Photoluminescence in modulation-doped GaAs/$Ga_{1-x}Al_xAs$ heterojunctions

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(Received 27 April 1998; revised manuscript received 20 July 1998)

We investigated the physical properties of modulation-doped GaAs/$Ga_{1-x}Al_xAs$ heterojunctions by photoluminescence. We found two types of transitions and attributed them to the space direct excitons in the GaAs flat band region and the space indirect excitons near the heterojunction notch. We propose the vertical transport of photoexcited carriers prior to the actual exciton formation under the influence of the two-dimensional carrier gas. Therefore, the photoluminescence intensities of the two types of excitons are correlated. The vertical transport model explains successfully the experimental results of the optically detected cyclotron resonance, the photoluminescence intensity oscillation with magnetic field and the long rise time of the photoluminescence intensity for the excitonic transitions. [S0163-1829(99)09011-6]

I. INTRODUCTION

Modulation-doped GaAs/$Ga_{1-x}Al_xAs$ heterojunctions are known to be useful for the fabrication of high-mobility devices. Such structures are also frequently used to demonstrate the fundamental physics related to a two-dimensional carrier gas (2DCG), in particular, the quantum Hall effect. Many experimental methods have been applied to reveal the many-body interactions in the 2DCG, where photoluminescence (PL) is known to provide direct information regarding the carrier self-energies. However, publications based on optical spectroscopy are in some cases still controversial concerning the sample structures used and the spectral line shapes obtained. Buhmann *et al.* have introduced a *p*-type δ-doping layer in GaAs for *n*-type heterojunctions in order to trap photoexcited holes near the 2DCG.[1] The PL spectrum obtained has only a broad feature and develops into Landau levels in high fields, whereas in heterojunctions without such a *p*-type δ-doping layer, several authors observed sharp peaks at energies close to the GaAs excitons that do not develop into Landau levels in high fields.[2–4] However, two-dimensional behaviors such as PL intensity oscillations in the field and the peak energy discontinuity have still been observed. Because of these two-dimensional properties, the PL peaks were assigned to transitions from the first and second subbands of the heterojunction notch and no discussion about the transitions from GaAs excitons was carried out.[3] This assignment, however, is not in good agreement with the research results published before. Yuan *et al.*[5] have found a broad PL line shape besides a group of sharp peaks. They named the broad peak the *H* band and assigned the rest of the peaks to the GaAs excitons. They attributed the *H* band to the transition from the 2DCG to the photoexcited holes in the GaAs flat band region. It was shown later by magneto-PL experiments that this *H* band might correspond to the recombination of electrons from the second subband instead of the lowest subband in heterojunctions.[6] This assignment seemed to be confirmed by recent time-resolved PL measurements and theoretical calculations of the transition lifetime by Lundström *et al.*[7] Some other authors[8] referred the *H* band to defect pairs situated at the interface although their arguments become weak as the sample quality improves. The above controversy calls for the necessity of a careful reexamination of the origin of these transitions.

To review the investigation on the quantum confined Stark effect (QCSE) (Ref. 9) would be a great help to achieve a deeper understanding of the PL in modulation-doped heterojunctions. The QCSE shows that the exciton peak energy in the quantum well can be reduced by several tens of meV by applying an external electric field of several tens of kV/cm along the sample's normal direction. Hence, due to the strong interface electric field in heterojunctions[10] the transitions with energies close to or even coinciding with the GaAs excitons can hardly be assigned to the transitions from the 2DCG.

Most publications also showed that the PL peaks in heterojunctions shift diamagnetically,[2] which suggests that the transitions are excitonic. Similar conclusions can be found for double heterostructures[11] and for heterojunctions under selective excitation.[12] According to the phase filling theory of Schmitt-Rink,[13] excitonic transitions cannot originate from the 2DCG. Even considering simply the screening effect, we find it quite suspect to assign peaks with an energy

TABLE I. Parameters of GaAs/$Ga_{1-x}Al_xAs$ heterojunctions.

Sample	1 (n)	2 (n)	3 (p)	4 (p)	5 (p)	6 (p)
Al%	33	33	60	56	50	50
n,p ($10^{11}/cm^2$)	4.3	3	1.99	6.72	8.3	
μ ($cm^2/V\,s$)	2×10^6	2.4×10^5 (2 K)	7100 (0.5 K)	5600 (0.5 K)	3128 (77 K)	
$m_h^*(m_o)$			0.48		0.5	0.48
$m_e^*(m_o)$	0.073	0.078				

exactly coinciding with GaAs excitons to transitions from a 2DCG.

In order to clarify more exactly the excitonic nature of the PL transitions in heterojunctions, we will review the PL spectra in modulation doped quantum wells (QW's) for comparison. In modulation doped QW's with well width below a certain value, the PL originates from band-band transition involving the 2DCG. The zero-field PL spectrum has an exponential intensity decrease on the high-energy side due to the Boltzman distribution of the photoexcited holes and a broadened band edge on the low-energy side due to the band-edge fluctuations. The PL line shape can be well described taking into account the breaking of the selection rule for the dipole transitions.[14] The line shape is non-Gaussian and, under some conditions, the Fermi edge singularity can be observed when the carrier localization plays an important role.[15] In magnetic fields, the PL spectra develop into discrete peaks characterizing both diagonal (obeying the dipole selection rule) and nondiagonal (breaking the dipole selection rule) transitions between Landau levels of the conduction and valence band.[14] The peaks shift linearly in low magnetic fields and the energy positions oscillate slightly in high magnetic fields due to the oscillation of the electron and hole self-energy.[16] These phenomena are, in principle, different than those observed in heterojunctions without an intentional δ-doping layer. Therefore, the origins of the transitions in QW's and heterojunctions should be different. In QW's, the transitions are from the 2DCG while in heterojunctions the transitions are excitonic and should not involve the 2DCG.

The following question arises: if these transitions do not involve the 2DCG, why do they still exhibit the two-dimensional behavior of the 2DCG? In our previous publications, we have briefly introduced a carrier vertical transport model to solve this problem.[17] In this paper, we will present our experiments and analysis in more detail. After a brief introduction of our measurement system, we will first present our results of magneto-PL spectroscopy. From the peak positions, diamagnetic shifts and the rotation symmetry argument we will demonstrate that the PL spectra we observed are the recombinations of GaAs excitons or excitons near the flat band region and they do not involve the 2DCG. However, in optically detected cyclotron resonance (ODCR), the PL intensities of these excitons do exhibit the properties of the 2DCG. This extraordinary experimental behavior is explained by a vertical transport model of the photoexcited carriers. Following this discussion, we present further experimental evidence for the vertical transport model with time-resolved PL spectra and the PL intensity oscillations with magnetic field.

II. EXPERIMENTAL SYSTEMS AND SAMPLE PARAMETERS

We have constructed two ODCR measurement systems, one with a solonoid and one with a split coil magnetic cryostat. The sample normal could be rotated with respect to the magnetic field in the split coil magnetic cryostat. For the ODCR and the magneto-PL experiments, argon ion lasers were used as excitation source. The PL spectra were measured by a 1-m Jobin-Yvon spectrometer with a high spectral resolution and collected by a photomultiplier. The microwaves are generated by a backward wave oscillator (BWO) (Ref. 18) with a maximum output power of 200 mW. The microwave frequencies could be varied continuously between 35 and 183 GHz. The microwaves were chopped to on and off states. A photon counter detected the photoluminescence signal with microwaves on (A) and off (B) separately. The differential signal, $A-B$, reflects the microwave effect on the photoluminescence intensity. The ODCR signal was obtained by adjusting the spectrometer to follow the peak position with increasing magnetic field under the microwave excitation. For the time-resolved PL experiments, a mode locked titanium sapphire laser with a frequency doubler was used as excitation. The pulse width of the mode locked laser was 120 fs. The time-resolved PL spectra were measured by a Hamamatsu streak camera (C4334) with a time resolution of 5 ps.

The samples studied were modulation doped n- and p-type heterojunctions. The sample parameters are listed in Table I. The values of the effective masses of electrons and holes confined in the notch are measured by the ODCR. The samples investigated in this work were not δ doped in the GaAs layer for preventing the escape of the photoexcited carriers as mentioned before. Photoexcited holes in n-type or electrons in p-type heterojunctions were allowed to drift away from the heterointerface.

