

## Longitudinal-optical phonon and shake-up excitations in the recombination spectra of semiconductor quantum wells

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The observation of both LO-phonon and inter-Landau-level magnetoplasmon shake-up excitations in the magnetophotoluminescence spectra of quantum wells containing high densities of free carriers is reported. The inter-Landau-level magnetoplasmon excitations occur at energies greater than the bare Landau-level separations, in agreement with the predictions of many-body theory. Clear evidence for LO-phonon-magnetoplasmon coupled-mode behavior is observed.

The recombination of a high-density electron gas with photocreated minority carrier holes can lead to a number of different final-state excitations of the system. For example, in semiconductor quantum wells (QW's) with which this paper is concerned, longitudinal-optical- (LO) phonon lattice excitations<sup>1-3</sup> and Fermi-sea particle-hole pair shake-up excitations<sup>4</sup> have both been reported in the last few years.<sup>1,2,4</sup> For metals, both phonon and shake-up excitations have been discussed to explain the observation of asymmetric line shapes in x-ray photoemission spectra, in addition to the observation of collective-mode plasmon satellites.<sup>5</sup> For bulk semiconductors the observation of a plasmon satellite in photoluminescence (PL) spectra has also been suggested.<sup>6</sup>

The present work reports the observation of both LO-phonon satellite and shake-up satellite recombination in the magneto-PL spectra of  $\text{Al}_y\text{Ga}_{1-y}\text{As-In}_x\text{Ga}_{1-x}\text{As-GaAs}$  modulation-doped QW's. In magnetic field the conduction- and valence-band densities of states are quantized into discrete Landau levels (LL's). The use of magnetic field is critical for the observation of the LO-phonon and shake-up excitations in high-density systems since the otherwise broad PL spectrum then breaks up into discrete peaks. As a result both the lattice and Fermi-sea processes give rise to a series of discrete peaks to lower energy, thus permitting spectroscopic study of the various excitations.

It has been shown previously that the strength of both the LO-phonon<sup>1,2</sup> and shake-up satellites<sup>4</sup> increases with the localization of the photocreated holes in the system. The hole localization is determined by the disorder in the structure. The present samples are of high perfection with relatively weak disorder. Nevertheless both the lattice LO-phonon and Fermi-sea shake-up excitations are observed weakly in the spectra. Clear effects of interaction between the shake-up and LO-phonon satellites are observed as the magnetic field is varied.

The experiments were carried out at 4.2 K on a series of modulation-doped, pseudomorphic strained-layer  $\text{Al}_y\text{Ga}_{1-y}\text{As-In}_x\text{Ga}_{1-x}\text{As-GaAs}$  QW's ( $y \approx 0.23$ ,  $x \approx 0.10$ ) with electron densities in the range

$(7-10) \times 10^{11} \text{ cm}^{-2}$ .<sup>7,8</sup> Attention is concentrated on a particular sample with  $n_s = 7.2 \times 10^{11} \text{ cm}^{-2}$ , and well width 150 Å. PL was excited by 25 mW/cm<sup>2</sup> of 633-nm radiation from a He-Ne laser.

The PL spectrum from the sample with  $n_s = 7.2 \times 10^{11} \text{ cm}^{-2}$  at  $B = 0$  is shown in Fig. 1(a). It shows a peak at 1.417 eV together with a long tail to higher energy. The electron Fermi energy  $E_F$  (25.4 meV) is marked on the spectrum, corresponding to  $n_s = 7.2(\pm 0.2) \times 10^{11} \text{ cm}^{-2}$  for an electron effective mass of  $0.068m_0$ .<sup>7</sup> The PL intensity falls off very rapidly from its peak at 1.417 eV with only a very weak feature at  $E_F$ . This is characteristic of a QW of low disorder with electron-hole recombination being fully allowed only for vertical transitions close to  $k = 0$ .<sup>9,10</sup> Transitions from electrons in states up to  $k_F \approx 2 \times 10^6 \text{ cm}^{-1}$  are observable due to the residual disorder which breaks translational symmetry and permits weakly  $\Delta k \neq 0$  transitions; the finite temperature of the photocreated holes (20-25 K) also permits  $k \neq 0$  transitions.<sup>11</sup>

