complexes. We also find that, because of a hidden symmetry in the Hamiltonian of this system, these states have an infinite optical recombination time for an ideal system. We propose that this unanticipated anomaly should lead to observable effects in time-resolved photoluminescence experiments.

In the strong magnetic field limit we consider, all particles in the charged exciton complex are confined to their lowest Landau levels and have their spins aligned with the magnetic field. Our basic conclusions follow from an exact mapping<sup>9,10</sup> between spin-polarized particle-hole systems and spin-1/2 electron systems, which holds in this limit. Our analysis exploits recent advances<sup>11</sup> in understanding the elementary charged excitations of the incompressible ground state in spin-1/2 electron systems at Landau level filling factor  $\nu \equiv N/N_\phi = 1$ . [Here  $N_\phi = A/(2\pi\ell^2) = AB/\Phi_0$  is the Landau level degeneracy,  $\ell$  is the magnetic length, and  $\Phi_0$  is the electronic magnetic flux quantum.] In the mapping the hole Landau level is associated with the minority ( $\downarrow$ ) spin

Landau level and the occupied states in the electron Landau level are associated with empty states in the majority spin ( $\uparrow$ ) Landau level.<sup>12</sup>

$$N_h = N_\downarrow, \quad (1)$$

$$N_e = N_\phi - N_\uparrow.$$

For example, charge neutral states ( $N_e = N_h$ ) of electron-hole systems correspond to  $\nu = 1$  ( $N = N_\uparrow + N_\downarrow = N_\phi$ ) states of the spin-1/2 system. Generally the total charge ( $Q \equiv N_e - N_h$ ) and total particle-number ( $L \equiv N_e + N_h$ ) of the electron-hole system are given by  $Q = N_\phi - N$  and  $L = Q + N - 2S_z$  where  $2S_z = N_\uparrow - N_\downarrow$ .  $X^-$  states have  $Q = 1$  and  $L = 3$  so they correspond to spin-1/2 states with one particle removed from a full Landau level and  $S_z = N/2 - 1$ . The eigenstates of electron-hole and spin-1/2 systems have a one-to-one correspondence under this mapping<sup>9</sup> and corresponding eigenenergies differ by a known constant:

$$E_{eh} = \tilde{E}_{1/2} - N_e I, \quad (2)$$

where  $I$  is the binding energy of an isolated exciton in this limit. [For an ideal 2D system  $I = (\pi/2)^{1/2}(e^2/\epsilon\ell)$ .] Here all energies are measured with respect to the corresponding non-interacting electron values which increase with field in proportion to the quantized kinetic energies of the Landau levels and  $\tilde{E}_{1/2}$  is the energy of the spin-1/2 system measured with respect to the energy of the fully spin-polarized  $\nu = 1$  state.

Recently, progress has been made in understanding the elementary charged excitations of the  $\nu = 1$  ( $N = N_\phi$ ) ground state for spin-1/2 particles. This ground state has total spin quantum number  $S = N/2$  and is therefore spin aligned by an arbitrarily weak magnetic field. Its charged excitations have the unusual property, first noticed in numerical exact diagonalization calculations<sup>13</sup> and dramatically evident in recent experiments,<sup>14</sup> that they can carry a large spin. It can be shown<sup>15</sup> that, for  $N = N_\phi - 1$ , a single low-energy spin-multiplet with orbital degeneracy  $\approx N_\phi$  and energy  $\tilde{E}_{1/2} = \epsilon_K$  occurs with  $S = N/2 - K$  for each  $K = 0, 1, \dots$ .  $\epsilon_{K=0} = I$  and for large  $K$  these elementary charged excitations of the  $\nu = 1$  state can be identified<sup>11,15,16</sup> with the topologically charged Skyrmion spin textures of the underlying

ferromagnetic  $\nu=1$  ground state.<sup>17</sup> In the  $K \rightarrow \infty$  limit  $\epsilon_K \rightarrow 3I/4$ . Numerical Hartree-Fock<sup>18</sup> and exact diagonalization<sup>19</sup> calculations indicate that  $\epsilon_K$  decreases monotonically with  $K$  between these limits. Using the mapping, since  $S_z = N/2 - 1$  states occur in both  $S = N/2$  and  $S = N/2 - 1$  multiplets, it follows that for  $N_e = 2$  and  $N_h = 1$  there are two low-lying states with energies  $E_{eh} = \epsilon_{K=0} - 2I = -I$  and  $E_{eh} = \epsilon_{K=1} - 2I$ . The first of these states corresponds to a single exciton and an unbound electron ( $X + e$ ) while the second has lower energy and corresponds to a single exciton bound to an electron ( $X_1^-$ ) with binding energy  $\epsilon_{K=0} - \epsilon_{K=1}$ . *There are no other bound states between an electron and a single exciton.*<sup>20</sup> (This lone  $X_1^-$  bound state in the strong magnetic field limit contrasts with the solitary singlet and three triplet<sup>21</sup> bound  $D^-$  states for two 2D electrons bound to an external charge.) The existence of a bound  $X^-$  state was noticed previously in the numerical calculations of Ref. 8. The same analysis can be carried out for larger values of  $K$  and has unexpected implications:  $K$  excitons and an additional electron form a bound  $X_K^-$  complex with energy  $(\epsilon_K - \epsilon_{K=0}) - KI$ . The ionization energy of this complex is  $\epsilon_{K=0} - \epsilon_K$  and dissociation energy is  $\epsilon_K - \epsilon_{K-1}$  for the reaction  $X_K^- \rightarrow X_{K-1}^- + X$ . Note that without the excess charge there are no bound multiple-exciton complexes in the strong magnetic field limit. (All binding energies are independent of both electron and hole masses in the strong magnetic field limit.)