III. BLUESHIFT AND DIAMAGNETIC SHIFT

Figure 1(a) shows typical PL spectra with increasing laser excitation power similar to those in other publications (e.g., Ref. 5). Two types of transitions are observed. They are a broad peak, shifting to higher energies with increasing excitation intensity, and a group of narrow peaks, remaining fixed. All narrow peaks are located exactly at the energy positions typical for excitons of three-dimensional GaAs. They correspond extremely well to the published data listed below: the upper polaron branch of free excitons FX's at 1.5153 eV, the lower polaron branch of FX's at 1.515 eV, the excitons bound to neutral donors (D°,X) at 1.5141 eV, the excitons bound to ionized donors (D^+,X) at 1.5133 eV

and the three spin-split states of the acceptor bound excitons (A°,X) at 1.5128 eV for $J=\frac{1}{2}$, 1.5124 eV for $J=\frac{3}{2}$, and 1.5122 eV for $J=\frac{5}{2}$.[19] Because of this exact coincidence of the peak positions with literature values, these transitions can obviously not be assigned to be from the 2DCG.

The broad transition peak, referred to as the *H* band, lies between the GaAs exciton and the GaAs donor-acceptor pair transitions. The most outstanding feature of the *H* band is its blueshift with increasing laser power as shown in Fig. 1(a). Some other features of the *H* band published so far include the peak shift with the temperature,[5] the Al composition in the barrier and the carrier concentration, or by means of an external electric and magnetic field.[8,20] Furthermore, the lifetime of the *H* band is extremely long and depends on its transition energy.[21,22] The *H* band disappears completely when the heterointerface is removed.[5]

However, not every sample exhibits an obvious *H* band. Especially samples with a high mobility of the 2DCG show mainly a group of features at the energies of the GaAs excitons. Nevertheless, through careful analysis of the spectra in the excitonic region with increasing excitation intensity, we can identify a peak with the same characteristic blueshift as the *H* band, although it is not so pronounced, see Fig. 1(b). Because of this characteristic blueshift, we believe that this slightly broadened peak that overlaps the GaAs excitonic transitions should have the same origin as the *H* bands at low-energy positions. The recombination energies of the *H* band are found, therefore, to be able to approach the energies of the FX's. The above experimental results suggest that the *H* band should also be of excitonic origins in GaAs. We will show that the excitons for the *H* band are distorted to a varying extent by the corresponding interface electric field. In the following, we will present the diamagnetic shifts and the rotation symmetry of the *H* bands at lower- and higher-energy positions.

Figure 2 depicts the magnetic-field-induced energy shifts for *H* bands of two different samples. A clear diamagnetic behavior can be found in both cases at low fields although the energy shifts are different. This implies that the *H* band is of excitonic origins. The *H*-band emission cannot come from the 2DCG since the transition involving the 2DCG can only be a band-band transition due to the phase filling effect.[13] With increasing magnetic field a band-band transition will shift linearly to higher energies, which contradicts our observations and also the experimental results published so far.

The nonlinearity of the diamagnetic shift is energy dependent, with a stronger shift for the *H* band at lower energies. Since a smaller energy shift indicates a larger exciton binding energy, the binding energy of the *H*-band excitons increases with their increasing recombination energy.

The reduction of the recombination and binding energy of the *H*-band excitons can be understood with the theory of the QCSE. According to the QCSE, the recombination energy and the exciton binding energy will be reduced by an external electric field in QW's.[9] Similarly, the interface electric field within the heterojunction should reduce the recombination and binding energy of the excitons located near the heterointerface. A lower recombination energy, and therefore a smaller exciton binding energy, reflects a stronger electric field at the place where the exciton recombines. Therefore, the *H* band with lower energies can be identified as the emission of excitons closer to the heterointerface where the interface electric field is stronger.

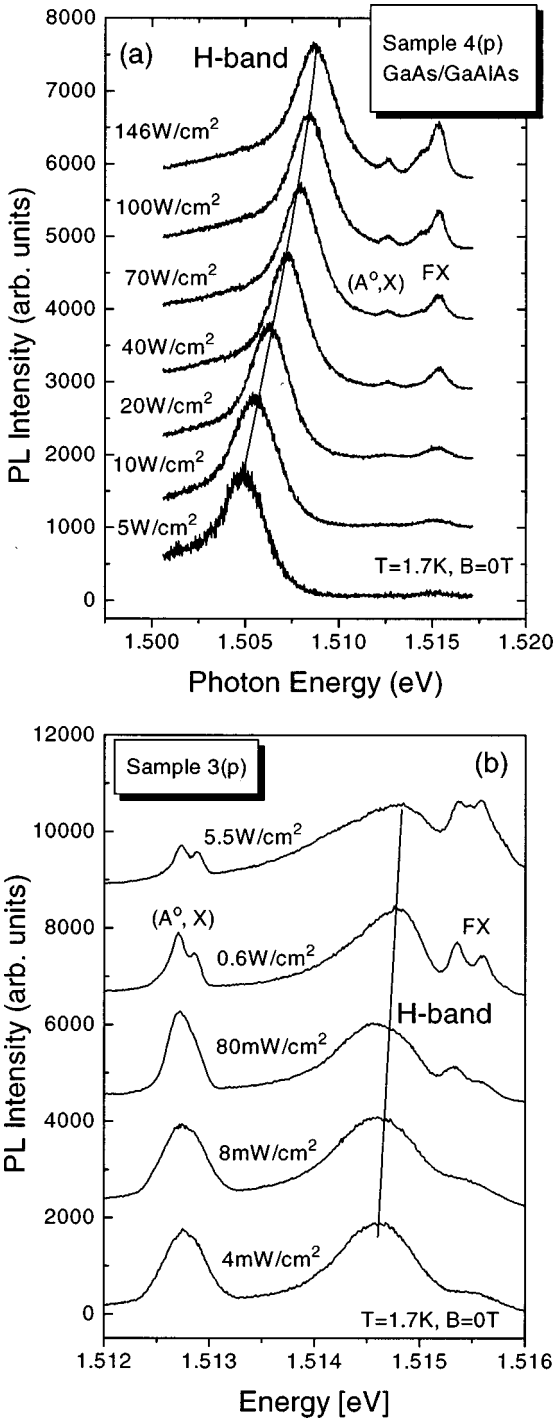


FIG. 1. (a) The *H* band in a *p*-type heterojunction, which lies energetically lower than the GaAs excitons. (b) The *H* band in a *p*-type heterojunction that overlaps with GaAs excitons. The common feature of these *H*-band emissions is their relative broad linewidth and their blueshift with increasing laser excitation density.

The above identification can be proved by the anisotropy of the diamagnetic shifts when the sample normal is rotated with respect to the magnetic field. We compare now the diamagnetic shifts in the Faraday and the Voigt configuration for the *H* bands at higher and lower energy. In the Faraday configuration the magnetic field is applied perpendicular to

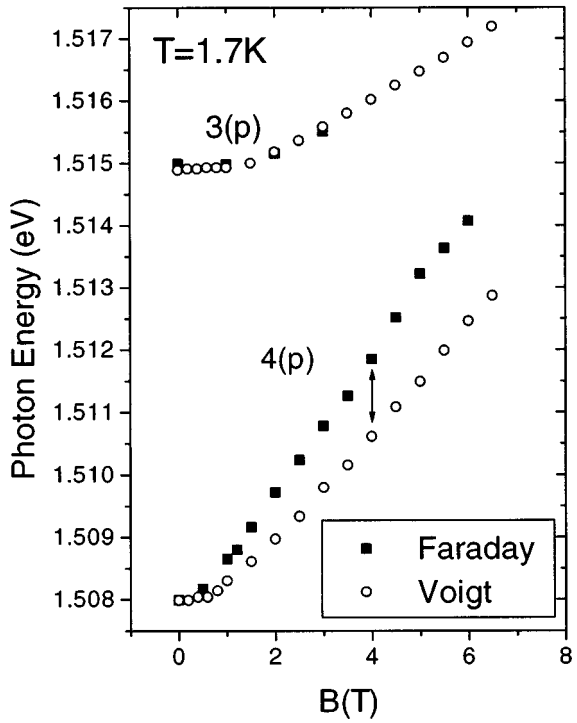


FIG. 2. Diamagnetic shift of the *H* band for samples 4 and 3 in the Faraday and Voigt configuration. The diamagnetic shift differs strongly for the two configurations in the case of the low-energy *H* band in sample 4 but not for the high-energy *H* band in sample 5.

the plane of the 2DCG, while in the Voigt configuration they are parallel. The low-energy *H* band for sample 4 shows a larger diamagnetic shift in the Faraday configuration than that in the Voigt configuration. While the diamagnetic shifts for the high-energy *H* band (sample 3) are almost the same for both configurations. This shows that the rotation symmetry is broken to a larger extent for the *H*-band excitons with lower emission energies. Therefore, these excitons are exposed to a stronger interface electric field. Instead, the rotation symmetry of the *H*-band excitons with higher emission energies is broken to a minor extent due to a weaker electric field.

