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Citation: *Journal of Applied Physics* **92**, 1227 (2002); doi: 10.1063/1.1489495

View online: <http://dx.doi.org/10.1063/1.1489495>

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Magnetoabsorption spectra of intraexcitonic transitions in GaAs-(Ga,Al)As semiconductor quantum wells

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(Received 11 March 2001; accepted for publication 2 May 2002)

We present a theoretical study, within the effective-mass approximation, of the magnetoabsorption spectra of intraexcitonic terahertz transitions of light-hole and heavy-hole confined magnetoexcitons in GaAs-(Ga,Al)As quantum wells. The semiconductor quantum wells are studied under magnetic fields applied in the growth direction of the semiconductor heterostructure. The various magnetoexciton states are obtained in the effective-mass approximation by an expansion of the exciton-envelope wave functions in terms of products of hole and electron quantum-well states with appropriate Gaussian functions for the various excitonic states. Intramagnetoexciton transitions are theoretically studied by exciting the allowed excitonic transitions with σ^+ (or σ^-) far-infrared radiation circularly polarized in the plane of the GaAs-(Ga,Al)As quantum well. Theoretical results are obtained for the intramagnetoexciton transition energies and magneto-absorption spectra associated with excitations from $1s$ -like to $2p_{\pm}$, and $3p_{\pm}$ -like magnetoexciton states, and found in overall agreement with optically detected resonance measurements. © 2002 American Institute of Physics. [DOI: 10.1063/1.1489495]

I. INTRODUCTION

Optical measurements of semiconductor heterostructures are of great value in understanding the physical nature of confined electrons, holes, and Coulomb-bound states such as impurities and excitons. In particular, a quantitative description and full comprehension of the role played by impurities and excitons in low-dimensional semiconductor heterostructures are not only of basic scientific relevance but also of considerable importance due to the prospects for building new optoelectronic physical devices. Excitonic features essentially dominate the optical properties of semiconductor heterostructures, such as GaAs-(Ga,Al)As quantum wells (QWs) and multiple quantum wells (MQWs), and effects due to magnetic fields applied along the growth axis of GaAs and Ga_{1-x}Al_xAs semiconductor layers provide valuable information on carrier subbands and exciton states via magneto-optical studies. Magnetoexcitons are observed as a series of hydrogenic-like ground and excited states in GaAs-Ga_{1-x}Al_xAs QWs and MQWs under magnetic fields in the perpendicular direction to semiconductor layers. The allowed transition energies among the various magnetoexciton states are found in the far-infrared (FIR) or terahertz region (i.e., energies of the order of 10 meV or 2.4 THz).

Recently, several internal excitonic transitions were observed by Salib *et al.*,¹ who found dominant $1s \rightarrow 2p_+$

heavy-hole (hh) magnetoexciton transitions in $L = 80 \text{ \AA}$ and $L = 125 \text{ \AA}$ (well width) GaAs-Ga_{0.7}Al_{0.3}As MQWs under growth-direction applied magnetic fields. Their magnetoexciton experimental spectra also revealed “weak” and “very weak” features which they tentatively assigned to hh $1s \rightarrow 3p_+$ and $1s \rightarrow 4p_+$ transitions, respectively. Also, the optically detected resonance (ODR) spectra by Salib *et al.*¹ presented other features which were attributed either to hole cyclotron resonances (CRs) or termed as of uncertain origin. Independently, Cerne *et al.*^{2,3} monitored changes in the excitonic photoluminescence spectra which were induced by FIR radiation with the electric-field polarized in the plane of the QW, and observed FIR $1s \rightarrow 2p_+$ hh magnetoexciton absorption in GaAs-Ga_{1-x}Al_xAs QWs under magnetic fields applied along the growth direction. More recently, ODR spectroscopic measurements were used by Nickel *et al.*⁴⁻⁶ in the study of electron and hole CRs and various internal magnetoexcitonic transitions in a number of GaAs-(Ga,Al)As MQW structures. Their results indicated (see final paragraph of Ref. 4) the need of further work to confirm the $2p_{\pm}$ assignments, to resolve the nature of the higher-energy intraexcitonic transitions, and to observe light-hole (lh) CR and associated intraexcitonic transitions.

