PHYSICAL REVIEW B VOLUME 50, NUMBER 7 15 AUGUST 1994-I

Shake-up intersubband processes in quantum-well luminescence

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(Received 31 March 1994)

Satellite peaks have been observed in undoped and *p*-doped GaAs/$Al_xGa_{1-x}As$ quantum wells (QW's) in selective photoluminescence spectra upon excitation resonant with the free-exciton states. The satellites are interpreted as being due to shake-up transitions involving QW hole subbands, in which valence-band holes are shaken up from the lowest heavy-hole (hh) band to the higher light-hole or the $n=2$ hh band; thus these shake-up satellites are of intrinsic origin. The prerequisite for the shake-up process is sufficient overlap of the wave function of a free exciton and an adjacent weakly localized hole. This localization is proposed to be due to interface roughness as evidenced by the Stokes's shift for the free exciton. Calculations have shown that interface roughness can give rise to the required localization by producing localized states that are weakly bound to each subband state. The measured intersubband transition energies agree with independent experimental results and theoretical predictions. A striking enhancement of the shake-up satellite intensity is observed with increasing applied magnetic field. This fact is consistent with an increasing localization of the exciton due to the compression of the wave function. The localization becomes increasingly important when the wave function of the exciton is comparable with or smaller than the lateral size of the island characterizing the interface. From the magnetic-field dependence of the shake-up satellite intensity we can roughly estimate the island size. In fact, the magnetic-field dependence can be utilized for probing the interface quality.

I. INTRODUCTION

Shake-up effects have been reported for radiative Auger processes, in which part of the recombination energy is transferred to a free or bound carrier.[1,2] Accordingly, the energy of the photon emitted in the presence of the shake-up process is smaller than in the corresponding case without shake-up. The energy difference corresponds to the energy needed for the electron-hole excitation. This process has been extensively studied for the case of bulk semiconductors.[2,3] A well-known example is the so-called two-electron (hole) transitions, TET's (THT's) of a bound exciton (BE), in which the interaction between an exciton and the neutral impurity binding the exciton results in a shake-up process. While one electron (hole) radiatively recombines with the exciton hole (electron), the second electron (hole) is simultaneously shaken up into an excited state of the donor (acceptor).[3] The orbital momentum and its projection should be conserved in these transitions according to the selection rules applicable for the shake-up processes.[2] This means that *s*-like states dominate in the final state after the shake-up process in contrast to the selection rules applicable to infrared absorption processes, in which transitions to *p*-like excited states are allowed. Corresponding shake-up or THT's (TET's) do not occur for free excitons (FE's) in bulk material, because exciton localization is a prerequisite to achieving sufficient overlap between the exciton and the additional hole (electron).

More recently, many-body shake-up processes have been reported for quasi-two-dimensional (2D) systems.[4] Shake-up processes occur in the presence of a 2D electron gas (2DEG), when optical transitions creates excitations in the 2DEG. The redshift in the satellite spectrum caused by the shake-up corresponds to the excitation created. An interesting effect is observed in the presence of a magnetic field. A series of low-energy satellites corresponding to inter-Landau-level excitations of the 2DEG have been reported.[4] The efficiency of the shake-up process is shown to be dependent on the localization of the valence-band hole.

In the present paper we report on another type of low-energy satellite observed in association with a localized exciton in selective photoluminescence (SPL) spectra of undoped or *p*-type quantum wells (QW's). These satellites have earlier been interpreted as shake-up transitions involving QW hole subbands.[5] Valence-band holes are shaken up from the lowest heavy-hole (hh) band to higher light-hole (lh) or hh bands. A prerequisite for such a shake-up transition is the localization of the states attached to the QW subbands. The localization of these states is caused by the interface roughness; thus these transitions are intrinsic. We now present magneto-optical data supporting the earlier proposed model.

II. EXPERIMENT

Samples grown by molecular-beam epitaxy were used in this study. Fifty periods of GaAs QW's sandwiched between 150-Å $Al_{0.3}Ga_{0.7}As$ barriers were grown at nominally 680 °C without interruption on top of a semi-insulating GaAs (100) substrate with a 0.35-μm undoped

GaAs buffer layer. Samples with a QW width in the range $40 < L_z < 150$ Å were employed in this investigation. Most of the QW structures were intentionally undoped (with $[p]$ in the 10^{14}-cm^{-3} range for the GaAs QW's), but also p-doped QW's were studied.