We would like to emphasize that *H*-band excitons can be located so far away from the heterointerface that the binding energies of these excitons are hardly influenced by the interface electric field. Figure 3 compares the diamagnetic shift of the high-energy *H* band at 1.5145 eV with the diamagnetic shift of the GaAs excitons. The experiment shows that the diamagnetic shifts are almost the same. A similar energy shift for all types of excitons at low magnetic fields indicates a similar exciton binding energy for both the *H*-band excitons and the GaAs excitons. Hence, these *H*-band excitons should be located in an area where the interface electric field is extremely weak and very close to the flat band region in the GaAs.

Summarizing, the recombination energy and the binding energy of the *H*-band excitons are reduced by the interface electric field of different strengths. Their properties depend on their location in real space. The PL in heterojunctions, therefore, consists of standard GaAs excitons in the flat band region and the *H*-band excitons that are distorted by the interface electric field to different extents.

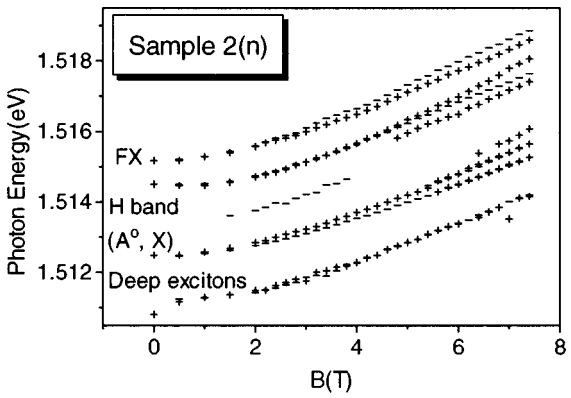


FIG. 3. Diamagnetic shift of excitonic transitions in an *n*-type heterojunction sample for σ^+ (denoted by $+$) and σ^- (denoted by $-$) polarizations.

IV. MICROWAVE INFLUENCED PL AND ODCR

Till now we have assigned all the transitions observed in the heterojunction samples to excitons not directly involving the 2DCG. However, we will demonstrate in the following that the PL intensities of all types of excitons are strongly influenced by the 2DCG. We will first demonstrate the vertical movement of the photoexcited carriers during their lifetime, driven by the interface electric field. Then we will show that these photoexcited carriers, which recombine later as the *H*-band excitons and the GaAs excitons, have been or are being influenced by the 2DCG. The effect of the 2DCG is reflected in the intensity of the exciton recombination. We have studied this mechanism by microwave-influenced PL and the ODCR experiments.

It is known that microwaves applied in addition to the laser excitation reduce the intensity of the exciton recombinations.[23] Therefore, if we assume that the total amount of photoexcited carriers remains unchanged, more free carriers are available in the material with microwave excitation. Typical microwave-induced PL intensity variations for the heterojunctions are shown in Fig. 4. We refer to the PL spectrum with microwave excitation as spectrum *A* and the PL spectrum without microwave excitation as spectrum *B*. Their difference, $A-B$, represents the microwave effect on the PL. For all heterojunction samples investigated, we always observed a PL intensity decrease for the *H* band and a PL intensity increase for the GaAs excitons if the laser and the microwave power were kept reasonably low, see Fig. 4. If the broad *H* band is overlapped with bound excitons, the differential spectrum shows a negative background with an upward peak at the bound exciton lines, see sample 5 in Fig. 4. For most samples, the integrated signal of the differential spectra vanishes. This demonstrates that the increase of the PL intensity of the GaAs excitons occurs at the expense of the *H*-band excitons. This important experimental fact demonstrates clearly an internal correlation between the recombinations of the *H*-band excitons and the GaAs excitons. As discussed in the previous section, the *H*-band excitons are situated closer to the heterointerface than the GaAs excitons. Under microwave excitation, a spatial redistribution of the photoexcited carriers occurs. The experiment shows that the extra free carriers due to the reduction of the recombination

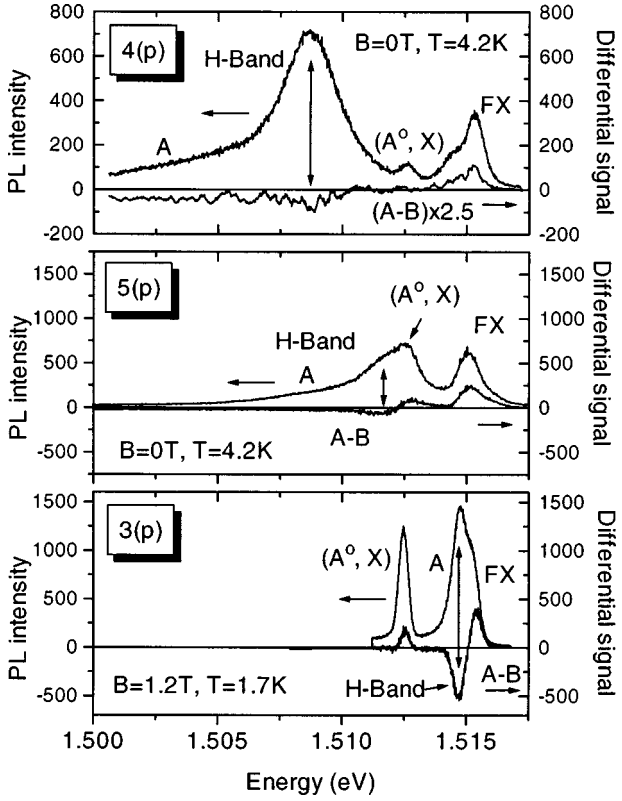


FIG. 4. The PL intensity with (A) microwaves and the differential spectra ($A-B$) by chopping the microwaves of sample 4 (upper), 5 (middle), and 3 (lower). The spectra of sample 3 are taken at the resonant magnetic field to enhance the microwave influence. The H band always exhibits a negative PL intensity change while the GaAs excitons show a PL intensity increase with microwaves.

rate of the H-band excitons are now transferred to the flat band region to form GaAs excitons.

Now we should point out the large conceptual difference between the PL of common bulk or epilayer samples and the PL of heterojunction samples. In bulk or epilayer samples, the carriers lose their phase coherence and relax to the band edge after the laser excitation. These carriers will barely move in real space. However, this is not the case in heterojunction samples. In this case, a giant spatial drift of the photoexcited carriers occurs during their lifetime, as demonstrated by the experiment in Fig. 4. Due to this drift, the carriers that finally recombine far from the heterointerface as the H-band or the GaAs excitons were created in an area very close to the 2DCG where they could interact with the 2DCG. The cyclotron resonance of the 2DCG, shown in Fig. 5, detected on both the H-band and the GaAs excitons is a good demonstration of this interaction.