In Figs. 1(b) and 1(c) at 4.1 and 9.4 T, respectively, the PL spectrum breaks up into a series of LL transitions, labeled  $(N_e, N_h)$  arising from recombination between electrons and holes in LL's of index  $N_e$  and  $N_h$ , respectively. The dominant transition (0,0) arises from electrons and holes in the lowest-energy states. Both Landau quantum-number-allowed ( $N_e - N_h = 0$ ) and nominally forbidden  $N_e - N_h = +1, +2$  transitions are observed to higher energy. Transitions such as (1,1), (2,2) arise due to the weak thermal population of  $N_h = 1, 2$  levels.<sup>11</sup> The  $(N_e, 0)$  ( $N_e > 0$ ) transitions are allowed by the weak disorder for the same reasons that  $\Delta k \neq 0$  transitions are allowed at  $B = 0$ .<sup>12,13</sup>

The transition energies are plotted in Fig. 2 as a function of magnetic field. The energy spacing between the  $(N_e, 0)$  peaks is given by the electron cyclotron energy  $\hbar\omega_c = \hbar eB / m_e^*$ , with  $m_e^* = 0.068m_0$ .<sup>7,13</sup> The splitting between (1,0) and (1,1), (2,0), and (2,1) is given by the hole LL splitting with  $m_h^* = 0.15m_0$ , as expected in a strained-layer  $\text{In}_x\text{Ga}_{1-x}\text{As}$  QW with  $x \approx 0.1$ .<sup>13</sup> LL filling

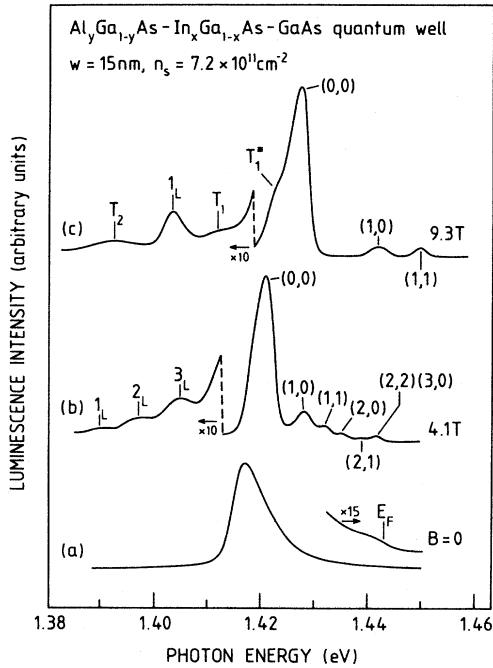


FIG. 1. PL spectra at 4.2 K for quantum well with  $n_s = 7.2 \times 10^{11} \text{ cm}^{-2}$ , at  $B = 0, 4.1,$  and  $9.3 \text{ T}$ . For  $B = 0$  (a) a broad PL band arising from recombination of electrons in states from  $E = 0$  up to  $E_F$  is observed. In finite  $B$  (b) and (c) the spectrum breaks up into Landau-level transitions labeled  $(N_e, N_h)$ . To lower energy LO-phonon satellite ( $N_L$ ) and shake-up excitations ( $T_n$ ) are observed.

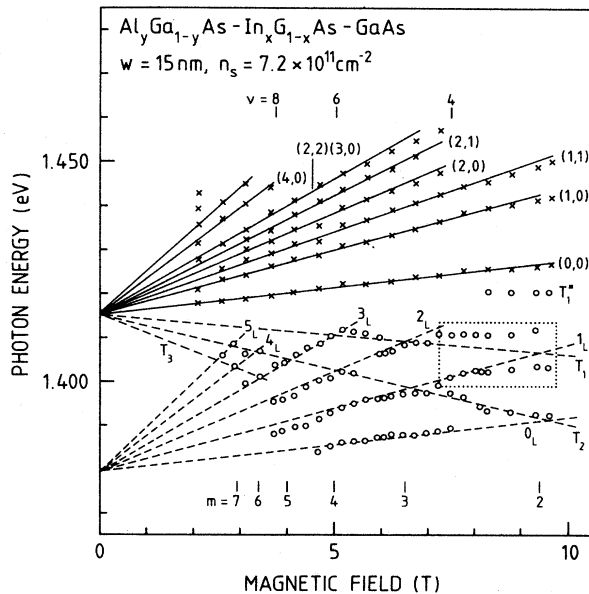


FIG. 2. PL transition energies against magnetic field for sample of Fig. 1. Transitions between electron and hole Landau levels  $(N_e, N_h)$  are indicated by crosses (full lines). To lower energy (open circles, dashed lines) LO-phonon satellites ( $N_L$ ) and shake-up satellites ( $T_n$ ) of the  $(N_e, 0)$  transitions are observed. Resonant coupling between the  $T_1$  and  $1_L$  satellites is seen in the boxed region between 7 and 10 T. Landau-level filling factor ( $\nu$ ) and magnetophonon resonance ( $m$ ) indices are indicated.

factors ( $\nu$ ), including spin, are indicated on the figure.<sup>14</sup> No spin splittings are resolved.