To estimate binding energies and optical matrix elements we perform microscopic calculations using the symmetric gauge which has single-particle states with definite angular momentum in electron and hole Landau levels. The wave function which describes the state  $X + e$  (an exciton and an unbound electron) is given in the corresponding occupation number representation by<sup>9</sup>

$$|\Psi_{X+e}\rangle = \sum_{m=1}^{N_\phi} e_m^\dagger h_m^\dagger e_0^\dagger |0\rangle, \quad (3)$$

where  $e_m^\dagger$  creates an electron with angular momentum  $-m$  and  $h_m^\dagger$  creates a hole with angular momentum  $m$ . Estimates of the binding energies of the  $X_K^-$  complexes for small  $K$  can be obtained by using the following variational wave functions:

$$|\Psi_{X_K^-}\rangle = \sum_{m_K > \dots > m_1 = 0}^{N_\phi} \left[ \prod_{k=1}^K \frac{a_{m_k}}{\sqrt{(m_k + 1)}} e_{m_k+1}^\dagger h_{m_k}^\dagger \right] e_0^\dagger |0\rangle. \quad (4)$$

This form for the variational wave function is motivated by the fact that for  $a_m$  independent of  $m$  it becomes exact<sup>15</sup> in the case of  $\delta$  function repulsive interactions between the electrons and attractive interactions between the electrons and the hole. The wave functions with constant  $a_m$  also become exact in the large- $K$  limit where they correspond in the spin language to the classical field theory Skyrmions.<sup>15,16</sup> For the physically relevant case of Coulombic electron-electron and electron-hole interactions we let  $a_m \propto \exp(-\lambda m)$  where  $\lambda$  is a variational parameter. This form of the wave function is motivated by our expectation that longer-range interactions would favor more compact bound states and by

TABLE I. Binding energies for the simplest  $X_K^-$  exciton complexes. The variational values have been obtained by optimizing the parameter  $\lambda$ . The binding energy for  $K \rightarrow \infty$  is related via the mapping to the energy of Skyrmion states in quantum Hall ferromagnets. All energies are in units of  $(e^2/\ell)$ .

$K$	$\lambda$	Variational binding energy
1	0.071	0.0529
2	0.045	0.0828
3	0.034	0.1018
4	$\approx 0.03$	$\approx 0.117$
$\infty$	0	0.313

comparison with small system exact-diagonalization calculations.<sup>22</sup> Table I shows the binding energies for the simplest  $X_K^-$  complexes calculated from the wave functions (4) by optimizing the variational parameter  $\lambda$ . As expected, the exponential factor becomes irrelevant for  $K \rightarrow \infty$  where the classical field theory energy becomes exact. The validity of this wave function for  $K=1$  has been checked by comparing with results obtained by exact diagonalization of the Hamiltonian. It is possible to diagonalize the Hamiltonian of a system sufficiently large ( $N_\phi \leq 80$ ) to make finite-size corrections negligible. The exact binding energy obtained in this way,  $0.0545(e^2/\ell)$ , is quite close to the variational one,  $0.0529(e^2/\ell)$ . Moreover, the overlap of the variational wave function (4) for  $K=1$  with the exact one is about 99%. The estimate for  $K=1$  is also in qualitative agreement with existing experiments. Electron-electron and electron-hole correlation functions for the  $X_1^-$  state are illustrated in Fig. 1 and compared with the electron-hole correlation function of an isolated bound exciton. In this figure we plot the probability of finding one electron or the hole at radius  $r$  when the other electron is at the origin. We see from this figure that the quantum mechanical sharing of the hole among the two electrons makes binding possible. No binding occurs if the elec-