The resonance occurs in a magnetic field of 1.24 T at a microwave frequency of 70 GHz for the p-type sample in Fig. 5. In this sample, the H band is mixed with both the (A°,X) and (D°,X) transitions. By applying microwaves, a broad negative differential signal is observed for the H band while the signal for the FX of the GaAs remains always positive. Throughout the negative background for the H-band differential signal, upward peaks can always be observed at (A°,X) transitions, demonstrating an increase of (A°,X) recombinations. Again the integration of each differential spectrum equals zero, which reflects the vertical transport and spatial redistribution of the photoexcited carriers. If the magnetic field is swept and the spectrometer is adjusted to follow the maximum of the PL intensity of the FX, a continuous positive ODCR signal is recorded, Fig. 5(b). The cyclotron resonance can be well attributed to the two-dimensionally confined holes although it is detected on the FX of GaAs. If it were the hole cyclotron resonance in the GaAs flat band region, then a more obvious ODCR signal of the electron cyclotron resonance should be observed due to their higher mobility. The electron cyclotron resonance is not observed in this sample. Hence the cyclotron resonance in our experiment represents the resonance of the 2DCG although neither the H-band excitons nor the GaAs excitons directly involve the 2DCG. The above conclusions appear to be contradictory, however, they reflect the nature of the vertical transport of the photoexcited carriers in the vicinity of the 2DCG: The carriers recombine near or in the flat band region, but they interacted with the 2DCG when they were created near the heterointerface. From this special property, we were able to extract the effective mass values of the two-

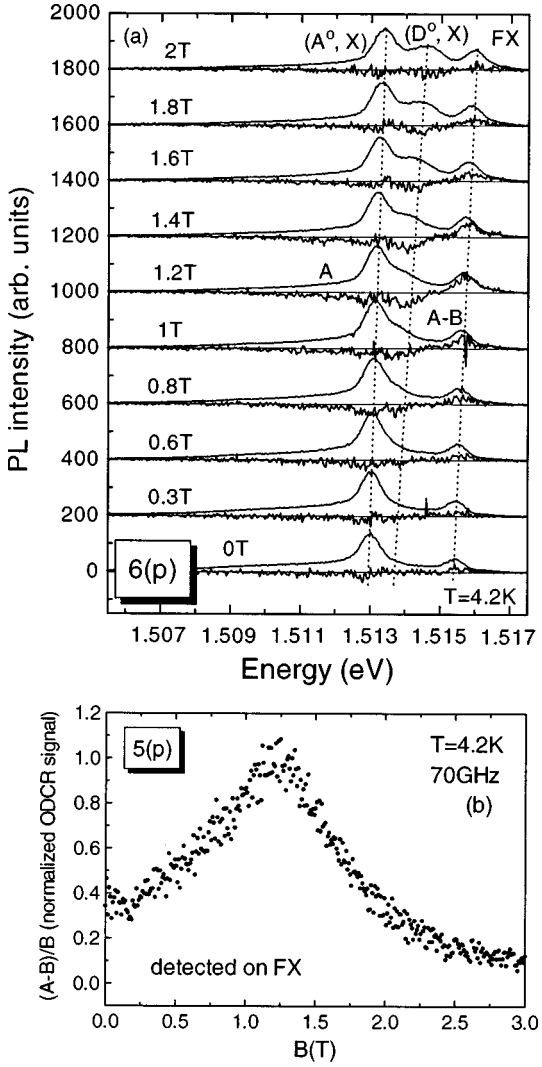


FIG. 5. (a) Differential PL spectra with microwave on and off at different magnetic fields. A resonant intensity decrease for the H band and a resonant intensity increase for the GaAs excitons are observed. (b) Normalized ODCR signal $(A-B)/B$ detected on the peak of the H band and the GaAs exciton transitions.

dimensional electrons in *n*-type heterojunctions and the two-dimensional holes in *p*-type heterojunctions. These values are listed in Table I.

Up to this point we have provided evidence for the vertical transport of photoexcited carriers that interact with the 2DCG. In order to cover the topic more generally, we consider more complex experimental results. In the case of a few high-quality samples only a negative differential PL signal has been observed, see Fig. 6(a) for the ODCR signal with low microwave attenuation in dB. But this is no proof for the absence of a vertical transport of the photoexcited carriers or the lack of a PL intensity correlation between the *H*-band excitons and the GaAs excitons. As a matter of fact, when we reduced both the laser intensity and the microwave power, the negative differential signal for the *H* band and the positive differential signal for the GaAs excitons were still observed [Fig. 6(a), ODCR signals with high microwave attenuation]. To understand this phenomenon, we need to compare the corresponding PL spectra for various microwave powers in Fig. 6(b). The experiment shows that the PL intensity of the *H* band quenches easily with the microwave excitation. When the *H* band dominates the PL spectrum and when the spectrally integrated PL intensity is not reduced [Fig. 6(b), the curves with attenuation of 18–9 dB], an increase of the microwave power reduces the *H*-band PL intensity and transfers it to the GaAs excitons. Under this condition, an enhanced PL intensity of the GaAs excitons has been observed, see Fig. 6(a). If the *H*-band emission already disappears at high microwave powers [Fig. 6(b), attenuation of 6–0 dB curves], a further increase of the microwave power reduces the spectrally integrated PL intensity. Therefore only a negative differential PL signal has been observed for the excitons of the GaAs layer, see Fig. 6(a).

For the samples like that of Fig. 6, the ODCR signal recorded with increasing magnetic field is very complicated to analyze. The recorded curves are displayed in Fig. 7. The ODCR signal detected on the *H* band with a microwave attenuation of 24 dB resonantly reduces at the magnetic field of 0.19 T (peak 1 indicated in Fig. 7) corresponding to an effective mass of $0.078m_0$. Compared with the effective mass of $0.0665m_0$ for the bulk GaAs, this effective mass is large and can be attributed to the quantum confinement and the non-parabolicity of the conduction band. The strongest resonant peak of the 0-dB curve detected on the (A°,X) (peak 2 indicated in Fig. 7) corresponds to an effective mass of $0.065m_0$. It may be attributed to three-dimensional electrons or less confined electrons. Further features shown in Fig. 7 are not well understood and can be partly attributed to the complexity of the PL intensity variation described for Fig. 6. A more detailed discussion over these results will be given in the next section.

V. VERTICAL TRANSPORT MODEL OF THE PHOTOEXCITED CARRIERS

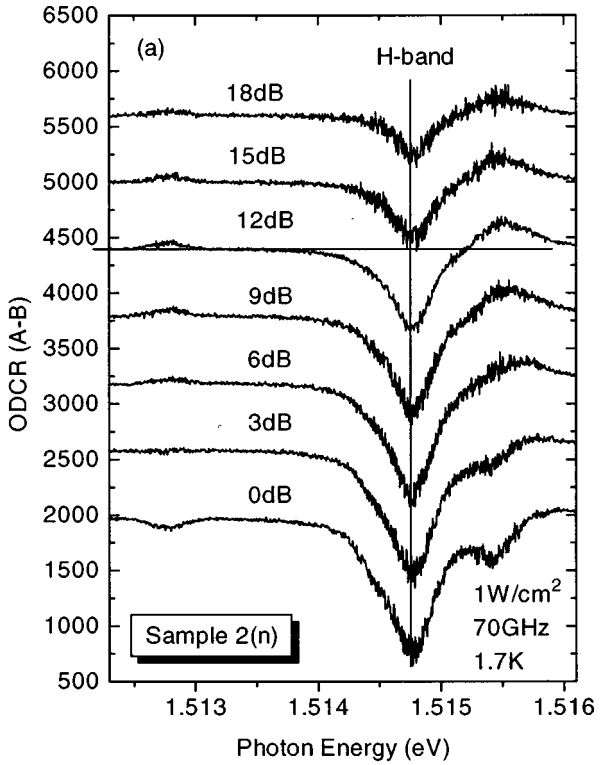
Now we begin to construct our model of the vertical transport of photoexcited carriers from the analysis of the experimental results discussed so far. The model will be demonstrated for an *n*-type heterojunction. It can be similarly applied for *p*-type samples.

In PL experiments, the laser excites electrons from the valence band to the conduction band within a certain penetration depth. In heterojunction samples, the photoexcited holes will drift away from the interface and the photoexcited electrons will drift towards the interface driven by the interface electric field, see Fig. 8. Because the interface electric field in the vicinity of the heterojunction reaches values up to 50 kV/cm,[10] most of the photoexcited holes drift to a certain distance away from the heterointerface before they are captured by the photoexcited electrons to form excitons. A simple estimation with a hole mobility of 500 $cm^2/V\,s$ and a collision time of 10^{-13} s gives a mean free path of 250 Å for photoexcited holes at 50 kV/cm. The effective notch width is typically about 50 Å. Within this distance there are no holes available for the recombination with the 2DCG. In fact, this region is depleted of holes by the interface electric field. This explains why no band-band transitions can be observed in modulation doped heterjunctions.

