Magneto-optical spectroscopy of free- and bound-electron-hole excitations in the presence of a two-dimensional electron gas

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Experimental and theoretical studies of the energy diagram of magneto-optical transitions observed in both luminescence and luminescence excitation spectra of an *n*-type modulation-doped quantum well are presented. The transitions related to the lowest populated electronic subband are well understood in terms of interband transitions with many-body corrections included within the density-functional approach. The excited subbands are differently renormalized and show a two-particle excitonic correlation.

I. INTRODUCTION

Interband magneto-optical experiments on modulation-doped quantum-well structures (MDQW's) have shown a number of effects related to the specific behavior of a two-dimensional (2D) electron gas subjected to perpendicular magnetic fields.¹ Most of the studies have been limited to the analysis of luminescence spectra only,¹⁻³ and there are very few reports on absorption-type experiments.⁴⁻⁶ Much effort has been focused on the properties beyond the oneelectron approximation, $1^{-3,5,6}$ whereas problems related to the complexity of the valence-band structure^{7,8} have been tentatively avoided. In fact, there is a lack of systematic studies of both luminescence and absorption which might demonstrate to what extent one could reproduce the rich energy spectrum of interband magnetooptical transitions in MDQW's on the basis of generally accepted theoretical concepts concerning many-body effects including the complexity of the valence band structure. To answer this question, we have studied a wellcharacterized $GaAs/Ga_xAl_{1-x}As$ MDQW in a series of magneto-optical experiments in which we have probed both occupied and empty electronic states using combination of luminescence and luminescence excitation spectroscopy. The experimental results, i.e., the energy ladder of the observed transitions, are compared with the calculated energy structure, taking into account the effect of band gap renormalization, the mixing of valence band Landau levels, and the formation of excitonic resonances for transitions involving empty electronic subbands.

After describing the experiments, we present our data

in Sec. III. Optical spectra measured for different magnetic fields allow us to clearly distinguish between transitions involving Landau levels of the lowest occupied electric subband and transitions related to the states of higher empty electric subbands. Transitions of the former type show a convincingly linear field dependence and, as will be shown in Sec. IV, they can be well described as interband transitions between one particle levels of the conduction and valence bands, although these levels are calculated in the potential shape locally modified by the renormalized band gap. Transitions involving energy levels of the empty electric subbands behave differently, showing, for example, diamagnetic shifts characteristic of excitonic resonances. The excitons, discussed in Sec. V, can be efficiently formed out of the empty states around k = 0 of the higher electric subbands although they are screened by electrons occupying the lowest electronic subband. Binding energies of these excitons are experimentally derived and compared with recent theoretical calculations.⁹

It is shown that the energies of transitions related to the lowest occupied electric subband are well reproduced in calculations, when the effect of the band-gap renormalization is locally included in the self-consistent solution of Poisson and Schrödinger equations (functional density approach). In contrast, for higher electric subbands better agreement is obtained when many-body effects are neglected.

II. EXPERIMENTAL DETAILS

The sample investigated, having well-defined optical spectra at zero magnetic field, was chosen from several

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structures grown by molecular beam epitaxy. This was a single quantum-well modulation doped on one side. The structure was grown on a semi-insulating GaAs substrate in the following sequence: GaAs buffer layer, undoped barrier of $Ga_{1-x}Al_xAs$ with composition of x = 0.34 and thickness d = 2000 Å, GaAs quantum well of d = 250 Å, $Ga_{1-x}Al_xAs$ spacer of $d_1 = 150$ Å and x = 0.43, layer of $Ga_{1-x}Al_xAs$ doped with Si donors with doping level of $N_d = 10^{18}$ cm⁻³, and d = 500 Å capped by 50 Åthick GaAs layer. The sample was initially characterized with low temperature magnetotransport and cyclotronresonance measurements. When cooled in darkness it shows well-defined quantum Hall plateaus. Under these conditions the free electron concentration and the low field Hall mobility were found to be 5.0×10^{11} cm⁻² and 3.5×10^5 cm²/V s, respectively. As usually and also in the case of our sample, the laser illumination destroys the observation of well-defined quantum Hall plateaus. This is due to the appearance of a parallel conduction channel in the $Ga_{1-x}Al_xAs$:Si layer. After illumination, the electron sheet concentration $n_s = 7.6 \times 10^{11} \text{ cm}^{-2}$ obtained from measurements of Shubnikov-de Haas oscillations has been found to be indepedent of further changes in the illuminations intensity in the range considered. The effective electron mass $m^* = 0.072m_0$ was measured by far-infrared Landau-emission experiments. This value is 7% higher than the band edge mass of GaAs, and agrees well with calculations which include the nonparabolicity of the Γ_6 conduction band. Luminescence spectra have been measured at liquid helium temperature (4.2)K) in magnetic fields up to 20 T supplied by Bitter coils. The sample was excited in Faraday configuration with the 514.5 nm line of the Ar⁺ laser or with the Ti:Sa tunable laser pumped by the Ar^+ laser. Experiments were performed for different σ^+ and σ^- helicities of both the excited and the emitted light. In luminescence spectra, signals related to both the quantum well and the bulk GaAs are observed. Bulk emission was found to be relatively weak under high energy Ar⁺ excitation indicating efficient trapping of photoexcited carriers into the quantum well. The 2D-related signals have been selected from the spectra using the conventional technique of tilted field experiments. The peak assignment has been additionally verified by luminescence experiments under resonant excitation below the barrier band edge.