To lower energy below the principal  $(0,0)$  transitions, a series of satellite lines  $N_L$  ( $N_L = 0, 1, 2, \dots$ ) and  $T_n$  ( $n = 1, 2, \dots$ ), etc. is visible in Figs. 1(b) and 1(c) and in the fan chart of Fig. 2. The  $(N_e, 0)$  LL fan is replicated at 36 meV lower energy by the  $N_L$  satellites, in which a GaAs-like LO-phonon vibrational excitation of the QW material is emitted in the  $(N_e, 0)$  recombination. The allowed  $(N_e, N_h)$  recombination lines with  $N_e = N_h$  ( $N_e, N_h \geq 1$ ) show no evidence for LO-phonon satellite replication. For the present sample with  $E_F (25.4 \text{ meV}) < \hbar\omega_{\text{LO}} (36 \text{ meV})$  the parent LL's of the  $N_L$  satellites depopulate before the  $N_L$  satellites are resonant with  $(0,0)$ . As a result the resonant polaron anticrossing between the  $(0,0)$  and  $N_L$  final states, discussed in Ref. 7, is not seen.

The  $T_n$  satellites by contrast arise from the emission of Fermi-sea shake-up excitations. In the  $(N_e, 0)$  electron-hole recombination process an inter-LL magnetoplasmon excitation of the Fermi sea occurs in which an additional electron is promoted to a partly filled or empty LL. Since a Fermi-sea excitation is created in the shake-up, the  $T_n$  satellites occur to lower energy below  $(N_e, 0)$ . Neglecting many-body interactions, the  $T_n$  peaks are expected at energies  $\Delta N_e \hbar\omega_c^e$  below the  $(N_e, 0)$  recombination lines, where  $\Delta N_e$  is the increase in Landau quantum number of the electron which is promoted to higher energy.

In contrast to the previously reported observation<sup>4</sup> of shake-up processes in a sample with low  $n_s$  where only the  $N_e = 0$  LL was populated, a given  $T_n$  satellite in Figs. 1 and 2 can arise from a variety of inter-LL excitation processes. In Ref. 4, it was shown that the strength of the shake-up satellites falls off roughly linearly with the energy of the inter-LL excitation; a  $\Delta N_e = 1$  excitation is significantly stronger than a  $\Delta N_e = 2$  process. Thus the  $T_1$  peak occurring fairly close to  $\hbar\omega_c^e$  below  $(0,0)$  is very likely a  $\Delta N_e = 1$  shake-up satellite of  $(0,0)$  [rather than a  $\Delta N_e = 2$  satellite of  $(1,0)$ ]. Since the  $N_e = 1$  level is filled up to  $\nu = 4$  at 7.4 T, the shake-up can only occur by inter-LL excitation between  $N_e = 1$  and  $N_e = 2$  (between  $\nu = 6$  and 4), and between correspondingly higher LL's at lower magnetic field. The  $T_2$  satellite is probably a  $\Delta N_e = 2$  satellite of  $(0,0)$  with second electron promotion from  $N_e = 0-2, 1-3,$  or  $2-4$  depending on the LL filling factors.

Closer inspection of the energy separation of the  $T_1$  satellite from  $(0,0)$  in Fig. 2 shows that it occurs at an energy  $\sim 1.25$  times greater than the  $(0,0), (1,0)$  splitting of  $\hbar\omega_c$ . Such behavior is expected since in the correct many-body, magnetoplasmon description of inter-LL transitions the excitation energy is only equal to  $\hbar\omega_c$  for magnetoplasmon wave vector  $q = 0$ .<sup>15-17</sup> The maximum in the magnetoplasmon density of states occurs for  $q \sim 1/l_0$ , where  $l_0$  is the magnetic length  $(\hbar/eB)^{1/2}$ , at energies  $\sim 20\%$  greater than  $\hbar\omega_c$  (at 7 T in GaAs).<sup>18</sup> Coupling to states at  $q \sim 1/l_0$  ( $1.1 \times 10^6 \text{ cm}^{-1}$  at 7 T) is allowed by the weak disorder in the system,<sup>4,17</sup> and by the magnetoplasmon-LO-phonon coupling at the  $T_n - N_L$  satellite resonances discussed later. By contrast with the

