## Excitons in dense two-dimensional electron-hole magnetoplasmas

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We report clear excitonic effects in the optical spectra of neutral, dense magnetoplasmas in an  $In_{0.53}Ga_{0.47}As/InP$  quantum well at low temperatures. The observed suppression of the density dependence of the uppermost occupied  $(j_e = j_h = N)$  Landau-level transition energies in the range of filling factors N < v/2 < N+1 is direct evidence that electrons and holes from the Nth Landau levels bind into magnetoexcitons. The effect disappears with increasing temperature. Good agreement between experimental results and theoretical calculations is obtained by including both the inter-Landau-level coupling and screening effects.

Electron-hole correlations in a neutral electron-hole (e-h) plasma are expected to cause strong excitonic effects at sufficiently low temperatures.<sup>1,2</sup> In particular, bound e-h pairs (excitons) may condense at the Fermi level, in analogy with Cooper pairs in superconductors.<sup>3</sup> Since the excitons are neutral the condensed phase is insulating and called "excitonic insulator."<sup>3</sup> The excitonic insulator state has never been identified experimentally in bulk crystals. Semiconductor quantum wells (QW's) are promising new candidates for the observation of a condensed excitonic phase mainly because of the increased exciton binding energy. Clear signatures of exciton condensation have been predicted at high plasma densities,<sup>4,5</sup> which are preceded by excitonic effects even above the critical temperature.<sup>4-6</sup>

Excitons can be observed in high-density plasmas only if the plasma temperature is low enough to avoid immediate thermal ionization. Indications of excitonic effects have been deduced previously from the magnetoluminescence of highly excited GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's.<sup>7,8</sup> However, the excitonic effects claimed at the reported high electron temperatures of more than 300 K are difficult to reconcile with hitherto-accepted theoretical concepts<sup>9</sup> and are still a matter of controversy.<sup>10,11</sup> On the other hand,  $In_xGa_{1-x}As/InP$  QW's display significantly larger carrier lifetimes<sup>12</sup> and hence lower plasma temperatures than GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW's.<sup>13</sup> These conditions should strongly favor the observation of excitonic effects in the (magneto-) optical spectra.

We have performed experimental and theoretical studies of excitonic effects in the magnetoluminescence spectra of  $In_{0.53}Ga_{0.47}As/InP$  single QW's. At low temperatures we observe a significant change in the plasma density dependence of the Landau-level transition energies when the Fermi energy is tuned through a Landau level. For empty and filled Landau levels the transition energy decreases with increasing density. In contrast, Landau levels at the Fermi energy display no significant density dependence. Quantitatively this behavior can be understood by the filling-factor-dependent interplay of the Pauli repulsion and the exchange interaction of the magne-However, previous theories<sup>14,15</sup> toexciton states. magnetic quantum for the limit developed  $(\gamma = a_B / l_H \gg 1)$  are insufficient for a quantitative description of the present experiments carried out at intermediate magnetic fields ( $\gamma \sim 2$ ). Here  $a_B$  and  $l_H$  are the exciton Bohr radius and magnetic length, respectively. We have therefore carried out calculations which take into account all interactions with higher Landau levels and the finite width of the quantum well and which provide the correct limiting behavior at infinite magnetic fields.<sup>14</sup> Agreement between the experimental and theoretical results is achieved only by proper consideration of screening. The strong excitonic effects which we are able to identify suggest that carrier temperatures of about 50 K are not very far from the transition temperature to the excitonic insulator state.<sup>16</sup>

We have measured photoluminescence excitation spectra for the empty single QW and photoluminescence spectra of the magnetoplasma in the same QW for a wide range of densities, temperatures, and magnetic fields. The undoped InP/In<sub>0.53</sub>Ga<sub>0.47</sub>As/InP single-QW structures (well width 15 nm) were grown by low-pressure metal-organic vapor phase epitaxy (MOVPE).<sup>18</sup> The nonequilibrium carriers were excited with an Ar<sup>+</sup>-ion laser ( $\lambda$ =514.5 nm). The emitted light was dispersed

46 12 765



FIG. 1. Emission spectra of the electron-hole magnetoplasma in an  $In_{0.53}Ga_{0.47}As$  single QW (well width 15 nm) for different excitation intensities at 8.65 T and  $T_{bath}=2$  K. All spectra are presented in the same scale.

with a grating monochromator and detected by a cooled Ge detector. The sample was mounted in a He cryostat with a superconducting solenoid (H < 8.7 T). Great care was taken to provide a high spatial and temporal *e*-*h* plasma homogeneity. In particular, lateral inhomogeneity in the plasma density was suppressed by preparing  $50 \times 50$ - $\mu$ m mesa structures and transient effects were avoided by using a cw excitation laser.

