Exciton dynamics in GaAs/$Ga_{1-x}Al_xAs$ heterojunctions and GaAs epilayers

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We present a study of the exciton dynamics in modulation doped GaAs/$Ga_{1-x}Al_xAs$ heterojunctions and GaAs epilayers. The comparison permits to identify the features characteristic for the heterojunctions. In particular, we analyze the rise time of the transient photoluminescence (PL) intensity. In general, we find a longer PL rise time for the lower-energy excitons indicating that the time required for the energy relaxation process increases with increasing binding energy of the excitons. Moreover, the rise time of the free excitons turns out to be conspicuously longer in heterojunctions than in epilayers although the time integrated PL spectra of the two systems are similar. From our analysis we conclude that the long rise time observed in the heterojunctions is closely connected with the vertical drift of the photoexcited carriers driven by the interface potential. In fact, we find that the carriers (i.e., holes in *n*-doped samples) photoexcited in the heterojunctions drift vertically from the heterointerface to the flat band region, where they finally recombine after forming excitons (bimolecular formation) with oppositely charged and locally excited carriers (i.e., electrons in *n*-doped samples).

I. INTRODUCTION

The exciton recombination process occurs in a time scale of a few hundred picoseconds to nanoseconds. This time domain is easily accessible to time-resolved photoluminescence (PL) experiments and therefore many studies of the exciton recombination process have already been carried out.[1] Instead, the much faster processes of energy relaxation of the carriers and exciton formation have been studied only recently and in only few PL experiments. In one of these new studies,[2] assisted by the recent development of the upconversion technique, the fine difference between the dephasing time of coherent excitons and the momentum scattering could be distinguished in the PL signal. In general, it is believed that the processes of energy relaxation and exciton formation should occur within 20 ps.[3] There are though a few reports of exceptionally long rise times well over 20 ps attributed to the momentum relaxation of excitons with large wave vectors towards the radiative zone around $K \approx 0$.[4] In quantum wells, PL rise times as long as 400 ps have been reported.[3,6] Furthermore, a rise time as long as 1 ns has been surprisingly reported for GaAs epilayers,[5] although this latter result could not be confirmed by our study.

In contrast, the dynamics of the excitons in heterojunctions is still unclear. Even the assignment of the peaks observed in the PL spectra of heterojunctions is controversial, except for the samples δ doped close to the interface.[7] The fact that the magneto-PL intensity in heterojunctions shows oscillations with the field similarly to the in-plane magnetoresistance[8] leaded to the incorrect interpretation that the observed PL signal arises from recombinations of the two-dimensional electron gas (2DEG). With this assignment of the PL signal, it had been difficult to explain why the PL peaks are located at the energies typical for the recombination of the excitons of the flat band region. This is not possible because the quantum confined stark effect in the interface electric field shifts the recombination energy. Moreover, we point out that discontinuities and oscillations on the energy scale observed in an energy-field contour plot of the magneto-PL intensity has still not found a reliable explanation.[9] Recently, we have shown that the PL signal in heterojunctions arises not from the 2DEG but from carriers photoexcited near the heterointerface and recombining in the flat band region after a vertical transport process.[10] The driving force for the vertical transport of the carriers is the self-built interface electric field arising in modulation doped heterojunctions and directly coupled to the 2DEG. This interface field drives the different charges of the photoexcited carriers to the two opposite directions, with the electrons (holes) drifting towards the heterointerface and the holes (electrons) towards the flat band region in *n*-type (*p*-type) samples. After reaching the flat band region, the holes capture free electrons excited there, forming excitons (bimolecular formation). The existence of long living free electrons in the flat band region has been confirmed by the observation of a long decay time for the electron-acceptor transitions and also by recently the nongeminate bimolecular formation of excitons.[11,12] According to the vertical transport model,[10] the carriers first drift along the interface electric field under the influence of the screening by the 2DEG, and finally reach the flat band region where they form excitons and recombine. Therefore, one understands why the PL signal shows oscillations typical for the 2DEG although the recombination does not involve the 2DEG directly. Our identification of the final location of the recombining carriers is consistent with the analysis presented in Ref. 13 although we do not need to involve acoustic phonon emission in our interpretation. In our previous work,[14] we have compared the

magneto-PL spectra of a heterojunction with the spectra of a quantum well sample in which the recombinations of the 2DEG are observed in the PL signal. The magnetic field dependence of the PL peaks in these two samples is totally different,[14] although the host material for the 2DEG is GaAs for both samples. This demonstrates that the optical transitions connected to the PL signal observed in quantum wells and in heterojunctions have a different origin. The key point in our interpretation is the vertical transport of the photoexcited carriers from the interface to the flat band region before they form excitons and recombine.