(0,0)- $T_1$  splitting, the  $T_2$ - $T_1$  separation is much closer to the (1,0)-(0,0)( $\hbar\omega_c$ ) separation (see Fig. 2). This is expected from the theory of Ref. 15, where it is shown that the density-of-states maximum for  $\Delta N_e=2$  excitations is shifted above  $2\hbar\omega_c$  by roughly the same energy as  $\Delta N_e=1$  is shifted above  $\hbar\omega_c$ , with the result that the  $T_2$ - $T_1$  separation is expected to be close to  $\hbar\omega_c$ , as observed. The shoulder on the low-energy side of (0,0) labeled  $T_1^*$ , occurring  $\sim 1.30\hbar\omega_c$  below (1,0) [Figs. 1(c) and 2], is very likely a  $\Delta N_e=1$  satellite of (1,0), gaining intensity by resonant interaction with (0,0).

Expanded spectra in the satellite region at closely spaced magnetic fields are shown in Fig. 3, and in Fig. 4 the variation with field of the intensities of the  $N_L$  satellites, as a percentage of the intensity of the (0,0) zero-phonon line, is shown. Clear interaction between the  $N_L$  and  $T_n$  satellites is observed. In the absence of many-body effects with the inter-LL excitation energies equal to multiples of  $\hbar\omega_c$ , resonances between the  $N_L$  and  $T_n$  satellites will occur at magnetic fields given by the "magneto-phonon" resonance (MPR) condition  $m\hbar\omega_c = \hbar\omega_{LO}$ , where  $m (>0)$  is an integer.<sup>19</sup> Since the (0,0)- $T_1$  spacing is  $\approx 1.25\hbar\omega_c$ , the resonances are expected at

$$(m + 0.25)\hbar\omega_c = \hbar\omega_{LO} . \quad (1)$$

Magnetic fields corresponding to this modified MPR condition, for  $m=2-7$  are marked on Figs. 2 and 4.<sup>20</sup> At resonance the  $N_L$  and  $T_n$  excitations correspond to LO-

phonon-magnetoplasmon coupled modes.<sup>21,22</sup> Resonant anticrossing between  $T_1$  and  $1_L$  is seen very clearly in Fig. 2 at the onset of the  $m=2$  resonance (the dotted, boxed region), with splitting between the two modes at resonance of 8 meV. It is worth noting that a splitting at resonance for LO-phonon-magnetoplasmon coupled modes of  $\sim 6$  meV, reasonably close to our experimental findings, was calculated in Ref. 22 for an  $\text{In}_x\text{Ga}_{1-x}\text{As}$  heterojunction although at the lower  $n_s$  of  $4 \times 10^{11} \text{ cm}^{-2}$ .

The variation of intensities of the  $N_L$  satellites with  $B$  is plotted in Fig. 4 as a percentage of the (0,0) intensity. A series of oscillations of satellite intensity is seen, with maxima occurring to a good approximation at fields given by Eq. (1) ( $m=7$  to 3), corresponding to the crossing points of  $N_L$  and  $T_n$  in Fig. 2. Several points should be noted. At fields beyond which the parent LL of an  $N_L$  satellite begins to empty (e.g., fields greater than  $\nu=6$  for the  $N_e=2$  parent LL of the  $N_L$  satellite), the  $N_L$  satellite intensities are expected to be reduced by the  $N_e$  depopulation with  $B$ . In field ranges where such depopulation effects are strong, dashed rather than solid lines are drawn through the experimental points of Fig. 4. Furthermore, it is only for fields above 5 T for  $T_1$  and 7 T for  $T_2$  that the  $T_n$  lines are resolved as separate features from  $N_L$ ; at lower fields the intensities plotted, close to resonance, arise from unresolved  $T_n, N_L$  satellites.