For the SPL and PL excitation (PLE) measurements an Ar^+ ion laser was used to pump a titanium-doped sapphire solid-state laser. The emitted light from the samples, perpendicular to the incident beam, was focused on the slits of a 1-m double-grating monochromator. The PL signal from the monochromator was detected with a dry-ice-cooled GaAs photomultiplier.

The magneto-optical measurements have been performed in a solenoid magnet system with a maximum field of 16 T applied perpendicular to the QW layers. The same laser and detection system as described above was used in the magneto-optical experiments. The sample temperature could be continuously varied from 1.6 K up to room temperature. The laser light was coupled via an optical fiber on to the QW sample. The emission light from the sample was coupled via the same fiber to be monitored in the detection system.

III. SELECTIVE LUMINESCENCE

Interface roughness is known to influence the optical properties of a QW structure. The roughness gives rise, e.g., to line broadening in the PL spectra due to local fluctuations in the subband energies[6] and a redshift, usually denoted the Stokes shift, between the exciton energy as observed in emission (PL) versus absorption (PLE).[5,7] The Stokes shift is explained in terms of exciton trapping at interface irregularities.[7] The absorption (PLE) spectrum reflects the density of states for the energy distribution of the excitons, while the corresponding PL spectrum is redshifted due to exciton capture before the recombination. The localized exciton (LE) is therefore a more adequate notation for the free exciton in PL. Accordingly, the Stokes shift demonstrates experimentally the existence of localization effects in the QW.

By comparing PL and PLE spectra of the QW's, the Stokes shifts for the different samples have been estimated. The Stokes shift exhibits a sample dependence reflecting the smoothness of the interface achieved at growth. For instance, by using interrupted growth smoother interfaces, i.e., the interfaces with atomically flat areas or islands of larger diameter, are achieved. There is also a more systematic trend with an increasing Stokes shift with decreasing QW width. This can be explained in terms of an increasing ratio between the interface volume (where interface fluctuations occur) in comparison to the total volume with decreasing QW width. A change in the QW width L_Z gives rise to a relatively larger change in the potential energy E of a confined state for a more narrow QW according to $dE/dZ \approx (\hbar^2\pi^2/m)(1/L_Z)^3$. Consequently, the localization becomes increasingly important, when the QW width is reduced.

We originally observed the shake-up satellites together with the "normal" THT satellites, corresponding to acceptor excitations from the 1S ground state to the first excited 2S state, in the SPL studies of p-doped QW's.[8] Both these features are weak, typically 3–6 orders of magnitude lower intensity than the exciton peaks, with the high-energy satellite as the least weak. The satellites usually render undetectable, if the excitation is shifted off resonance. However, there were striking differences in their behavior. While the THT satellites were obviously enhanced upon excitation resonantly with the BE state, as expected, a corresponding intensity enhancement for the satellites was achieved by excitation resonantly with the FE state. This is clearly illustrated in the SPL spectra shown in Fig. 1 for a 50-Å-wide QW, doped with Be to a level of 3×10^{17} cm^{-3} in the central 10 Å.

The observed intensity correlation with the FE instead of the BE implies an intrinsic origin of the satellites. This implication leads us to perform corresponding SPL investigations of undoped QW's. In fact, the corresponding satellites appeared also in the intentionally undoped structures. In most cases, two different satellites were detectable. The energy separations between the LE and the shake-up satellites are plotted as a function of QW width in Fig. 2. In the same figure, the well width dependence of the energy separations between the FE^{hh} and the FE^{lh} as derived from PLE spectra are plotted. Also the theoretically predicted well width dependences of the energy separations between the $n=1$ hh subbands and partly the $n=1$ lh and partly the $n=2$ hh subbands are plotted in Fig. 2. The resemblance between these sets of data implies a similar origin, i.e., intersubband transition energies. It should, however, be remembered that the PLE data do not measure the intersubband energies directly, but include also the exciton binding energies.