From the theoretical point of view, a variational procedure within the effective-mass approximation, with hydrogeniclike envelope wave functions for the exciton states, was performed by Duque *et al.*⁷ in an investigation of lh and hh magnetoexcitonic transition energies in GaAs-Ga_{1-x}Al_xAs

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QWs. A further theoretical study, within the effective-mass approximation, considered an expansion of the magnetoexciton-envelope wave functions in terms of products of hole and electron QW states with appropriate Gaussian functions^{8–10} for the various excitonic states.¹¹ Both theoretical approaches result in very good agreement between the two calculations for the hh $1s \rightarrow 2p_{\pm}$ magnetoexciton transitions, whereas for higher-energy magnetoexciton transitions, the two calculations give quantitatively different results for the transition energies.¹¹ A comparison with experimental data indicated that, although some of the theoretical magnetoexciton transition energies agree quite well with experimental measurements, other calculated transitions only reproduce qualitative features of the experiment. Also, both theoretical calculations indicated that some of the calculated transitions should be (and were not) observable in the experimental spectra. In the hope of achieving a better understanding of the terahertz transitions of confined magnetoexcitons in GaAs–Ga_{1-x}Al_xAs QWs and of the ODR experimental data by Salib *et al.*,¹ Cerne *et al.*,^{2,3} and Nickel *et al.*,^{4–6} in this work, we present a detailed theoretical study on the magnetoabsorption spectra of intraexcitonic transitions in GaAs–Ga_{1-x}Al_xAs QWs under magnetic fields applied along the QW growth direction.

The work is organized as follows. Section II presents the magnetoabsorption coefficient associated with intraexcitonic transitions excited by circularly polarized light (σ^+ or σ^-) in the plane of the GaAs–Ga_{1-x}Al_xAs QW. Theoretical results, comparison with experiment, and discussion are shown in Sec. III for the intraexcitonic $1s \rightarrow np_{\pm}$ transitions. Finally, in Sec. IV, we present our conclusions.

II. THE MAGNETO-ABSORPTION COEFFICIENT FOR INTRAEXCITONIC TRANSITIONS

Here, we work in the effective-mass approximation, and consider exciton states in GaAs–Ga_{1-x}Al_xAs QWs of width L in the presence of a magnetic field parallel to the growth direction of the heterostructure. The values of the square potential-well barriers $V_c(z_e)$ and $V_v(z_h)$ are assumed to be 65% and 35% of the total energy-band-gap discontinuity, respectively, and therefore dependent on the Al concentration. Also, we consider the spin–orbit splitting to be large enough so that the interaction between $J=3/2$ and $J=1/2$ states may be disregarded, and, for simplicity, we take the relative motion of the carriers and that of the center of mass as independent, although one may only make this separation in the plane of the well.^{12,13} Moreover, we assume independent excitons by discarding the off-diagonal elements in the hole Hamiltonian,^{14,15} i.e., effects due to hole-subband mixing are neglected in the calculation. Also, image-charge effects are not considered and the electron–hole (e–h) Coulomb interaction is assumed to be screened by an average static dielectric constant of the GaAs and Ga_{1-x}Al_xAs bulk materials. In particular, we are interested in the $\alpha(\omega)$ magnetoabsorption coefficient for intraexcitonic transitions. In the dipole approximation, $\alpha(\omega)$ associated with transitions from initial state i to final state f is essentially given by

$$\alpha_{i \rightarrow f}(\omega) \propto \frac{1}{\omega} \left| \langle F_{M,M',m}^f(\boldsymbol{\rho}, z_e, z_h) \right| \times \hat{\boldsymbol{\epsilon}} \cdot \mathbf{P}_r \left| F_{M,M',m}^i(\boldsymbol{\rho}, z_e, z_h) \right|^2 \times \delta(E_{M,M',m}^f - E_{M,M',m}^i - \hbar\omega), \quad (1)$$