III. EXPERIMENTAL RESULTS

The luminescence and luminescence excitation (pseudoabsorption) spectra observed without magnetic field are presented in Fig. 1. The observed structures can be interpreted as follows. The broadband observed in luminescence can be assigned to the recombination E_0H_0 between the lowest occupied electron, E_0 , and the highest heavy hole, H_0 , electric subbands. The sharp, excitonlike line, observed both in the luminescence and luminescence excitation is related to the E_1H_0 excitation between the first empty electric subband E_1 and the H_0 hole subband. The higher energy peaks observed in



FIG. 1. Photoluminescence and photoluminescenceexcitation (pseudoabsorption) spectra at zero magnetic field. The observed structures are assigned to E_iH_j (E_iL_j) transitions between subsequent conduction band E_i and valence band heavy hole H_j (light hole L_j) subbands. The quantum-well energy structure is presented schematically in the inset.

the photoexcitation spectrum are assigned to the E_1L_0 , E_1H_2 , E_2H_1 , and E_3H_2 transitions between subsequent conduction (E_i) and valence (L_i, H_i) subbands, where L and H denote, respectively, the light and heavy hole states. Note that contrary to undoped structures in our nonsymmetric quantum well most of the dominant transitions involve electric subbands with different quantum numbers. This is a consequence of the symmetry of wave functions in the growth direction which, in fact, are better overlapped in the case of electric subbands with different quantum numbers than in case of subbands with the same quantum number.

When a sufficiently high magnetic field is applied (see Fig. 2), we are able to resolve peaks in previously featureless parts of the zero-field luminescence and luminescence-excitation spectra. Some of the structures observed in luminescence correspond to band-edge recombination in the GaAs substrate. These emission lines can be easily recognized since they dominate the luminescence spectrum when the selective excitation energy is chosen to be in the range where the quantum-well absorption vanishes.⁶

The measured spectrum is very rich and the observed transitions depict strong circular polarization. The Landau level fan chart of the dominant transitions observed in luminescence and luminescence-excitation spectra for the σ^+ and σ^- circular polarizations is shown in Fig. 3. The solid and broken lines are the results of calculations presented in Secs. IV and V, can be use for the moment as guides for the eye.

The optical transitions observed in the magnetic field can be divided into groups related to different electric subbands. For the lowest, occupied E_0 subband one can observe a nearly linear increase of the transition energies with the magnetic field. In the first approximation, it can be described within a simple Landau level geometry with the splitting for the electron and hole



FIG. 2. Photoluminescence and photoluminescenceexcitation spectra for σ^+ (a) and σ^- (b) circular polarizations at B = 9.5 T. Each set of arrows indicate transitions corresponding to the same conduction band subband E_i .

Landau levels defined by a single reduced effective mass $m_r = 0.65 m_0.^{10}$ Transitions related to fully occupied electronic Landau levels are visible only in the emission experiments. Upon increasing the magnetic field, when a given Landau level depopulates, the related optical transition becomes observable in the absorption-type spectra. When a given transition is visible in both the luminescence and luminescence-excitation spectra we do not observe any difference between the emission and absorption energies, within an experimental error estimated to be about 0.5 meV.