The holes drifting vertically may capture electrons that drift towards the interface and form excitons. These excitons are distorted by the interface electric field and become space indirect. The space indirect excitons are not accelerated by the interface field because they are neutral in charge. But their recombination energy, as well as their binding energy, is reduced by the interface electric field according to the QCSE.[9] The recombination of these excitons contributes to the broad *H* band shown in Fig. 1. The reduction of the binding energy due to a larger spatial separation of electron and hole increases the photon energy emitted by the *H*-band excitons. However, in addition to this, a large reduction of the potential acting on electrons and holes takes place. This leads to a strong actual reduction of the transition energy. This concept explains why the transition peak of the *H*-band is always energetically lower than that of the GaAs free excitons.

The exciton binding energy and the emitted photon energy of the *H* band depend on the location of the *H*-band excitons. When they are located closer to the interface, where the electric field is stronger, the binding energy of the *H*-band excitons becomes smaller and the photon energy emitted is also smaller. How far the *H*-band excitons will be located from the interface depends on the vertical mobility of the photoexcited carriers and the strength of the interface electric field. With a stronger interface electric field or a higher carrier mobility, the final distance of the *H*-band excitons from the interface is larger and the final electric field which affects the *H*-band excitons is smaller. This explains the different energy positions of the *H*-band excitons for different samples, as shown in Fig. 1, and also their sensitivity to other sample parameters like mobility or Al composition, as well as experimental parameters such as temperature and laser excitation power, etc., as discussed previously.

If the laser excitation power is increased, the interface electric field or the notch potential is flattened by the Coulomb force between the photoexcited carriers and the 2DCG. The reduction of the interface electric field results in an increase of the recombination energy and the binding energy of the *H*-band excitons. The energy increase depends on the location of the *H*-band excitons in real space. When they are located closer to the notch, where the interface electric field is initially stronger, the electric field is more obviously reduced with increasing laser excitation power. Thus a larger



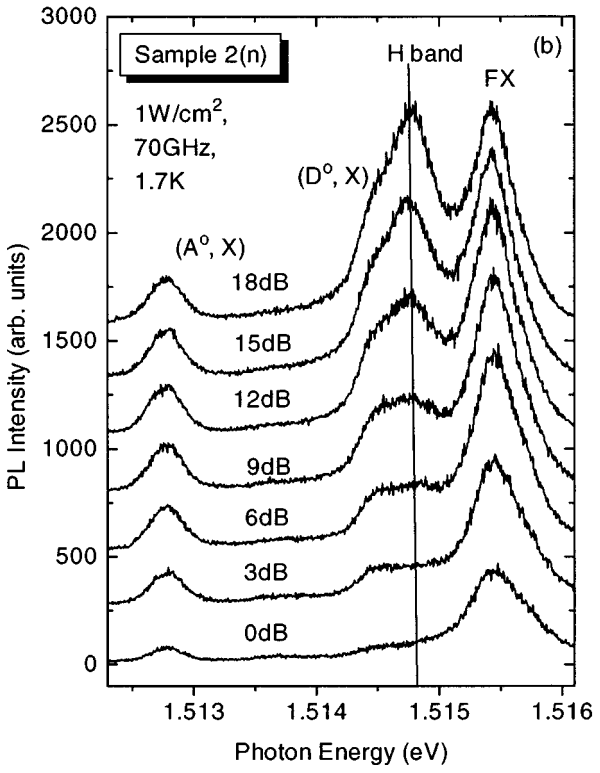


FIG. 6. (a) Differential spectra of sample 2 for different attenuations of the microwave power. (b) PL spectra for different attenuation of the microwave power. For small microwave powers, the *H*-band PL intensity quenches and the differential signal is positive for GaAs excitons. At big microwave powers the *H* band disappears. A further increase of the microwave power only quenches the PL intensity for all transitions.

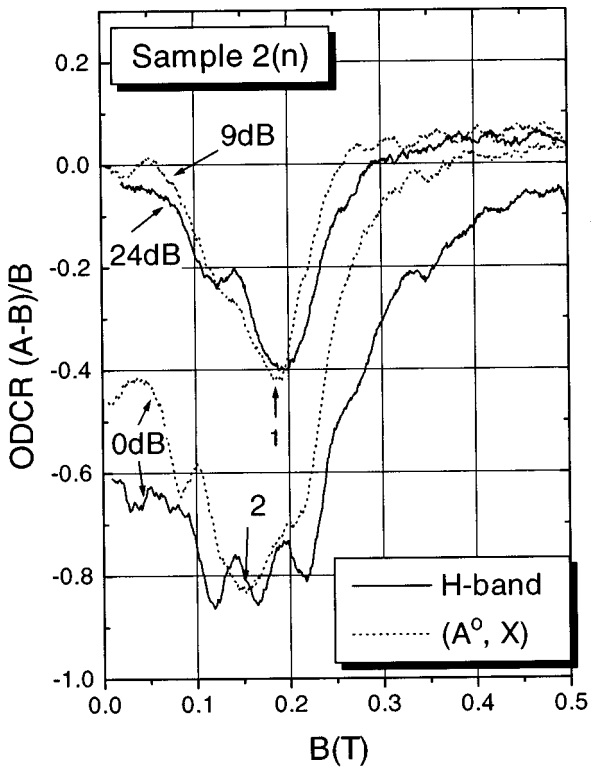


FIG. 7. ODCR detected on the *H* band and on the acceptor-bound excitons for different microwave powers. At low microwave powers, the ODCR exhibits CR for the 2DCG. At higher microwave powers, the ODCR curves become extremely complex.

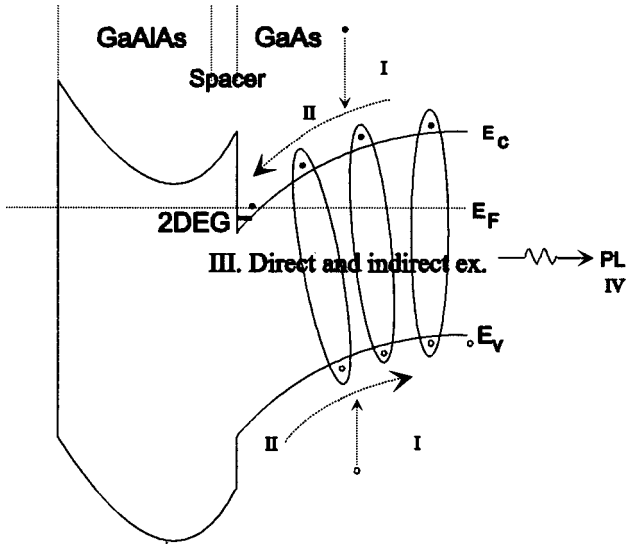


FIG. 8. Dynamics for the photoexcited carriers in an *n*-type heterojunction. Four steps are involved in the PL process. They are (I) carrier relaxation, (II) vertical transport, (III) exciton formation, and (IV) exciton recombinations. The excitons in the flat band region are not influenced by the interface electric field and will emit excitons of GaAs. The *H*-band excitons are subjected to the interface electric field of different strength and, therefore, their recombination energies are reduced to different extent. It is attributed to the breadth of the *H* band.

blueshift is observed in Fig. 1(a). Contrary to this, if the excitons are located far away from the notch where the interface electric field is initially weak, the potential variation is also small and only slight blueshifts can be detected, see Fig. 1(b).

When the *H*-band excitons are located closer to the heterointerface, the exciton binding energy is smaller, so a larger diamagnetic shift can be expected. While for the *H*-band excitons located near the flat band region, the exciton binding energy is larger and a smaller diamagnetic shift is observed in Fig. 2. If the *H*-band excitons are subjected to extremely weak electric fields, they have a similar exciton binding energy like those excitons in the flat band region. These *H*-band excitons exhibit a similar diamagnetic shift like the GaAs excitons shown in Fig. 3.