where $\hat{\boldsymbol{\epsilon}}$ corresponds to the photon polarization, M and M' are magnetic quantum numbers for the conduction and valence band, respectively, \mathbf{P}_r is the relative mechanical momentum of the e–h pair, and $F_{M,M',m}^p(\mathbf{r}_e, \mathbf{r}_h) \equiv F_{M,M',m}^p(\boldsymbol{\rho}, z_e, z_h)$, where $\boldsymbol{\rho}$ is the e–h relative coordinate in the plane of the QW, is the exciton-envelope wave function associated to the p th exciton state, with angular momentum m in the z direction. In Eq. (1), the exciton-envelope wave functions $F_{M,M',m}^p(\boldsymbol{\rho}, z_e, z_h)$ may be written in terms of single-particle $f_k^M(z_e)$ and $f_k^{M'}(z_h)$ solutions of the effective-mass equation for electron or hole motion, respectively, along the z axis of the QW, i.e.,^{8–10}

$$F_{M,M',m}^p(\boldsymbol{\rho}, z_e, z_h) = \sum_{k,k'} \psi_{k,k'}^{M,M',m,p}(\boldsymbol{\rho}, \phi) f_k^M(z_e) f_{k'}^{M'}(z_h), \quad (2)$$

with

$$\psi_{k,k'}^{M,M',m,p}(\boldsymbol{\rho}, \phi) = \sum_j C_{k,k',j}^{M,M',m,p} v_{m,j}(\boldsymbol{\rho}, \phi), \quad (3)$$

and

$$v_{m,j}(\boldsymbol{\rho}, \phi) = \rho^{|m|} e^{im\phi} e^{-\rho^2/\lambda_j^2}, \quad (4)$$

with the expansion in Eq. (3) made in a restricted set of Gaussian functions with appropriate λ_j length parameters.^{8–10} Note that the exciton has a total z -direction angular momentum \mathcal{M} (which is a good quantum number),¹⁵ which is given by the sum of the z -direction angular momenta of the envelope function m and those of the Bloch functions of electron M and hole $-M'$, i.e., $\mathcal{M} = m + M - M'$. In the calculations, we assume the hh and lh exciton Hamiltonians,^{14,15} and the GaAs conduction-band effective-mass and dielectric constant as $m_e = 0.0665$ (in units of the free electron mass m_0) and $\epsilon = 12.5$, respectively; the relevant mass parameters and the Luttinger valence-band parameters are taken as in Bauer and Ando.¹⁵ We label the magnetoexciton energy states as $n\ell m (M, M')$, which correspond to $n\ell m$ -like exciton states composed of a M electron (with $M = \pm 1/2$) and a $-M'$ hole (with $M' = \pm 1/2, \pm 3/2$). By writing the relative mechanical momentum as $\mathbf{P}_r = \mathbf{p}_r + e/c\mathbf{A}_r$, where e is the proton charge, \mathbf{A} is the vector potential associated with the $\mathbf{B} = B\hat{z}$ magnetic field (\hat{z} is the QW growth direction), and choosing the symmetric gauge, one finds

$$\hat{\boldsymbol{\epsilon}} \cdot \mathbf{P}_r = \frac{\hbar}{\sqrt{2}i} e^{\pm i\phi} \left(\frac{\partial}{\partial \rho} \pm \frac{i}{\rho} \frac{\partial}{\partial \phi} \mp \frac{Be}{2\hbar c} \rho \right), \quad (5)$$

for the photon circularly polarized in the QW plane, i.e., $\hat{\boldsymbol{\epsilon}} = 1/\sqrt{2}\hat{x} \pm 1/\sqrt{2}\hat{y}$, corresponding to polarization σ^+ (or σ^-). By defining

$$N_{mm'jj'} = \langle v_{m',j'}(\boldsymbol{\rho}, \phi) | \hat{\boldsymbol{\epsilon}} \cdot \mathbf{P}_r | v_{m,j}(\boldsymbol{\rho}, \phi) \rangle, \quad (6)$$