The purpose of this work is to give a decisive experimental proof of the vertical transport model by the comparison of the PL rise times observed in heterojunctions and epilayer samples. In fact, the vertical transport is a movement of the carriers in real space. Therefore, in heterojunctions the exciton formation is delayed until the carriers reach the flat band region. The issues of the paper are threefold: (a) verify the assignment of the PL signal observed in heterojunctions, (b) confirm the model of the vertical drift of the carriers, (c) characterize the PL signal of the carriers drifting vertically. The paper is organized as follows. We first identify the PL peaks observed in the heterojunctions by comparison with the PL spectra of the GaAs epilayer sample. Then, we discuss the magnetic field dependence of the PL peaks, before coming to the discussion of the time-resolved PL data. Finally, we characterize the PL signal of the carriers drifting vertically in the heterojunctions on the basis of the analysis of the free exciton (FX) dynamics.

II. EXPERIMENTAL DETAILS

The samples used in this study were MBE grown GaAs epilayers and modulation doped GaAs/$Ga_{1-x}Al_xAs$ heterojunctions. In the heterojunctions, the electron mobilities were better than 5×10^5 $cm^2/V\,s$. The PL spectra of all epilayer and heterojunction samples were well reproducible. Therefore, we selected three representative samples, i.e., one epilayer sample and two *n*-type heterojunction samples with 33% Al. The magneto-PL measurements were carried out in a cryostat with a superconducting magnet enabling fields up to 7.5 T with a sample bath temperature of 1.7 K. Instead, the time-resolved PL measurements were performed at 2 K. For the optical excitation, for the magneto-PL experiments we used an argon-ion laser and for the time-resolved PL experiments we used the frequency doubled output of a femtosecond mode locked titanium-sapphire laser with a central wavelength of 395 nm and a pulse width of 120 fs. The laser repetition rate was 82 MHz. The time-resolved PL spectra were detected with a Hamamatsu streak camera (C4334). The best time resolution per CCD pixel in the streak camera is 5 ps. The time resolution of the real spectra depends further on the synchronization of the detector with the excitation laser. The effective resolution was approximated by 20 ps per 1 ns time span. The spectral resolution was better than 1 Å.

III. MAGNETOPHOTOLUMINESCENCE

In Fig. 1, we show the PL spectra of the epilayer and the hetero No. 1 samples under constant wave (CW) excitation.

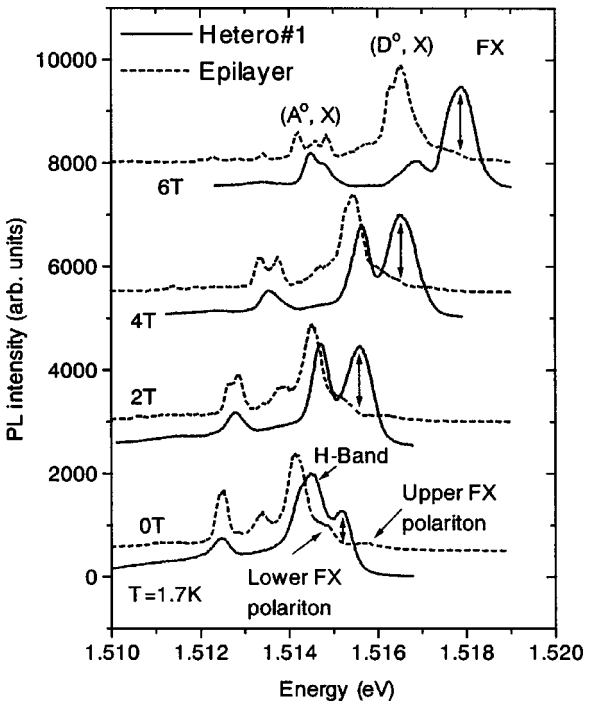

FIG. 1. Comparison between the PL spectra of the hetero No. 1 sample and the epilayer sample at 0, 2, 4, and 6 T.