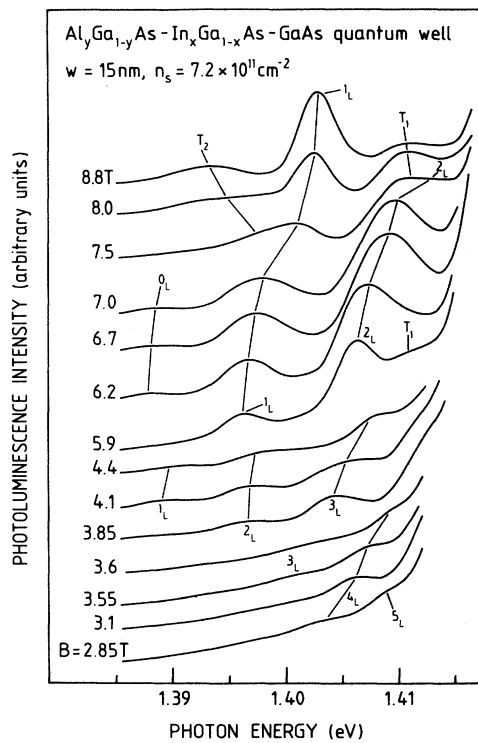


FIG. 3. Expanded spectra in the  $N_L, T_n$  satellite region between 2.85 and 8.8 T. The resonance enhancements of, for example,  $4_L$  at 3.1 T,  $3_L$  at 3.85 T,  $2_L$  at 6.7–7.5 T, and  $1_L$  from 7.5 to 8.8 T are clearly visible.

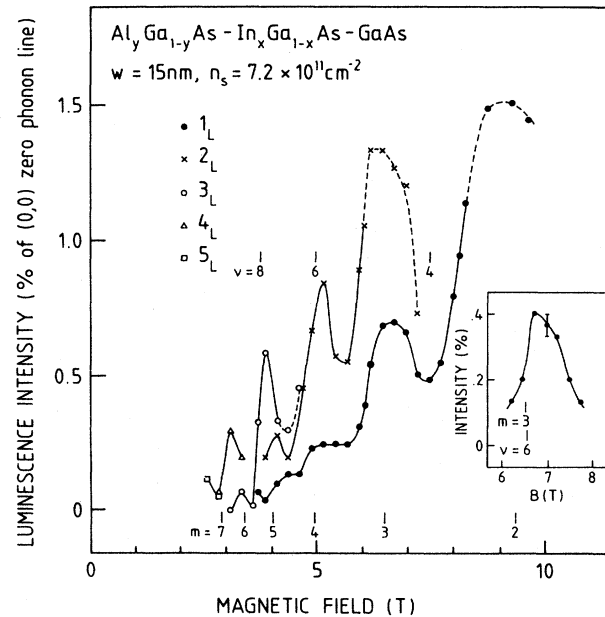


FIG. 4. Integrated intensities of the  $N_L$  satellite lines vs magnetic field for sample with  $n_s=7.2 \times 10^{11} \text{ cm}^{-2}$ , as a percentage of the intensity of the (0,0) zero-phonon line. The lines drawn through the experimental points are guides to the eye. The lines are drawn dashed in magnetic-field regions where the  $N_L$  intensity is strongly reduced by Landau-level depopulation (e.g.,  $2_L$  beyond  $\nu=6$ ). For  $B < 5$  T,  $T_n$  lines are not resolved from  $N_L$ . Good correlation of the intensity maxima with fields corresponding to  $N_L, T_n$  crossing (integer  $m$  values) is seen. The inset shows the variation of  $1_L$  intensity with  $B$  in the region of  $m=3$  for the sample with  $n_s=9.5 \times 10^{11} \text{ cm}^{-2}$ .

We suggest that the oscillations of satellite intensity arise from the mixing of the otherwise unperturbed LL's by the electron-LO-phonon interaction at the MPR condition. The mixing will give rise to increased inter-LL scattering at resonance.<sup>23</sup> This in turn will lead to enhanced shake-up satellite intensities, and hence magnetoplasmon-LO-phonon coupled-mode intensities, at fields close to integer  $m$  values, in agreement with the results of Fig. 4.

Variations in satellite intensity due to oscillations in screening with LL filling factor can be excluded as an explanation for the findings of Fig. 4. Very similar variations in  $N_L, T_n$  intensity with  $B$  have been found in a sample of the same design as that discussed in Figs. 1-4, but having 30% higher  $n_s$  ( $n_s = 9.5 \times 10^{11} \text{ cm}^{-2}$ ). The LL's will hence depopulate at  $\sim 30\%$  higher fields than those for the lower  $n_s$  sample. The variation of the  $1_L$  intensity with  $B$  for this sample is shown in the inset to Fig. 4. Once again a peak in intensity close to an integer  $m$  value is observed, supporting the attribution of the inten-

sity oscillations to  $T_n, N_L$  resonant interaction.