Due to the drastic decrease of the interface electric field in the sample normal direction, the rotational symmetry of the *H*-band excitons at different locations is broken to a different extent. When the photoexcited carriers drift to a certain distance from the heterointerface, the final *H*-band excitons are subjected only to a weak electric field. Their diamagnetic shift is almost space isotropic, as observed for the sample 3 (Fig. 2). The opposite occurs if the vertical transport is weak and the *H*-band excitons are situated closer to the heterointerface. These excitons feel a much stronger interface electric field and the anisotropy of the diamagnetic shift becomes stronger, see sample 4 in Fig. 2.

The photoexcited carriers that form excitons far away from the heterointerface in the GaAs flat band region may have been created already in this region by the exciting laser light. But they also may have been originally created closer to the heterointerface and vertically drifted to the flat band region. Nevertheless, as the finally experienced electric field at the location of recombination is negligibly small, the exciton recombination energy and the exciton binding energy are not reduced. These are space direct excitons and they emit photons with energies typical for GaAs bulk or epilayer materials, see Fig. 1.

The application of microwaves probes the dynamic process of the vertical transport. According to Romestain and Weisbuch,[23] the exciton emission intensity is reduced by the microwave excitation. If we assume that the total amount of photoexcited carriers remains constant, the reduction of the exciton emission intensity should be balanced by an increased density of free carriers. In the *n*-type structure shown in Fig. 8, the additional photoexcited holes, freed by the microwave excitation, drift to the GaAs flat band region and recombine with the free electrons in that area. The existence of the free electrons can be confirmed by the relatively long lifetime of the electron-acceptor transitions. Therefore the reduction of the *H*-band PL intensity is balanced by the increase of the PL intensity on the exciton lines in the GaAs flat band region with microwave excitation, shown in Fig. 4.

Modulation doping increases the carrier mobility of the 2DCG. While the carriers near or in the flat band region are believed to have a much smaller mobility. Thus, the microwave energy should be absorbed mostly by the 2DCG and then transferred to the area near the *H*-band excitons or the excitons in the GaAs flat band region by thermal conduction. According to this argument, the effective carrier temperature will be less enhanced as the distance to the interface increases. This argument is reasonable since we do observe experimentally that for most samples only the PL intensity for the *H*-band decreases, which means that only the carriers for the *H*-band excitons are heated through the thermal conductance (Fig. 4). The PL intensity of the GaAs excitons increases, demonstrating that the effect of thermal conduction for these excitons is less important than the carrier vertical transport. For most samples, the energetically integrated intensity of the differential spectra equals zero, which indicates a huge temperature difference between the carriers for the *H*-band excitons and for the GaAs excitons, see Figs. 4 and 5. By sweeping the magnetic field, the ODCR resonant signal is negative for the *H*-band and is positive for the excitons in the GaAs flat band region for these samples.

However, for samples where the thermal conductance leads to a similar temperature increase for both the carriers of the *H*-band excitons and the carriers for the GaAs excitons, a negative ODCR signal of the 2DCG can be observed for both types of excitons (Fig. 7). But due to the larger distance from the heterointerface, the excitons in the flat band region require more microwave power than the *H*-band excitons in order to reach a similar ODCR intensity of the 2DCG. Experimentally, to obtain the same amount of ODCR signal on the GaAs excitons as on the *H* band we must use a 15 dB lower attenuation of the microwave power (compare the 9-dB curve on the GaAs excitons and the 24-dB curve on the *H*-band excitons in Fig. 7).

Furthermore, the carriers which were initially photoexcited in the GaAs flat band region are also heated by the microwaves directly. This intrinsic process can reduce the PL intensity of the GaAs excitons and contribute to the ODCR signal of bulk GaAs.

The ODCR signal for the excitons in the GaAs flat band region may come from three possible origins: first, the photoexcited carriers of these excitons may be heated directly by the microwave absorption. The exciton formation probability is reduced by the increased carrier temperature, which induces a negative ODCR signal of the bulk electron cyclotron resonance. Secondly, the photoexcited carriers of these excitons may be indirectly heated by the microwaves through the thermal conductance to the 2DCG, yielding a negative cyclotron resonance signal of the 2DCG (Fig. 7, the 9-dB curve). Thirdly, the density of photoexcited carriers may also increase due to the reduction of the *H*-band PL intensity and the vertical transport of these photoexcited carriers. This results in a positive ODCR signal of the 2DCG (Fig. 5). The experimental ODCR signal on the exciton transitions in the GaAs flat band region is a combination of these three effects and therefore the observed ODCR curves are extremely complicated to analyze (Fig. 7).

We now end our discussion on the experimental observations presented so far. To summarize our model, the transitions in the heterojunction are excitonic. But they still reflect the properties of the 2DCG through the carrier vertical transport. In the following, we will present more experimental results which corroborate the special properties in heterojunctions presented so far.

VI. TIME-RESOLVED PL

The time-resolved PL is known for revealing dynamic properties of excitons and photoexcited carriers. The pub-

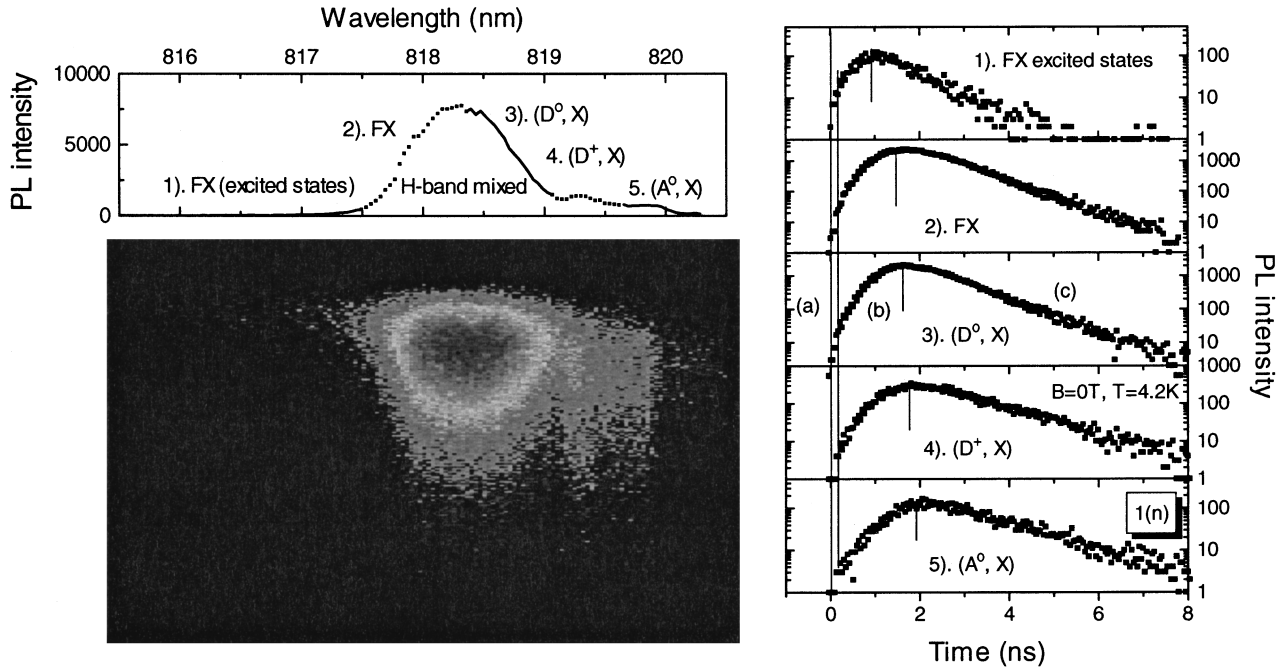


FIG. 9. On the lower left is the three-dimensional image of the time-resolved PL spectra of sample 1 at 4.2 K. The horizontal direction represents the wavelength and vertical direction is the time evolution. Excitation wavelength of the laser is 3950 Å. The upper left part is the time-integrated PL spectrum. Five excitonic transitions are identified according to the three-dimensional image and the energy references in the literature (Ref. 15). The right part is the time evolution of the energy-integrated PL intensity for five types of excitons. The PL intensity rise of all five types of excitons can be split into two stages with different rise times.

lished time-resolved PL on heterojunctions deals only with the extremely long lifetime of the *H*-band transitions.[7,21,22] We will report, to our knowledge for the first time, the extraordinary long rise time of the PL intensity for excitonic transitions in heterojunctions.