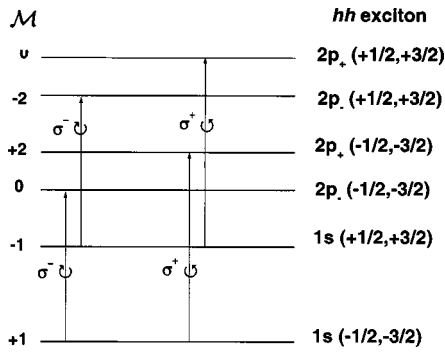


FIG. 1. Schematic diagram of allowed hh intramagnetoexciton transitions under left- (σ^-) and right-hand side (σ^+) circularly polarized light in the GaAs-(Ga,Al)As QW plane. On the left-hand side, \mathcal{M} is the exciton total angular momentum in the z direction.

and using Eq. (5), one obtains

$$N_{mm'jj'} = \frac{2\pi\hbar}{\sqrt{2i}} \delta_{m,m'\mp 1} (A_{jj'm} + B_{jj'm}), \quad (7)$$

with

$$A_{jj'm} = \int_0^\infty \left(\frac{|m|\mp m}{\rho} \right) e^{-\beta_{jj'}\rho^2} \rho^{2p} d\rho, \quad (8a)$$

and

$$B_{jj'm} = \int_0^\infty \left(-\frac{2}{\lambda_j^2} \mp \frac{Be}{2\hbar c} \right) e^{-\beta_{jj'}\rho^2} \rho^{2p+1} d\rho, \quad (8b)$$

with $p = (|m| + |m \pm 1| + 1)/2$, and $\beta_{jj'} = 1/\lambda_j^2 + 1/\lambda_{j'}^2$. If the initial state is the $1s$ -like exciton state, one finds

$$N_{0m'jj'} = \frac{2\pi\hbar}{\sqrt{2i}} \delta_{m',\pm 1} \left(-\frac{1}{\lambda_j^2} \mp \frac{Be}{4\hbar c} \right) \frac{1}{\beta_{jj'}}, \quad (9)$$

and that the $\alpha(\omega)$ magnetoabsorption coefficient for the intraexcitonic $1s \rightarrow np_\pm$ transitions reduces to

$$\alpha(\omega) \propto \frac{1}{\omega} \sum_{m',f} \left| \sum_{k,k',j,j'} (C_{k,k',j',f}^{M,M',m',f}) * C_{k,k',j}^{M,M',m=0,1s} N_{0m'jj'} \right|^2 \times \delta(E_{M,M',m'}^f - E_{M,M',m=0}^{1s} - \hbar\omega), \quad (10)$$

in which final states with $m' = +1$ (-1) correspond to transitions excited by right- (left-hand side) circularly polarized light σ^+ (σ^-). For the δ -function in Eq. (10), we introduced a phenomenological half-width $\Gamma \approx 2 \text{ cm}^{-1}$ by replacing the δ -function by a Lorentzian function in the evaluation of the absorption coefficient. An schematic picture of the allowed $1s \rightarrow 2p_\pm$ hh magnetoexciton transitions associated to the two optically active hh $1s$ ($-1/2, -3/2$) and $1s$ ($+1/2, +3/2$) magnetoexcitons is depicted in Fig. 1, for σ^- - and σ^+ -circularly polarized FIR radiation in the GaAs-(Ga,Al)As QW plane [one could draw an equivalent diagram for the two optically active lh $1s$ ($+1/2, -1/2$) and $1s$ ($-1/2, +1/2$) magnetoexcitons]. Note that the two σ^- (or σ^+) transitions in Fig. 1 are identical if hole-subband mixing is neglected. Also notice that, in the high-magnetic-field limit, when the Coulomb energy of the excitons may be viewed as a small perturbation on magnetic-field effects,