In conclusion, both LO-phonon and inter-Landau-level shake-up excitations have been reported in the magneto-PL spectra of QW's with high carrier density. Both  $\Delta N_e = 1$  and  $\Delta N_e = 2$  shake-up satellites were observed at energies close to the predictions of many-body theory for magnetoplasmon excitations in a high-density system. Clear evidence for LO-phonon-magnetoplasmon coupled mode behavior was observed. Very recently, Butov *et al.*<sup>24</sup> have reported experimental results which bear a number of similarities to those in the present paper. Most notably a clear observation of magnetoplasmon-LO-phonon coupled mode satellites in magnetoluminescence spectra is reported by these workers.

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<sup>3</sup>LO-phonon replication in bulk semiconductor spectra is a very-well-documented subject, see, e.g., J. J. Hopfield, *J. Phys. Chem. Solids* **10**, 110 (1959).

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<sup>5</sup>P. H. Citrin, G. K. Wertheim, and Y. C. Baer, *Phys. Rev. B* **16**, 4256 (1977).

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<sup>9</sup>M. S. Skolnick, J. M. Rorison, K. J. Nash, and S. J. Bass, *Surf. Sci.* **196**, 507 (1988).

<sup>10</sup>S. K. Lyo and E. D. Jones, *Phys. Rev. B* **38**, 4113 (1988).

<sup>11</sup>The hole temperature of 20-25 K is estimated from the relative intensities of the (1,1) to (0,0) magneto-PL transitions.

<sup>12</sup>M. S. Skolnick, K. J. Nash, S. J. Bass, P. E. Simmonds, and M. J. Kane, *Solid State Commun.* **67**, 637 (1988).

<sup>13</sup>S. K. Lyo, E. D. Jones, and J. F. Klem, *Phys. Rev. Lett.* **61**, 2265 (1988).

<sup>14</sup>The filling factors and hence the  $n_s$  value in the QW are determined from the magnetic fields at which the  $(N_e, 0)$  LL transi-

tions ( $N_e = 3, 2$ ) are no longer visible in the PL spectra.

<sup>15</sup>C. Kallin and B. I. Halperin, *Phys. Rev. B* **30**, 5655 (1984).

<sup>16</sup>A. H. Macdonald, *J. Phys. C* **18**, 1003 (1985).

<sup>17</sup>A. Pinczuk, J. P. Valladares, D. Heiman, A. C. Gossard, J. H. English, C. W. Tu, L. Pfeiffer, and K. West, *Phys. Rev. Lett.* **61**, 2701 (1988).

<sup>18</sup>The deviation of the maximum in the magnetoplasmon density of states from  $\hbar\omega_c$  should be field dependent, as predicted in Refs. 15 and 16 and observed in Ref. 4. Due to the weakness of the  $T_1$  satellites and the scatter of the experimental points, detailed analysis of the (0,0)- $T_1$  separation with field is difficult. For the same reason the observed (0,0)- $T_1$  separation of  $1.25\hbar\omega_c$  should be regarded as being in very reasonable agreement with the predicted value of  $1.20\hbar\omega_c$  at 7 T.

<sup>19</sup>P. G. Harper, J. W. Hodby, and R. A. Stradling, *Rep. Prog. Phys.* **36**, 1 (1973).

<sup>20</sup>As noted in Ref. 18, the deviation of the  $T_1$ -(0,0) splitting from  $\hbar\omega_c$  should be field dependent, varying as  $B^{1/2}$  as discussed in Ref. 14. Nevertheless within the uncertainty of the data, Eq. (1) will be a reasonable approximation to the experimental  $m$  values.

<sup>21</sup>H. C. Oji and A. H. Macdonald, *Phys. Rev. B* **34**, 1371 (1986).

<sup>22</sup>X. Wu, *Phys. Rev. B* **38**, 4212 (1988).

<sup>23</sup>In Ref. 4 and in the work of T. Uenoyama and L. J. Sham, *Phys. Rev. Lett.* **65**, 1048 (1990) it was shown that shake-up is strong in systems containing localized holes, the localized holes giving rise to enhanced electron inter-LL scattering.

<sup>24</sup>L. V. Butov, V. I. Grinev, V. D. Kulakovskii, and T. G. Andersson, *Phys. Rev. B* **46**, 13 627 (1992).