The intensities of the satellites are rapidly decreasing with the temperature and render undetectable already at about 10–20 K. The temperature dependence corresponds to an activation energy of a few meV, approximately similar to the localization energy as derived from the Stokes shift.

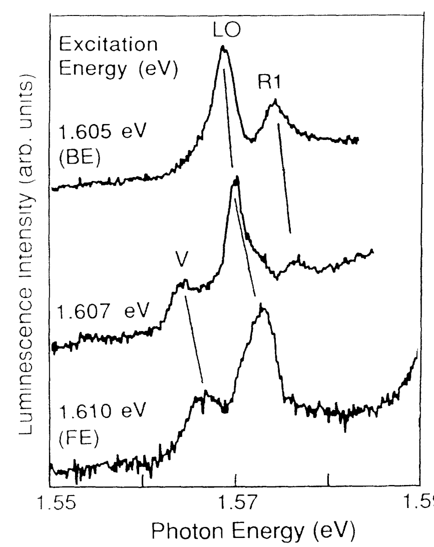

FIG. 1. SPL spectra of an acceptor doped 50-Å-wide QW measured at 1.6 K with excitation resonant with the BE (top spectrum), the FE (bottom spectrum) and in between (the middle spectrum).

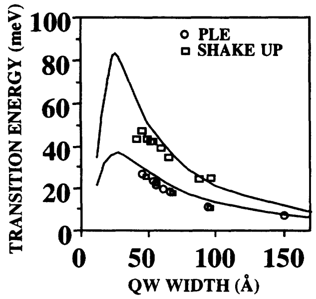

FIG. 2. The energy separations between, on one hand, the FE and the shake-up satellites obtained from the SPL spectra and, on the other hand, the FE^{hh} and FE^{lh} observed in PLE spectra. The solid lines represent the theoretically predicted energy separations between the hh and lh states and the $n=1$ and $n=2$ hh states, respectively.

IV. MAGNETIC-FIELD DEPENDENCE

In the presence of a magnetic field, the FE peak position exhibits a blueshift in PL spectra characterized by the diamagnetic shift. The magnetic-field dependence for the FE peaks for the case of a 50-Å-wide QW measured in PL and PLE, respectively, is shown in Fig. 3. The diamagnetic shift of the FE^{hh} in PL measures to 1.4×10^{-2} meV/T^2 in this case. It should be noted that the FE^{hh} peak is slightly downshifted in PL versus PLE with increasing magnetic field. The Stokes shift increases from ≈ 2.7 meV at 0 T to ≈ 3.9 meV at 16 T. This is explained in terms of an increasing degree of localization for the FE because of the compression of the wave function at higher magnetic fields. This point will be further expounded below. In the same figure, the magnetic-field dependence of the first shake-up satellite position is shown. The shake-up satellites are interpreted as being due to hole intersubband excitation processes involving an exciton and an adjacent hole, both trapped at the same localization potential. The hole may be excited from its *s*-like ground state (attached to the $n=1$ hh subband) to an *s*-like state attached to a higher-energy subband such

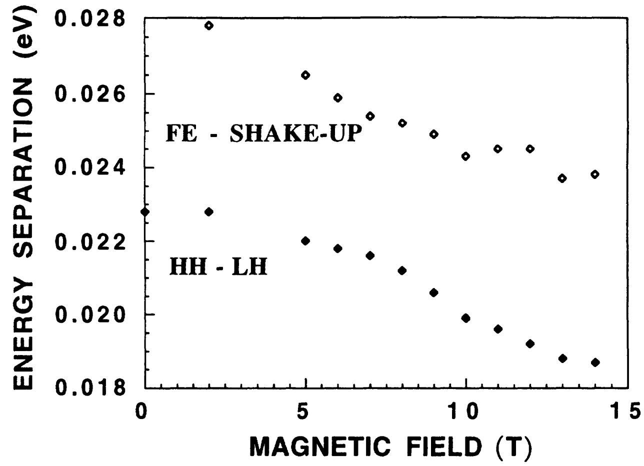

FIG. 4. The energy separations between the FE^{hh} and the FE^{lh} states as observed in PLE in comparison with the energy separation between the $n=1$ FE^{hh} as observed in PL and the first shake-up satellite as observed in SPL as a function of the applied magnetic field.