The long lifetime of the low-energy H band has been reproduced in our experiment. We found that if the energy position of the *H*-band emission increases, its lifetime becomes shorter and comparable to those of excitons in the GaAs flat band region. Similar results have been reported by Bergman *et al.*[21] A three-dimensional image of the time-resolved PL spectrum for the excitonic transitions of sample 1 is shown in Fig. 9. The horizontal axis represents the wavelength. It increases from left to right. The time develops from the upper to the lower direction. The time integrated spectrum is plotted on the upper side of the three-dimensional image. The time evolution of the PL intensity, energetically integrated for (1) the excited states of FX, (2) FX, (3) (D^0,X), (4) (D^+,X), and (5) (A^0,X) excitons is plotted on the right-hand side of the three-dimensional image. We found that the lifetime (*c*) of the excitonic transitions is typical for GaAs epilayers. However, the rise time is extraordinarily long. The PL rise can be separated into two time steps. A part (*a*) with quicker rise time is commonly observed with a time constant of several picoseconds. The part (*b*) has a much slower rise time, almost 1–2 ns. It cannot be explained by the exciton relaxation process, as this long rise time is also observed for the transitions of the free excitons and excited states of free excitons (Fig. 9). It can also not be explained by the carrier relaxation from the $Ga_{1-x}Al_xAs$ barrier to the GaAs, because this long rise time is still observed when we excite below the barrier. Within our model of the vertical transport of the photoexcited carriers, we identify the first part of the PL intensity rise (*a*) as the intrinsic formation of excitons by photoexcited carriers locally excited by the laser. The slow rise (*b*) is identified as the excitons formed by the photoexcited holes that are created near the interface, but vertically transported to an area near the flat band region. These holes recombine with free electrons in that area. Since the interface electric field decreases drastically after typically 100 Å, the vertical transport time can be much longer than the exciton formation time. This transport time for the photoexcited carriers is responsible for the PL intensity rise time of (*b*). The relative intensity ratio between the first rise of (*a*) due to the intrinsic exciton formation and the second rise (*b*) due to the vertical transport process depends on the amount of the vertically transported holes. The excitation intensity has been varied to change the density of holes reaching the flat band region in Fig. 10. For both (D^0,X) mixed with the *H* band and (A^0,X), the intrinsic part (*a*) is dominant when the laser excitation power is low. This can be understood as follows: if the excitation power is low, the total amount of holes accumulated in the flat band region is small. The chance for these vertically transported holes to capture free electrons in that area is small. With increasing laser excitation power, more photoexcited holes are generated and drift to the flat band region. Consequently the probability for exciton formation becomes higher. In this case, the second rise (*b*) becomes dominant due to the vertical transport contribution for the PL rise time. This nonlinear power dependence of the PL intensity is consistent with the experimental results in Fig. 1 where, with increasing laser excitation power, the relative PL intensity of the GaAs excitons increases more strongly than that of the *H*-band excitons. Thus, we have confirmed the vertical transport of the photoexcited carriers

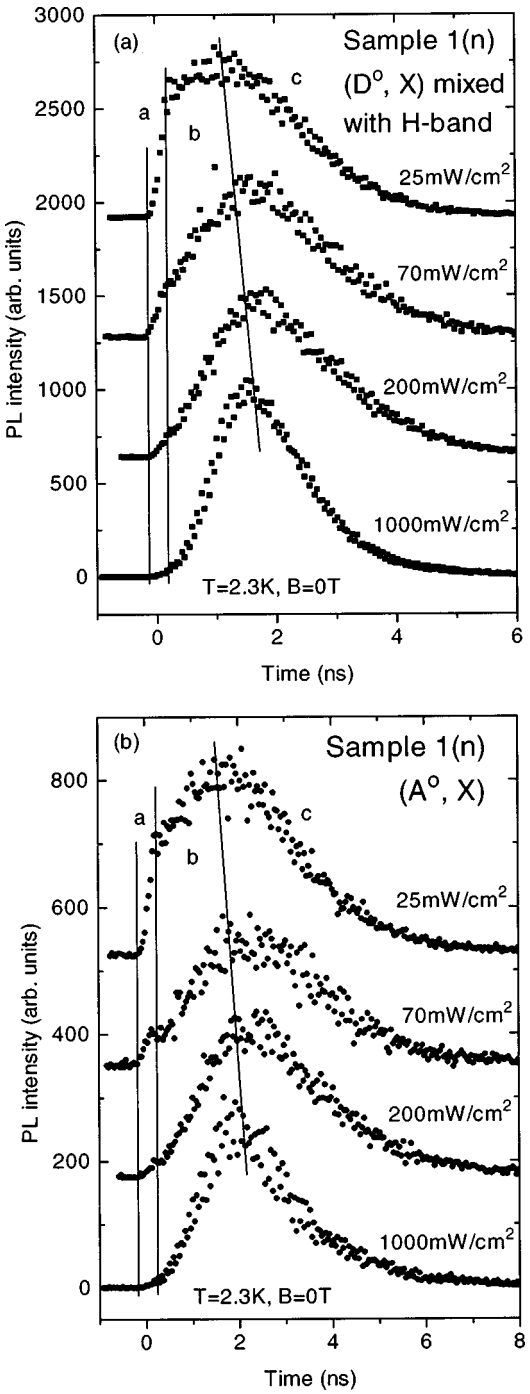


FIG. 10. The laser excitation density dependence of the two-step PL rise time on (a) the neutral donor-bound excitons (D^0,X) overlapped by the H band, and (b) the neutral acceptor-bound excitons (A^0,X).

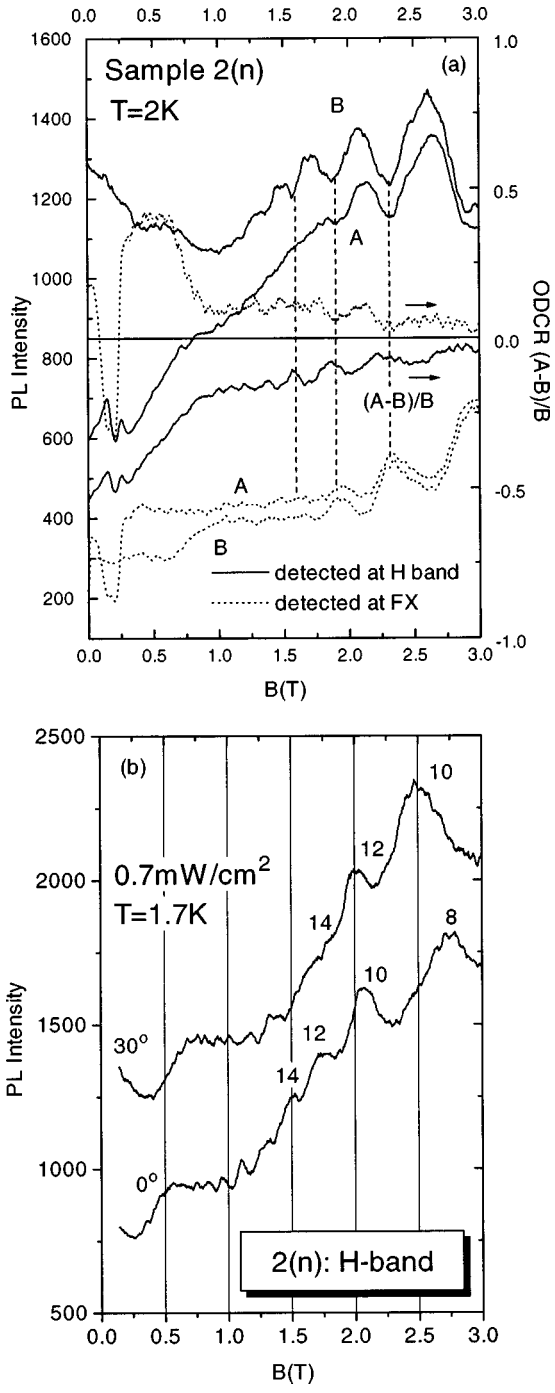


FIG. 11. (a) PL intensity oscillations with (A) and without (B) microwaves and the change of the PL intensity $(A-B)/B$ for the H band and the GaAs free excitons. The dotted lines are guidelines for peak comparison. (b) Two-dimensional properties of the ODSdH effect. It shows that the ODSdH effect reflects the oscillatory properties of the carriers confined in the notch.

for exciton recombinations in the heterojunctions by means of time-resolved spectroscopies.