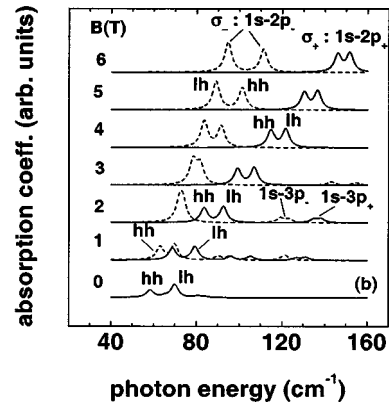
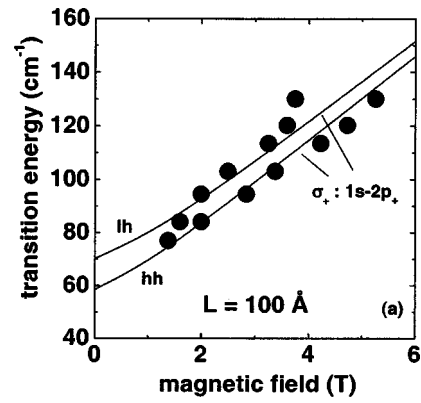


FIG. 2. Results for a $L = 100 \text{ \AA}$ GaAs-Ga_{0.7}Al_{0.3}As QW under a magnetic field applied along the growth direction of the heterostructure: (a) lh and hh $1s \rightarrow 2p_+$ calculated magnetoexciton transition energies (full curves) for the case of right-hand side circularly polarized light (σ^+) in the well plane. Also shown are the experimental data (full circles) taken from Cerne *et al.*,² (b) Intraexcitonic lh and hh $1s \rightarrow np_\pm$ magnetoabsorption coefficient for the case of left- (σ^-) and right-hand side (σ^+) circularly polarized light in the QW plane. The column of numbers on the left-hand side gives values of the applied magnetic field in T.

$\Delta m = +1$ transitions essentially correspond to the excitation of an electron from an electronic Landau level n_e to $n_e + 1$, whereas $\Delta m = -1$ would be associated to the promotion of a hole from the Landau level n_h to $n_h + 1$.

III. RESULTS AND DISCUSSION

In what follows, we compare our theoretical results with experimental measurements, which are performed for GaAs-Ga_{0.7}Al_{0.3}As superlattices (SLs), although we have ignored SL tunneling effects and performed calculations for single isolated GaAs-Ga_{0.7}Al_{0.3}As QWs. We would like to stress that, in the approach by Duque *et al.*,⁷ the exciton envelope wave functions were described as products of variational hydrogeniclike wave functions and electron and hole ground-state solutions of the effective-mass equation, along the z axis, for the barrier potentials of the GaAs-(Ga,Al)As QW. The present scheme considers the various magnetoexciton wave functions as products of symmetry-adapted Gaussian functions with appropriate hole and electron solutions of the QW potentials [contribution of the ground state and excited QW states, cf. Eq. (2)]. As mentioned before, for the hh $1s \rightarrow 2p_\pm$ magnetoexciton transitions, one finds very good agreement between the two ap-