For the transitions related to higher, empty electronic subbands a quite different nature is noticeable. The energies of the ground state transitions associated with each of the excited subbands show significantly weaker field dependence than the ground state of the lowest occupied subband. In fact the former transitions show nonlinear diamagnetic field dependence which indicates their excitonic character.

IV. INTERBAND TRANSITION ASSOCIATED WITH THE OCCUPIED ELECTRIC SUBBAND

Photoluminescence and photoluminescence-excitation spectra of the transitions related to the lowest electric subband show some characteristic features which suggest the choice of the theoretical approach to describe these spectra. The magnetic field dependences of the energies of the discussed transitions follow quite linear field dependences. This is particularly significant in the low field region and indicates exciton unbinding for these



FIG. 3. The Landau level fanchart of σ^+ (a) and σ^- (b) polarized transitions observed in luminescence (open circles) and luminescence excitation (closed circles) spectra.



FIG. 4. Part of luminescence-excitation spectrum where in the range between 1.55 eV and 1.57 eV the spectrum shows a number of transitions involving the same n = 1 electronic Landau level and different valence band levels.

transitions in agreement with theoretical predictions. 11,12 At high magnetic field, when the energy levels become well resolved and when all but the lowest electron Landau level is empty, very rich spectra of the zero-subband transitions are observed in absorption-type experiments (see Fig. 4). The rich absorption spectra in high fields is the manifestation of the complexity of the valence band structure. In consequence, one can expect that the magnetic field fan chart of the zero-subband-related transitions may be understood within the one-electron approximation, i.e., in terms of interband magneto-optical transitions, although all the valence band structure complexity has to be taken into account. However, as discussed in Sec. VI, such a simplified description is not realistic since it does not allow us to reproduce properly the absolute values of the observed transition energies. This discrepancy is thought to be of a many-body origin. In the first approximation it can be corrected using the concept of the band-edge renormalization. The following procedure has been applied to reproduce the Landau level fanchart of transitions related to the lowest electron subband. We have used the envelope function approximation, including, however, the local exchangecorrelation terms both for electrons and holes within the functional density approach (FDA).^{13,14} Using this approximation the Poisson and Schrödinger equations have been solved self-consistently for the case when the magnetic field B = 0 T. In the Poisson equation the charges of free electrons confined in the quantum well and ionized donors in the doped region of the barrier have been taken into account. The Fermi energy E_F has been assumed to be pinned to the neutral donor states in the doped region in the $Ga_{1-x}Al_xAs$ barrier, and constant for all the structure. Its value, as measured from the bottom of the conduction band in the doped region, has been adjusted to reproduce the confined carrier concentration derived from magnetotransport experiments.

The electronic Landau levels energies have been calculated including the effect of nonparabolicity as described in Ref. 15. Energies of valence levels have been obtained within the envelope approximation^{4,7} through diagonalization of the Luttinger Hamiltonian for the Γ_8 valence band, for $B \parallel [100]$. For B = 0 T, the Schrödinger equation has been solved numerically for $k_x = k_y = 0$. The effective potential for the valence band has been obtained within FDA including the electrostatic term derived from the free electron charge density and the exchange-correlation term.¹⁴ The calculated matrix elements of the k_z operator have been used to obtain the Landau level energies after diagonalization of the Luttinger Hamiltonian. The number of electric hole subbands used in the calculations has been limited to seven.

The solid lines presented in Fig. 3 reproduce the energies of the dominant optical transitions calculated using the procedure described above. The agreement with experimental results is very good for such a simplified description, both with respect to absolute values and the magnetic field dependence of transitions related to the lowest subband. It is worth emphasizing the rich structure (see Fig. 4) observed in the photoexcitation spectra for magnetic fields above 10 T in the energy range between 1.54 and 1.57 eV. On the basis of the performed calculations it can be clearly interpreted as transitions from the Landau levels related to deeper hole electric subbands to the spin split n = 1 electron Landau level of the lowest electric subband.

V. EXCITONIC RESONANCES RELATED TO EXCITED SUBBANDS

The transitions which involve the empty electric subband E_1 are clearly different reflecting an excitoniclike behavior and therefore a similar analysis to that performed for the E_0 -related transitions is not appropriate.