VII. OSCILLATIONS OF THE PHOTOLUMINESCENCE INTENSITY

Now we will focus on the interactions between the photoexcited carriers and the 2DCG, shown by their effects on the exciton recombination in the heterojunctions. The PL intensity of a heterojunction was recorded as a function of magnetic field when the spectrometer was adjusted to follow the maximum peak positions. Typical traces for exciton recombinations, both with and without microwave excitation, are shown in Fig. 11(a). The PL intensity of both, the H band and the free-exciton lines, oscillates with the magnetic fields. The maxima of the oscillation are periodic with the inverse of the magnetic-field strength. The experimental technique and the observed curves are similar to those of the Shubnikov–de Haas (SdH) effect and will be referred to as

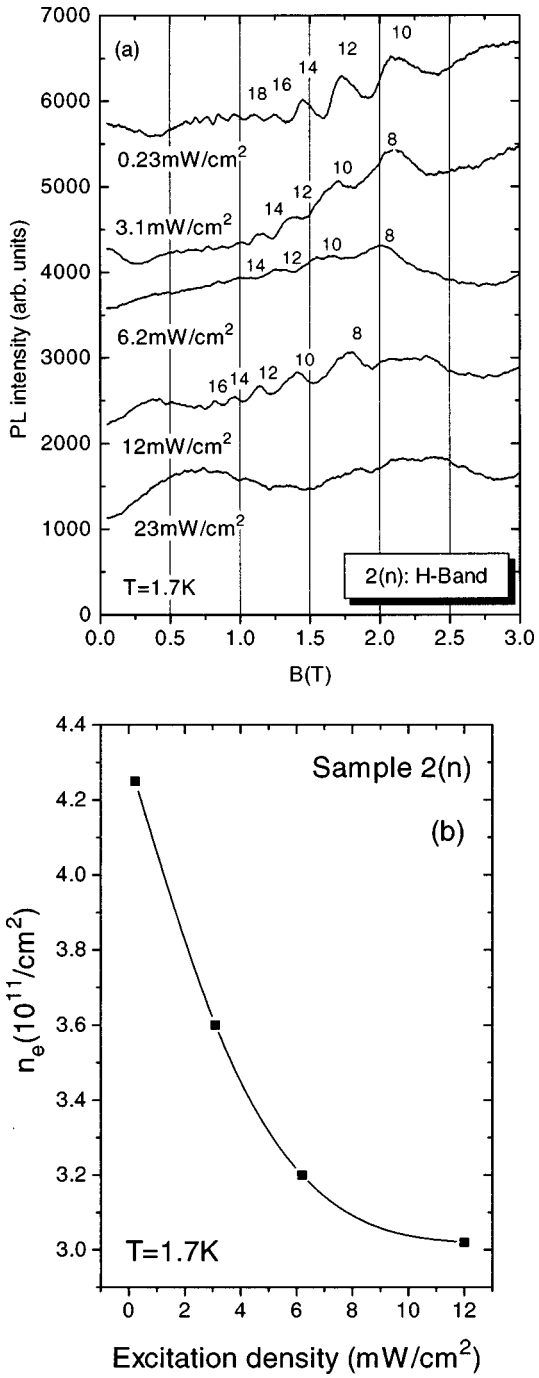


FIG. 12. (a) ODSdH effect at different laser excitation densities. The oscillation period of $\Delta(1/B)$ becomes larger for higher laser intensities. (b) The concentration of electrons confined in the notch as a function of the laser excitation intensity.

optically detected SdH (ODSdH) oscillations in the following.

The oscillation of the PL intensity displayed in Fig. 11(a) is two dimensional although it is detected on the *H*-band excitons or three-dimensional excitons of the GaAs. To verify the two-dimensional character, we rotated the sample normal 30° with respect to the external magnetic field. The ODSdH peaks shift to higher magnetic fields, shown in Fig. 11(b). The peak shifts satisfy the relation of $B(\perp) = B(30°)\cos(30°)$. Therefore the ODSdH effect detected on the *H*-band and the GaAs bound exciton lines represents the properties of the 2DCG confined in the notch. The above experimental result reveals the influence of the 2DCG on the carrier vertical transport more exactly.

It is well known that the in-plane transport of the 2DCG is determined by the electron density at the Fermi level. In magnetic fields, the density of states splits into broadened Landau levels. By sweeping the magnetic field, the density of states at the Fermi level oscillates, which induces the oscillation of the magnetoresistance and the Hall coefficient. In a two-dimensional system, the Hall resistance is quantized with a low dissipation current flowing in the plane of the 2DCG when the Fermi level lies between two Landau levels. By comparing the oscillating PL intensity and the oscillating differential PL intensity under microwave excitation, we found that the vertical transport of the photoexcited carriers is similarly modulated as the in-plane transport.

The relative change of the PL intensity caused by the microwave excitation, denoted as $(A-B)/B$, is plotted in Fig. 11(a). It reaches a minimum when the *H*-band PL intensity is at a minimum. The relative change of the PL intensity represents the in-plane conductivity of the sample in our microwave measurement. Hence, the *H* band has the smallest PL intensity when the in-plane conductivity is lowest. Therefore, when the Fermi level lies between two Landau levels, the vertical transport becomes more favored for the photoexcited carriers, similar to the carrier in-plane transport. Under this condition, most of the photoexcited carriers can escape and reach the flat band region. This reduces the *H*-band PL intensity to its minimum. The PL intensities of the (A^0,X) and the FX oscillate with opposite phase to that of the *H* band due to the PL intensity correlation discussed so far. For comparison we also plot in Fig. 11 the oscillation of the PL intensity detected on the FX. The antiphase oscillation and its relation to the SdH effect are similar to the results reported by Turberfield *et al.*[3] However, we emphasize that these oscillations are detected on transitions not involving the 2DCG. The PL intensity oscillation with microwaves disappears at low magnetic fields due to the extremely strong heating of the 2DCG. Cyclotron resonances are observed with complicated patterns in low magnetic fields as discussed before.

The oscillatory period of the ODSdH reflects the carrier concentration in the notch. We have recorded ODSdH oscillations under different laser excitation intensities, shown in Fig. 12. A peak shift, and therefore a change of the oscillatory period, is observed. The oscillatory period $\Delta(1/B)$ becomes smaller with increasing laser intensity, indicating a decrease of the electron concentration and thus a flattening of the space charge potential in the notch. With stronger laser excitation, more photoexcited holes drift into the GaAs flat band region and the electric field created between the 2DCG and photoexcited holes becomes stronger. It reduces the original interface electric field and flattens the space charge potential. The flattened space charge potential raises the subband energy and reduces the electron concentration in the notch. It raises also the photon energy emitted by the *H*-band excitons. So the reduction of the electron concentration is consistent with the blueshift of the *H*-band transition with increasing laser intensity (Fig. 1).

VIII. CONCLUSION

We have studied the photoluminescence of $GaAs/Ga_{1-x}Al_xAs$ heterojunctions. We discovered that the transitions are excitonic and therefore do not directly involve the 2DCG. However, we have demonstrated that prior to exciton formation, the vertical transport of the photoexcited carriers under the interface electric field is influenced by the 2DCG. Due to this vertical transport, the PL intensity of the *H*-band excitons and that of the excitons in the GaAs flat band region are correlated and indirectly reflect the properties of the 2DCG. The interaction between the photoexcited carriers and the 2DCG is responsible for the two-dimensional character of the ODCR and the PL intensity oscillations with magnetic field in heterojunctions. The vertical transport is also responsible for the extremely long rise time of the PL intensity for all observed excitonic transitions.

ACKNOWLEDGMENTS

The authors would like to thank Dr. H. Schäfer for the help in constructing the ODCR measurement system. Dr. E. Kurtz and Dr. R. Pittini are gratefully acknowledged for their careful reading of the entire manuscript as well as for the intensive discussions concerning the experimental results. The work carried out in Würzburg was funded by the Deutsche Forschungsgemeinschaft through Os 98/5.

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