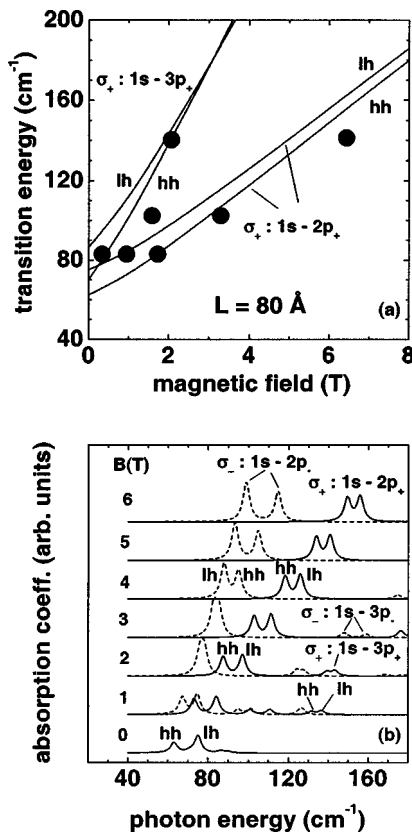


FIG. 3. Results for a $L=80 \text{ \AA}$ GaAs–Ga_{0.7}Al_{0.3}As QW under a magnetic field applied along the growth direction of the heterostructure: (a) lh and hh $1s \rightarrow 2p_+$ and $1s \rightarrow 3p_+$ calculated magnetoexciton transition energies for the case of right-hand side circularly polarized light (σ^+) in the well plane. Also shown are the experimental data (full circles) taken from Salib *et al.*,¹ (b) Intraexcitonic lh and hh $1s \rightarrow np_{\pm}$ magnetoabsorption coefficient for the case of left- (σ^-) and right-hand side (σ^+) circularly polarized light in the QW plane. The column of numbers on the left-hand side gives values of the applied magnetic field in T.

proaches, whereas quantitatively different results¹¹ are found for higher-energy hh $1s \rightarrow 3p_{\pm}$ and lh $1s \rightarrow 2p_{\pm}$ and $1s \rightarrow 3p_{\pm}$ magnetoexciton transitions. Here we just point out that the present scheme better describes higher-energy states as the magnetoexciton envelope wave functions include the effects of excited electron and hole states in its expansion.

The energies corresponding to lh and hh $1s \rightarrow 2p_+$ magnetoexciton σ^+ transitions are shown in Fig. 2(a) for a 100 \AA GaAs–Ga_{0.70}Al_{0.30}As QW. Notice that the experimental FIR data by Cerne *et al.*² are in good agreement with lh and hh $1s \rightarrow 2p_+$ intraexcitonic theoretical transitions. Note that the higher-energy experimental transitions, which Cerne *et al.*² do not assign to any specific intraexcitonic transition, are found to correspond to lh $1s \rightarrow 2p_+$ magnetoexciton σ^+ transitions. Figure 2(b) displays the calculated lh and hh intraexcitonic magnetoabsorption coefficient, for σ^- left- and σ^+ right-hand side circularly polarized light in the well plane, also in the case of a $L=100 \text{ \AA}$ GaAs–Ga_{0.7}Al_{0.3}As QW. One then clearly sees that the σ^- and σ^+ oscillator strengths of the lh and hh $1s \rightarrow 2p_{\pm}$ intraexcitonic transitions are of the same order of magnitude. This unambiguously indicates that both the lh and hh $1s \rightarrow 2p_-$ exciton transitions should be observable in the measured spectra. Also, one

clearly sees, in the low-magnetic-field regime, some weaker, higher-energy features in $\alpha(\omega)$, which correspond to lh and hh $1s \rightarrow 3p_{\pm}$ exciton transitions. These features would be observable in the $L=100 \text{ \AA}$ GaAs–Ga_{0.7}Al_{0.3}As QW provided one is able to perform the experiment with higher spectral resolution.

Figure 3(a) shows the energies for the σ^+ -excited lh and hh $1s \rightarrow 2p_+$ and $1s \rightarrow 3p_+$ exciton transitions in the case of a $L=80 \text{ \AA}$ GaAs–Ga_{0.7}Al_{0.3}As QW. A comparison of the present σ^+ -excited magnetoexciton theoretical transitions with available FIR measurements¹ indicates fair overall agreement, although the assignment of some of the experimental features to specific intraexcitonic transitions is uncertain. Again, results in Fig. 3 indicate that measurements with left-hand side σ^- circularly polarized FIR radiation would reveal the corresponding lh and hh $1s \rightarrow 2p_-$ and $1s \rightarrow 3p_-$ magnetoexciton transitions. We note that the present calculations indicate that the observed intraexcitonic transitions occur in both hh and lh magnetoexcitons, which contrasts with the assignment by Salib *et al.*¹ of the observed FIR resonances to hh transitions only. Also, results in Fig. 3 indicate that the experimental feature at $2T$ and $\approx 140 \text{ cm}^{-1}$ would correspond to the lh and hh $1s \rightarrow 3p_+$ transitions, and not to the hh $1s \rightarrow 4p_+$ transitions, as suggested by Salib *et al.*¹