Figure 1 shows the emission spectra of the InP/In<sub>0.53</sub>Ga<sub>0.47</sub>As/InP single QW at  $T_{bath} = 2$  K and H=8.65 T for different excitation levels, i.e., as a function of the carrier density  $n_{eh}$ . For small  $n_{eh}$  a single emission line is observed which corresponds to the radiative recombination of the 0-0 magnetoexciton consisting of the electron and hole in the zeroth Landau levels  $(j_e = j_h = 0)$ . With increasing density new lines appear in the emission spectra, corresponding to allowed  $(j_e = j_h)$ transitions between the occupied Landau levels. The observed saturations of the j-j emission line intensities with density indicate the complete filling of *j*th electron and hole Landau levels. Figure 1 shows that the effective electron temperature  $T_e$  is larger than the lattice temperature (=2 K). From the ratio of the emission line intensities for large values of  $j_{e,h}$ ,  $T_e$  is estimated to be in the range 30-50 K for  $n_{eh} \approx 10^{12}$  cm<sup>-2</sup>. This is clearly smaller than the Coulomb energy (15 meV at H=8.65 T), and excitonic effects can be expected to become important.

The density dependence of the optical transition energies,  $\hbar \omega_j(n_{eh})$ , at H=8.65 T is displayed in Fig. 2. The experimental points were determined from the emission spectra except for the values of  $\hbar \omega_j(n_{eh}=0)$ , which have been measured by photoexcitation. The plasma densities were determined by the filling of the Landau levels, which is directly presented by the number of emission lines as the oscillator strength of the different Landaulevel transitions was calculated to be almost identical. Figure 2 shows that a strong reduction of the transition energies  $\hbar \omega_j$  occurs both between the empty,  $j > \nu/2$ , and completely filled,  $j < \nu/2 - 1$ , Landau levels with in-



FIG. 2. Measured (points) and calculated (lines) Landau-level transition energies vs density at 8.65 T. The electronic temperature of 50 K estimated from the experimental spectra has been used in the calculation. The HF and PP lines correspond to Hartree-Fock and plasmon-pole approximations, respectively. The lines labeled HF (self-energy) correspond to calculations without excitonic corrections, and are in clear disagreement with experiment. Inset: dependence of the emission line spacing  $\Delta_{10}$  on the filling factor  $\nu/2$ .

creasing carrier density. In contrast, the transition energy remains approximately constant when the level is partly filled (i.e., coincides with the Fermi energy v/2-1 < j < v/2). Here v is the filling factor and the factor 2 accounts for the spin degeneracy of the Landau levels. An increase of temperature leads to a qualitative change of the density dependence of the transition energies. As displayed in Fig. 3, at temperatures of 100 K the pronounced plateaus in  $\hbar \omega_j(n_{eh})$  have disappeared completely. As a consequence, the energy difference  $\Delta_{j,j-1} = \hbar \omega_j - \hbar \omega_{j-1}$  turns out to be temperature dependent. The inset of Fig. 3 displays the temperature dependence of  $\Delta_{10}$  for a constant plasma density  $n_{eh} = 4 \times 10^{11}$  $cm^{-2}$  ( $v/2 \approx 1$ ). We observe a pronounced increase of  $\Delta_{10}$  for increasing temperature which is another indication of excitonic effects.

The dependence  $\hbar \omega_j(n_{eh})$  at low temperature can be qualitatively explained if the neutral magnetoplasma is described in terms of magnetoexcitons.<sup>13,15,17,18</sup> The *j*-*j* magnetoexciton is formed by the  $j_e$  electron and  $j_h$  hole at the Landau levels  $j_e = j_h = j$ . The interaction of magnetoexcitons within one Landau level is repulsive at small distances  $r < l_H$  ( $l_H$  determines the magnetoexciton radius) due to the Pauli exclusion principle and attractive at large distances due to the exchange interaction. In the magnetoplasma, these two contributions to the scattering amplitude cancel exactly in the high-magnetic-field limit and the magnetoexcitons can be considered as noninteracting.<sup>14</sup> Note that a partial cancellation of selfenergy and excitonic vertex correction has been discussed in Refs. 1 and 2 in the low-density limit and at zero mag-



FIG. 3. Density dependence of measured (circles) and calculated (lines) Landau-level transition energies at 8.65 T and T=100 K. Full and dashed lines correspond to Hartree-Fock (HF) and plasmon-pole (PP) approximations, respectively. The theoretical data have been shifted by  $\approx 10$  meV to take into account the lattice temperature-dependent band-gap shrinkage. Inset: the emission line spacing  $\Delta_{10}$  for  $n_{eh} = 4 \times 10^{11}$  cm<sup>-2</sup>  $(\nu/2 \approx 1)$  as a function of the temperature ( $T_e$  for theory,  $T_{bath}$ for experiments).

netic field. Thus the density independence of the Landau-level transition energies at the Fermi level suggests that electrons and holes from the uppermost occupied Landau levels are bound into magnetoexcitons. The exact cancellation of the two terms does not hold for the interaction of magnetoexcitons from different  $i \neq j$  Landau levels. In particular, if the Fermi level is increased above a given Landau level this does not lead to a further increase of the influence of the Pauli repulsion. This is due to the fact that, with increasing difference i-j, the overlap of the particle wave functions in k space vanishes. On the other hand, the exchange interaction with carriers in the newly populated bands is significant.<sup>18,19</sup> As a consequence, the filling of states at the Fermi level causes a decrease of transition energies of the Landau levels which are fully occupied or completely empty.