For the GaAs epilayer, the well known[15] PL peaks of the FX, neutral donor bound excitons (D^0,X) and neutral acceptor bound excitons (A^0,X) are indicated. All these peaks shift diamagnetically in the field. For the heterojunction we observed very similar PL peaks at nearly the same energies except one peak, called the *H* band, related to spatial indirect excitons near the heterointerface.[10] Furthermore, we observed also the same diamagnetic field dependence of all these peaks in the heterojunction as in the epilayer sample. Therefore, for the herojunction we assign the PL peaks displayed in Fig. 1 to the recombinations in the flat band region of the excitons FX, *H* band, (D^0,X), and (A^0,X), in correspondence with the assignments of the peaks observed in the epilayer sample. Still, the magneto-PL taken in the hererojunction shows some additional features, as can be seen in the contour plot of Fig. 2. The black filled circles show the PL peak positions for the GaAs epilayer sample for comparison. The three strong and narrow peaks have been identified as FX, neutral donor bound excitons (D^0,X), and neutral acceptor bound excitons (A^0,X). The spin splitting of (A^0,X) can be observed in high fields. Although the overall diamagnetic shift of the PL peaks is very similar in the epilayer and in the heterojunction (it reflects the three-dimensional character of the excitons recombining in the flat band region), in heterojunctions the PL signal displays oscillations of the intensity and the recombination energy of the FX (it reflects the two-dimensional character due to the screening effect of the 2DEG). The peak of each oscillation can be denoted with an electron filling factor ν. At these filling factors, quantum Hall plateaus are expected. Therefore, these oscillations apparently contradict the assignments of the PL peaks to recombinations of excitons in the flat-band region. The solution of this paradox is given by the vertical drift of the photoexcited carriers from the interface (where they are created) to the flat-band region (where they

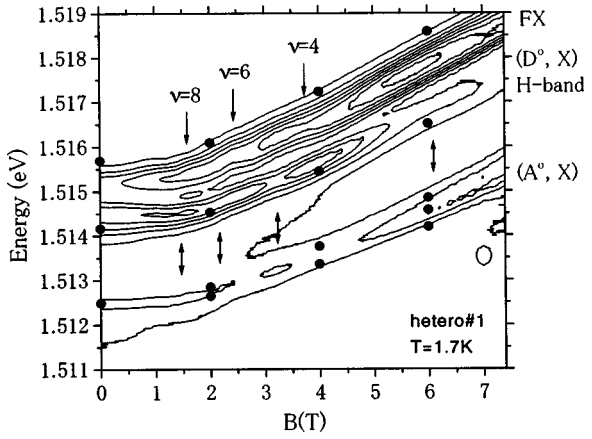

FIG. 2. Magnetic field dependence of the time integrated PL spectra of the sample hetero No. 1 (contour plot) and the epilayer sample (filled circle dots). The overall feature is the diamagnetic shift of the FX, (D^0,X), H band, (A^0,X) in the hetero No. 1 as well as in the epilayer sample. In the former sample, we observe also concurrent oscillations of the PL intensity and the peak energies.

recombine) under the screening effect of the 2DEG. In this sense, the vertical transport influences the density of carriers reaching the flat-band region, and at the same time involves the 2DEG indirectly over the screening effect.

IV. TIME-RESOLVED PHOTOLUMINESCENCE

The vertical drift of the photoexcited carriers influences the dynamics of the exciton formation. This is observed in the time-resolved PL spectra of heterojunctions shown in Fig. 3 for hetero No. 2 for four different excitation densities. In Fig. 3, gray indicates a high PL intensity while white indicates a low PL intensity. Black is the background color and marks the zero of the PL intensity. One recognizes in hetero No. 2 that the rise time of the PL signal strongly depends on the excitation power density [Fig. 3(a) through (d)]. For an excitation power density as low as 25 mW/cm^2, the PL signal of the FX and (D^0,X) shows a very sharp onset, but increasing the excitation power density to 70 mW/cm^2 or more, the rising time becomes longer. Such a long PL rise time for the FX and especially its dependence on the excitation power is characteristic for the heterostructures and has not been observed in any other sample structures. We also tuned the laser pulse to excite below the energy gap of $Ga_{1-x}Al_xAs$ and the same long rise time of the FX signal was observed. Therefore, the long rise time discussed in this paper is not related to energy relaxation of the excitons from the $Ga_{1-x}Al_xAs$ layer to the GaAs layer. Furthermore, nonradiative processes are neglected in our interpretation of the PL data. In fact, very long recombination times, from several nanoseconds to 50 μs have been reported