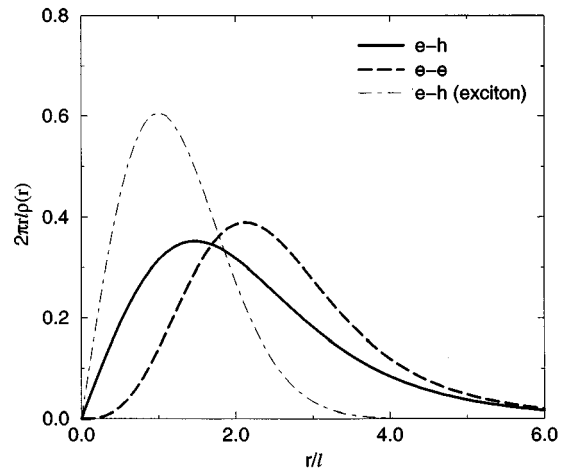


FIG. 1. Electron-electron and electron-hole correlations in  $X_1^-$  state and an isolated exciton. These graphs show the hole density and the density of the other electron with one electron fixed at the origin. Note that the hole density at the origin is finite while the other electron density vanishes.

tron and hole are treated as classical particles. The spin alignment of the two electrons guarantees that the electrons do not have a large probability of being close together; in this strong field limit, there is no bound  $X^-$  when the electrons form a singlet even if the Zeeman energy is discounted. Note that since the  $X_1^-$  is a charged particle its states occur in manifolds with degeneracy  $\sim N_\phi$  like the Landau level manifolds for an isolated electron.

The mapping between spin-1/2 and electron-hole systems has interesting implications for the optical recombination matrix elements of  $X_K^-$  states. Let us focus again on the simplest  $X_1^-$  state. As mentioned above, the eigenstates of the spin-1/2 system occur in spin multiplets because of the spin-rotational invariance of the Hamiltonian and the unbound  $X+e$  and bound  $X_1^-$  states correspond, respectively, to the  $S_z = N/2 - 1$  members of the  $S = N/2$  and  $S = N/2 - 1$  multiplets. The optical recombination operator,  $R = \sum_m e_m h_m$ , maps<sup>9</sup> to the total spin-raising operator of the spin-1/2 system and therefore *annihilates* the bound  $X_1^-$  state. In this ideal model the optical recombination time of the  $X_1^-$  state is infinite.<sup>23</sup> In real systems there are a number of effects which will make the optical recombination time finite, including Landau level mixing effects at finite magnetic field strengths which depend in detail on the complicated valence-band electronic structure in a quantum well. Even at infinite field the hidden symmetry is violated when electron-hole interactions differ from electron-electron interactions by more than a change of sign, i.e., when the envelope functions for electron and hole Landau layers are not identical. This is often the case in quantum-well electron-hole layers,<sup>24</sup> for example, when electrons and holes are intentionally displaced by an electrostatic potential to inhibit optical recombination. In Fig. 2 we show results for the optical recombination rate of  $X_1^-$  states obtained from exact diagonalization calculations for the case of narrow electron and hole layers separated by a distance  $d$ . The  $X_1^-$  recombination rate is normalized to the recombination rate of an isolated exciton. In this figure the binding energy of  $X_1^-$  is also shown. The  $X_1^-$  state is bound only for  $d < \ell$ . Results are shown for  $N_\phi = 50$ . The recombination rate at small  $d/\ell$  is small and still decreasing with system size. In realistic experimental situations, the popula-

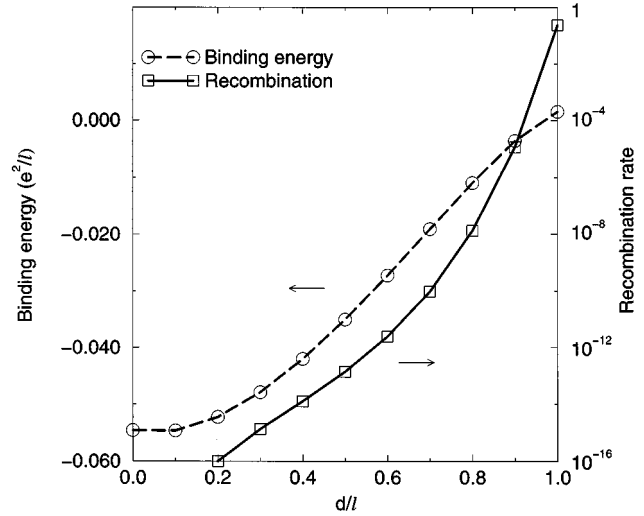


FIG. 2. Normalized optical recombination rates and binding energy as a function of electron-hole layer separation. The binding energy is in units of  $(e^2/\ell)$ . These results were obtained from exact diagonalization calculations for  $N_\phi = 50$ . The recombination rates at small  $d/\ell$  for this value of  $N_\phi$  are still decreasing with increasing  $N_\phi$ .

tion of excitons and charged excitons present in the system at strong magnetic fields will depend in part on kinetic effects not discussed here. Both excitons and charged excitons will tend to be localized; the finite-size effects we find in our calculations indicate that the actual recombination rate of a charged exciton will be sensitive to the disorder potential it experiences. Nevertheless, it follows from our work that a dramatic difference between neutral and charged exciton recombination times should occur and be observable in time-resolved photoluminescence experiments.

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