We stress that the present calculation does not include the effects due to the hole-subband mixing. This approximation should be reasonable provided the QW width is not too large, in which case the mixing between lh and hh valence states could be significant. Experimental FIR data⁶ for GaAs–(Ga,Al)As multiple QWs, with well widths of 125, 150, and 200 \AA , seem to be associated to two distinct hh $1s \rightarrow 2p_-$ magnetoexciton transitions, corresponding to strong hole-subband mixing. The experimental findings for these “large” $L \geq 125 \text{ \AA}$ well-width multiple QWs may be interpreted as follows: A 6×6 Kohn–Luttinger calculation for excitons would lift the degeneracy of both σ^- (or σ^+) transitions (cf. Fig. 1), although one should expect a very small spin splitting^{9,10} of the electron states for intramagnetoexciton transitions associated with “electron transitions” ($\Delta m = +1$) within the exciton, and therefore would obtain two near degenerate $1s \rightarrow 2p_+$ transition energies; on the other hand, a full 6×6 (or 4×4) calculation of the internal excitonic transitions related to “hole transitions” ($\Delta m = -1$) would certainly imply in a measurable difference for the two $1s \rightarrow 2p_-$ transitions, as hole-spin splitting is quite large.^{9,10} In that respect, a full quantitative understanding of the experimental data related to the $L \geq 125 \text{ \AA}$ multiple QWs would certainly require a full Kohn–Luttinger calculation for the intramagnetoexciton transitions.

IV. CONCLUSIONS

Summing up, theoretical results in GaAs–(Ga,Al)As QWs are obtained for intramagnetoexciton transition energies corresponding to excitations from $1s$ -like to $2p_{\pm}$ - and $3p_{\pm}$ -like magnetoexciton states. We have also presented results for the $\alpha(\omega)$ magnetoabsorption coefficient corresponding to the intraexcitonic $1s \rightarrow np_{\pm}$ transitions, for the case of

σ^- left- and σ^+ right-hand side circularly polarized photons in the QW interface. Our results demonstrate that the σ^- and σ^+ oscillator strengths of the lh and hh $1s \rightarrow np_{\pm}$ intraexcitonic transitions are of the same order of magnitude, and that the lh and hh $1s \rightarrow 2p_{-}$ exciton transitions should be observable in both the $L=80 \text{ \AA}$ and $L=100 \text{ \AA}$ GaAs–Ga_{0.70}Al_{0.30}As QWs measured spectra if one uses FIR σ^- circularly polarized photons. Theoretical results suggest that the higher-energy experimental magnetoexciton transitions in the FIR data for the $L=100 \text{ \AA}$ GaAs–Ga_{0.70}Al_{0.30}As QW, which were not assigned to any specific intraexcitonic transition, would correspond to lh $1s \rightarrow 2p_{+}$ magnetoexciton σ^+ transitions. Also, the present calculations unambiguously indicate that the observed intraexcitonic transitions occur in both hh and lh magnetoexcitons, which contrasts with previous assignment of the observed FIR resonances to hh transitions only. Finally, further experimental and theoretical studies, with both right- and left-hand side polarized FIR radiation in the plane of the QW, are forthcoming to unambiguously associate specific hh or lh intraexcitonic transitions to FIR features in the measured spectra.

ACKNOWLEDGMENTS

This research was partially supported by Red IX.E (CYTED), Millennium Scientific Initiative/Chile (Condensed Matter Physics, Grant No. P 99-135-F), Fondo Nacional de Ciencias/Chile (Grant Nos. 1010429 and 7010429), and Universidad Santa Maria (internal grant). The authors

also acknowledge the Colombian (CODI-Universidad de Antioquia) and Brazilian Agencies (Fapesp, CNPq, and FAEP-UNICAMP) for partial financial support.

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