For example, the excitonic character of the E_1 -related



FIG. 5. Photoluminescence-excitation spectra of $GaAs/Ga_{1-x}Al_xAs$ modulation-doped quantum well at low magnetic fields showing the evolution of magnetoexcitonic resonances related to the first excited electronic subband.

transitions can be clearly seen in the low field absorptionlike spectra presented in Fig. 5. Although the zero-field spectrum in this figure shows only the ground states of heavy and light hole resonances, the clear evolution of a series of the excited heavy hole excitons is observed when relatively weak magnetic fields are applied.¹⁶ The excited states become more pronounced in the presence of the magnetic field as a result of the effective shrinkage of their wave functions.¹⁷ At higher magnetic fields all the observed lines are split and spin components are observed in one of the σ^+ or σ^- circular polarization (Fig. 3) in the same sequence as for excitons of an undoped quantum well.¹⁸

To characterize quantitatively the observed heavy hole exciton E_1H_0 structure, the magnetic field dependence of the energy of the states has been calculated (see broken lines in Fig. 3) applying the two-dimensional approximation (i.e., two-dimensional hydrogen atom placed in a magnetic field,^{16,19} including nonparabolicity in the first approximation). The binding energy E_{B1} of the 1s heavy hole E_1H_0 ground exciton state and the reduced exciton effective mass $m*_{r1}$ are used as fit parameters and have been found to be $E_{B1} = 2.0 \text{ meV}$ and $m*_{r1} = 0.051m_0$, respectively. The third fit parameter, i.e., the separation energy between free electron and hole electric subbands (energy gap for this transition) has been found to be equal to 1534.2 meV.

The analysis of higher subband transitions is less accurate. The broadening of the observed structures is significant and increases the error of the estimated parameters. For the E_2H_1 transitions, the excitonic interaction is found to be still noticeable. The two-dimensional exciton approximation has been used to describe the magnetic field dependence of these transitions (Fig. 3—broken lines). Within this approximation an exciton binding energy $E_{B2} = 4.0$ meV and a reduced mass $m_{r2}^* = 0.051m_0$ have been estimated. For the E_3H_2 transitions, within the experimental error, the excitonic correction is not necessary and the magnetic field dependence of these transitions can be reproduced using the reduced effective mass $m_{r3}^* = 0.065m_0$.

VI. DISCUSSION

Our results clearly demonstrate that the main features of magneto-optical experiments on a conventional modulation-doped quantum-well structure can be well understood using quite simple theoretical models. The large variety of magneto-optical transitions, their hierarchy and polarization rules, originates from the complexity of the valence band structure and is satisfactorily explained using the envelope function approximation. The effects beyond the one electron approximation which have to be included are the band-gap renormalization and the formation of excitonic resonances at the edges of nonpopulated electric subbands.

The transition energies related to the lowest occupied electric subband are well reproduced in calculations of the interband Landau level transitions. These energies are calculated with a potential shape which is locally shifted according to the effect of the band-gap renormalization. It is found that the transitions related to the Landau levels of the lowest electric subband are renormalized independently of whether these levels are populated or partially emptied upon increasing the magnetic field. The theoretically predicted changes of the transition character when emptying the electronic levels²⁰ are not observed here. However, the electron concentration in our experiments is higher than the one considered in these calculations. On the other hand, our experiments show that transitions related to levels of higher empty electric subbands do not follow the same renormalization effect as the ones involving the lowest subband states. In fact the higher subband transitions are better reproduced in calculation when the effect of band-gap renormalization is neglected. These transitions show, however, a two-particle correlation excitonic effect.

For a more quantitative estimation of the applicability of the FDA to the higher, empty subbands, we compare (Table I) the calculated energy gaps between different electron and hole electric subbands, with the experimental values. Energies of the dominant transitions, measured for B = 0 T, have been shifted up by the exciton

| | Calculated energy gaps (meV) | | | | Experimental |
|---------------------|------------------------------|-----------|----------------|-----------|--------------------|
| | electrostatics | | plus many body | | energy |
| | d=250 Å | d = 240 Å | d=250 Å | d = 225 Å | gaps (meV) |
| $\overline{E_0H_0}$ | 1513.5 | 1514.8 | 1501.9 | 1503.2 | 1503.7 ± 1 |
| $\overline{E_1H_0}$ | 1538.8 | 1540.9 | 1529.2 | 1533.6 | $1534.2{\pm}.5$ |
| $\overline{E_1L_0}$ | 1544.8 | 1547.2 | 1534.4 | 1540.0 | 1539.5±1* |
| E_2H_1 | 1580.1 | 1585.3 | 1569.4 | 1582.4 | 1583.0 ± 2 |
| $\overline{E_3H_2}$ | 1635.7 | 1644.5 | 1624.4 | 1647.4 | 1643.0 ± 3 |
| $\overline{E_1H_2}$ | 1558.8 | 1562.1 | 1547.4 | 1555.3 | 1555.5 ± 1^{b} |
| $\overline{E_2H_0}$ | 1571.2 | 1575.1 | 1561.6 | 1572.8 | 1569.0 ± 2^{b} |

TABLE I. The comparison of the calculated energy separation between different conduction band and valence band electric subbands with the experimentally observed transition energies corrected where nesessary for the excitonic binding energies (see text).