For a more quantitative analysis of the transition energies calculations have been carried out in two approximation schemes labeled Hartree-Fock (HF) (Ref. 4) and plasmon pole (PP), taking into account all interactions with higher Landau levels and the finite width of the quantum well. In the PP calculations the dynamical screening effects in the self-energies were included in the plasmon-pole approximation,<sup>20</sup> while excitonic effects were incorporated by summing ladder diagrams for the statically screened electron-hole interaction; the temperature and magnetic-field-dependent polarizability has been calculated following Ando and Uemura.<sup>21</sup> Details of the calculations have been published in Ref. 22. The results of the calculations at T=50 K are shown in Fig. 2 by the solid (HF) and dashed (PP) lines. Effective masses  $m_e = 0.05$  and  $m_h = 0.2$  and a static dielectric constant of 12.5 have been adopted.

Figure 2 shows that the behavior of empty Landau lev-

els is well described by both the HF and PP calculations. The observed reduction of the 1-1 and 2-2 magnetoexciton energies under complete filling of the zero or zero and first Landau levels, respectively, is mainly the (exchange) self-energy effect discussed above. The small deviation of experiment and theory at higher Landau levels may be caused by the nonparabolicity of the valence and conduction bands. For partially filled Landau levels both HF and PP calculations predict the suppression of the density dependence of the transition energies due to excitonic corrections. However, quantitative predictions of the HF and PP calculations are different. In the HF approximation the inter-Landau-level mixing causes a (net) repulsive interaction between magnetoexcitons and, hence, an increase of the transition energy with  $n_{eh}$ . If the carrier temperature is reduced from 50 to 10 K even a very pronounced blueshift linear in  $n_{eh}$  is found<sup>22</sup> in the HF calculations. On the contrary, in the PP approximation screening effects dominate and transition energies always decrease with  $n_{eh}$ .

At low temperature the experimental curve is located between the two calculations. Therefore, screening effects ignored in the HF approximation are important but somewhat overestimated in the PP approximation. The static screening approximation used in the PP calculation of the vertex correction is mainly suspect for the hole contribution, since the dynamical response should be retarded by the relatively heavy mass.<sup>22</sup>

To emphasize the importance of the excitonic effects, the HF calculations taking into account self-energy only (without excitonic corrections) are also shown in Fig. 2. The monotonic reduction of  $\hbar \omega_j$  with density predicted by the calculation is in complete disagreement with experimental results exhibiting the density independence of Landau-level transition energies at the Fermi level.

As displayed in Fig. 3, the data at high temperatures (100 K) are described correctly by the PP approximation for all densities investigated. The HF calculation predicts only a very small reduction of the transition energies for increasing  $n_{eh}$ , in significant disagreement with the experiment. Finally, Fig. 2 also shows that screening effects are most important for completely filled Landau levels. Again, the HF approximation strongly overestimates the decrease of transition energies with plasma density, whereas the PP calculations are in reasonable agreement with the experimental data.

The superiority of the PP over the HF approximation is emphasized in the difference  $\Delta_{10}(n_{eh})$  shown in the inset of Fig. 2. The experiment shows that after a strong decrease of  $\Delta_{10}$  in the density range  $\nu/2 < 1$  caused by the exchange interaction of the j=1 e-h pair with particles at the zeroth Landau level, the magnitude of  $\Delta_{10}$  increases monotonically with density in the whole range  $1 \le \nu/2 \le 5$ . The PP approximation describes this fact well, whereas the HF calculations predict a strong maximum at  $\nu/2 \approx 2$  and a decreasing  $\Delta_{10}$  at the higher filling factor (i.e., densities). This maximum reflects the known HF artifact of vanishing electron mass at the Fermi level.

The disappearance of plateaus in the dependences  $\hbar\omega_j(n_{eh})$  at elevated temperatures and temperature dependence of  $\Delta_{10}$  at fixed  $n_{eh}$  displayed in Fig. 3 is con-

In conclusion, we have reported experimental and theoretical evidence of strong excitonic effects in confined electron-hole magnetoplasmas at low temperatures and high densities. Since excitonic effects at the Fermi energy are precursors of the transition to an excitonic insulator state, our results suggest that the observation of conden-

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sation phenomena will come within reach of experiments in the near future. We hope that our results encourage and provide additional momentum to the quest for superconductivity in coupled quantum wells.<sup>23,24</sup>

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