as the $n=2$ hh state or the $n=1$ lh state. Consequently, the emitted photon will be reduced in energy by approximately the subband energy separation. Accordingly, we compare in Fig. 4 the energy separation, on one hand, between the FE as observed in PL and the shake-up satellite and, on the other hand, between the FE^{hh} and the FE^{lh} as derived from PLE spectra. First, we note that the hh/lh band separation decreases with increasing magnetic field as expected.[9] Second, the magnetic-field dependences of the observed energy separations are not found to overlap, which might be expected at first sight. Instead, the PLE results are downshifted by ≈ 5 meV versus the satellite data. However, there are two important points to keep in mind. As described above, we do not measure the difference in intersubband energies directly in the PLE measurements, but the related exciton energies. It is well known that the binding energy of the FE^{lh} exceeds the corresponding FE^{hh} energy.[10] Also, a prerequisite for the shake-up process is the localization of the exciton as well

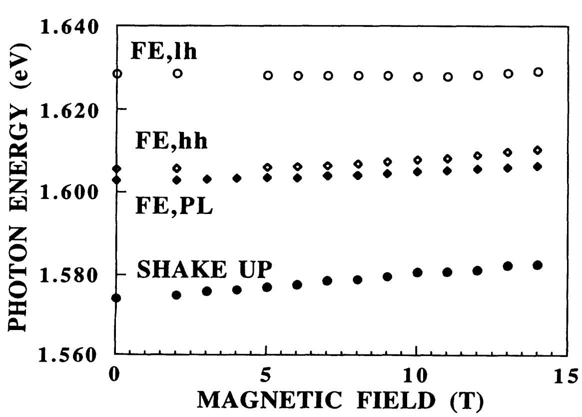

FIG. 3. The magnetic-field dependence for the energy positions of the FE's observed in PL and PLE together with the shake-up satellite observed in SPL.

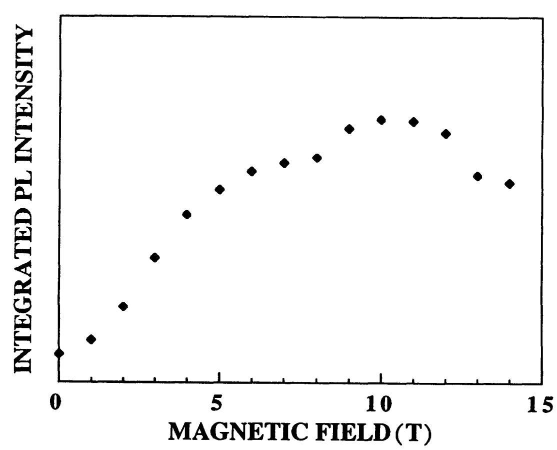

FIG. 5. The magnetic-field dependence of the intensity of the first shake-up satellite.

as the hole. This means that the initial state involves not a free hole, but a localized hole. Consequently, the upper set of data in Fig. 4 should correspond to the intersubband energy plus the hole localization energy. Both these factors tend to reduce the observed energy difference.

Another important observation to be stressed is the intensity enhancement of the shake-up satellite intensity with increasing applied magnetic field as illustrated in Fig. 5. As can be seen in this figure, the integrated intensity is constantly increasing to reach a maximum at about 10 T. The enhancement effect is striking, almost an order of magnitude at maximum in comparison with zero-field conditions. This point will be further expounded below.

V. DISCUSSION

As for the FE, most of our now considerable knowledge on the BE states is obtained from optical spectroscopy. Among many important steps in this development, the observation of the two-particle transitions (TPT's) of the BE should be noted here. The observation of these usually well-defined and narrow TPT lines thus constitutes an important way to provide detailed information on the impurity excited states and for accurate determinations of the impurity transition energies.[3,8]