^aFor the E_1L_0 exciton the same binding energy $E_B = 2.0$ meV is assumed as for the E_1H_0 exciton. ^bTransition energies observed at B = 0 T without excitonic corrections. binding energy, as discussed in Sec. V. For comparison, the calculated energy gaps, when many-body effects are totally neglected (electrostatic potential only) are also presented. One can see that it is impossible to reproduce the fundamental energy gap E_0H_0 when neglecting many-body effects, whereas, as pointed out above, within the FDA the agreement is very good. For the higher electric subbands, one can reach quite good agreement, neglecting many-body corrections, and assuming a slightly smaller quantum-well width than the value specified from the parameters of the growth process. To obtain a similar agreement within the FDA, the size of the quantum well should be reduced to unrealistic dimensions which are totally incompatible with the growth parameters. We therefore conclude that the rigid band-gap shift (which depends on the local electron density within the FDA) overestimates the role of many-body interactions for empty states. Our observation is, however, in agreement with the generally accepted picture²¹ that the single particle spectrum of the free-electron gas is strongly modified for the occupied states, while the modification of the higher energy states is substantially weaker.

The most prominent feature of transitions related to empty electronic subbands is their excitonic character despite the high concentration of electrons occupying the lowest electric subband. This observation confirms once more the fact that in two-dimensional structures screening plays a secondary role in the bleaching of the exciton resonance which in fact can be formed as long as there exist empty electronic states around k = 0. In our experiments clear structures corresponding to E_1H_0 heavy and E_1L_0 light excitons are observed (Fig. 5) with a binding energy of the heavy hole exciton equal to 2 meV. The same value has been found in calculations performed for the MDQW with the same parameters.⁹

We have estimated the binding energy of the next excitonic transition E_2H_1 to be higher (~ 4 meV) which might be due to the different symmetry of the related wave functions and it would be interesting to confirm this experimental result with calculations.

In addition we would like to mention that contrary to some previous reports on $Ga_{1-x}In_xAs/Ga_{1-x}Al_xAs$ structures²² we do not observe any evidence of the electron effective mass renormalization due to electronelectron interaction. Both, inter-Landau-level (cyclotron emission) and interband spectra are well described using the GaAs effective mass value $m^* = 0.66m_0$, corrected for the nonparabolicity of the Γ_6 band.

Several effects related to many-body interactions such as the Fermi edge singularity or shake-up $processes^{2,3,23}$

are not discussed in this paper. Experimentally, these effects are found not to affect the energy diagram of the optical transitions in the investigated structure. On the other hand, they are important for the intensity of the particular transitions. The intensity analysis is, however, beyond the scope of this paper and will be published elsewhere.

VII. CONCLUSIONS

We have presented an analysis of the energy diagram of magneto-optical transitions observed in both luminescence and luminescence-excitation spectra of an n-type modulation-doped quantum-well structure with electrons occupying the lowest conduction band electric subband. The aim of the analysis was to determine the possible simplest theoretical model which reproduces satisfactorily our experimental data. We have found that different approaches are necessary depending on whether the optical transitions involve Landau levels of populated electron subband or whether they are related to empty conduction band subbands. The transitions involving the lowest (occupied) electron subband can be understood in terms of interband transitions where many-body corrections are included within the functional density formalism. For the electron concentration considered, this approach satisfactory describes all the Landau level transitions independent of whether they are occupied or empty but only if they belong to the lowest conduction band subband. The same approach cannot be used to reproduce the energies of the transitions related to levels of higher (empty) conduction band subbands. These transitions are differently renormalized and they show excitonic two-particle correlation.

We believe our data could stimulate a theoretical approach to search for a self-consistent model describing the rich spectra of magneto-optical transitions which are observed both in luminescence as well as in absorption-type experiments on modulation-doped structures.

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