Corresponding TPT's do not occur for FE's in bulk material, since the prerequisite of exciton localization in order to achieve wave-function overlap between the exciton and another electronic particle is generally not fulfilled. However, the situation is different for QW's. Theoretical treatment of the localization effects have shown that any island size will localize a state in a QW, i.e., that interface roughness will always produce bound states with a localization energy, which depends on the radius and depth of the interface island.[7,11] The migrating FE's get trapped at an area with the locally lowest potential at low temperatures. The FE's are thus effectively localized at lowest temperatures, forming LE's (Ref. 12) and can be treated similarly to bound excitons.[13] The LE's can only pass the potential barriers by gaining energy from, e.g., optical or thermal excitation or by emitting an acoustical phonon.[7] The localization effects have been demonstrated by studies on the FE dynamical processes,[13,14] the FE linewidths in PL,[6,15] and by the difference for the FE energy between PL and PLE spectra, the Stokes shift.[7]

In this paper, we present results derived from SPL experiments on the LE in GaAs/$Al_xGa_{1-x}As$ QW's performed with and without an applied magnetic field. Upon excitation resonant with any of the LE states, satellites appear at lower photon energies. By analogy with extrinsic two-particle shake-up transitions at neutral acceptors or donors in doped QW's,[8,16] we interpret these satellites as corresponding intrinsic two-particle or shake-up transitions. A small equilibrium population of holes is assumed in the samples used, either p type or undoped with p-type background doping. The holes are localized due to the interface roughness at the locally lowest potential. By the laser excitation, an exciton is formed, which modifies the interface roughness potential binding the hole. This sudden perturbation may excite the hole from its ground state attached to the $n=1$ hh subband to a higher state. The shake-up satellite recombination energy will be redshifted versus the exciton by approximately the subband energy separation. We have in this study observed shake-up satellites corresponding to transitions from the $n=1$ hh subband to the $n=1$ lh and the $n=2$ subbands, respectively. This interpretation is supported by comparing the derived intersubband transition energies partly with theoretically predicted intersubband energies and partly with results achieved from PLE spectra shown in Fig. 2. The intersubband transition energies derived from the SPL measurements are slightly shifted towards lower energies compared to the calculated values. However, the satellite data are in very good agreement with the data for energy separation between the $n=1$ hh and the $n=1$ lh FE peaks measured using PLE. This close agreement between the results achieved from different experimental techniques, the satellites observed in SPL and PLE measurements, and theoretical predictions supports our interpretation of the satellite states as originating from an intersubband shake-up process.

In the presence of a magnetic field, the FE peak position exhibits a blueshift as illustrated in Fig. 3. What is notable in this figure is the different blueshift rate with increasing magnetic field for the FE^{hh} in PL versus the FE^{hh} in PLE. In other words, there is a significant magnetic-field dependence in the Stokes shift term. Another related effect is the striking satellite intensity enhancement with increasing magnetic field up to ≈ 10 T. In order to understand the underlying physical mechanism, a comparison with the corresponding magnetic-field dependence for the extrinsic shake-up processes is informative. For the case of the donor BE, a similar TET satellite enhancement in the presence of an applied magnetic field is observed,[16] which is in sharp contrast with the THT of the acceptor BE, for which no significant enhancement effect is observed.[8] The analogy with the donor BE implies that the enhancement effect is related to the wave-function extension. When the magnetic field is increased, the exciton wave function is increasingly compressed, resulting in an increasing degree of localization. This localization effect is strongest when the dimension of the "flat island" radius is comparable with the exciton Bohr radius.

The extension of the exciton in-plane wave function in the presence of a magnetic field applied in the z direction can be roughly estimated by

$$\langle \rho \rangle = \frac{\langle \Psi_{ex} | \rho | \Psi_{ex} \rangle}{\langle \Psi_{ex} | \Psi_{ex} \rangle} , \quad \rho^2 = x^2 + y^2 . \tag{1}$$

The simple exciton trial wave function is given by

$$\Psi_{ex} = N \Psi_e(z_e) \Psi_h(z_h) f_1(\rho) f(\rho) , \tag{2}$$

where

$$f(\rho) = e^{-\rho/\alpha} , \tag{3}$$

$$f_1(\rho) = \frac{1}{\pi^{1/4}\sqrt{L_m}} e^{-\rho^2/2L_m^2} , \tag{4}$$

α is a variational parameter, which reflects the extensioı of the exciton wave function in the xy plane at zero mag netic field. $\Psi_e(z_e)$ and $\Psi_h(z_h)$ correspond to the wav function of the free electron and free hole, respectively related to the lowest subband of the QW structure. L_m i the magnetic length, $L_m=\sqrt{\hbar/eB}$. The electron-hol wave function is divided into two parts: $f_1(\rho)$ is th lowest harmonic-oscillator-like wave function of the rela tive electron-hole motion in the xy plane with a magneti field applied perpendicular to this plane, while $f(\rho)$ cor responds to the simplified electron-hole trial wave func tion in the xy plane related to the Coulomb interaction The calculated magnetic-field dependence of the exten sion of the exciton wave function based on Eq. (1) is shown in Fig. 6 for some different values on α. Since the simplified $f(\rho)$ function in Eq. (4) is used, the wave-function extension described by $\langle \rho \rangle$ is not directly related to the $\Psi_e(z_e)$ and $\Psi_h(z_h)$, but depends indirectly on the QW width via the α parameter. Since the α parameter reflects the extension of the exciton wave function in the xy plane, it will be influenced by the QW width via the compression of the wave function in the z direction.

Following the arguments given above, the localization effect increases with increasing magnetic field until the dimension of the "flat island" is comparable with the exciton Bohr radius. The intensity enhancement of the satellite reaches its maximum at ≈ 10 T (Fig. 5). According to Bastard,[17] the exciton transversal radius is 160 Å at zero-field conditions for a 50-Å-wide QW surrounded by infinite barriers. This approximate value deviates from the experimental conditions in at least two respects; the barriers are taken as infinite and no exciton localization is taken into account. Assuming that these factors cancel and we keep at an exciton radius of 160 Å for a 50-Å-wide QW at zero magnetic field,[17] we can estimate the wave-function compression in Fig. 6 to about 90 Å at 10 T. From the same figure, we get a rough estimate of the accuracy for the radius determination. An error bar of ± 20 Å for the transverse exciton radius at zero field corresponds to an error bar of ± 10 Å at 10 T. We can accordingly draw conclusions about the interface smoothness and roughly estimate the dimensions of the "flat islands." Although the accuracy of this estimate is limited,

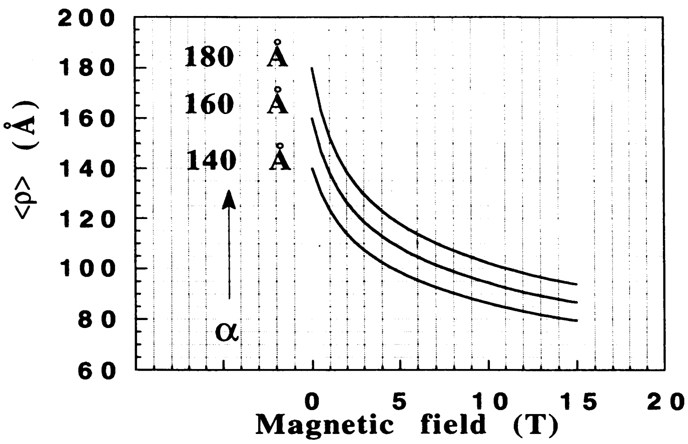

FIG. 6. The calculated magnetic-field dependence of the extension of the excitonic wave function. The calculations are performed for three different values on α, the exciton transversal radius at zero-field conditions.

it should be remembered that the corresponding information is difficult to experimentally achieve in a direct way and has never been demonstrated up to now, to the best of our knowledge.

VI. CONCLUSIONS

We report on shake-up transitions involving QW hole subbands in undoped QW's. These transitions are explained in terms of localized states attached to the QW subbands. We suggest that these states are localized by interface roughness; thus these transitions are intrinsic. Thus, the observation of the intersubband shake-up processes yield direct information about the hole intersubband energies without exciton interaction. The satellite intensity is found to be significantly enhanced in the presence of a magnetic field due to an increasing localization until the dimension of the "flat island" is comparable with the exciton Bohr radius in the plane of the well. From the magnetic-field dependence of the shake-up satellite intensity we can roughly estimate the radius of the "flat island." Accordingly the method demonstrated can be used as a way to probe the interface roughness.

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