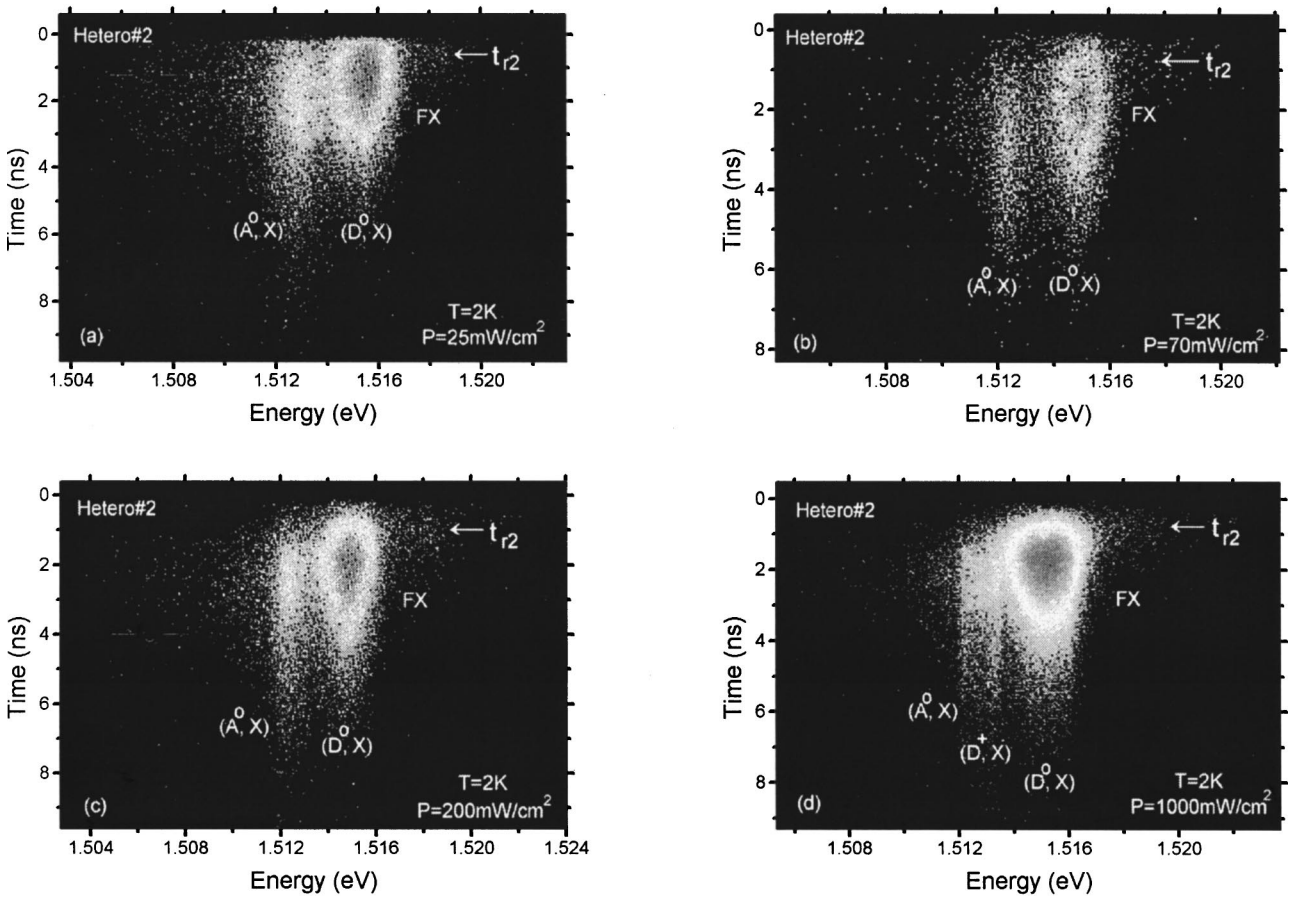

FIG. 3. Streak images of the transient PL of sample hetero No. 2. The gray level indicates the PL intensity going from the white (low PL intensity) to the dark (high PL intensity). Black indicates zero PL signal. The FX, (D^0,X), (D^+,X), and (A^0,X) peaks are identified. The arrow marked as t_{r2} signals the variation of the PL rise time from low excitation density to high excitation density with (a) 25 mW/cm^2, (b) 70 mW/cm^2, (c) 200 mW/cm^2, (d) 1000 mW/cm^2.

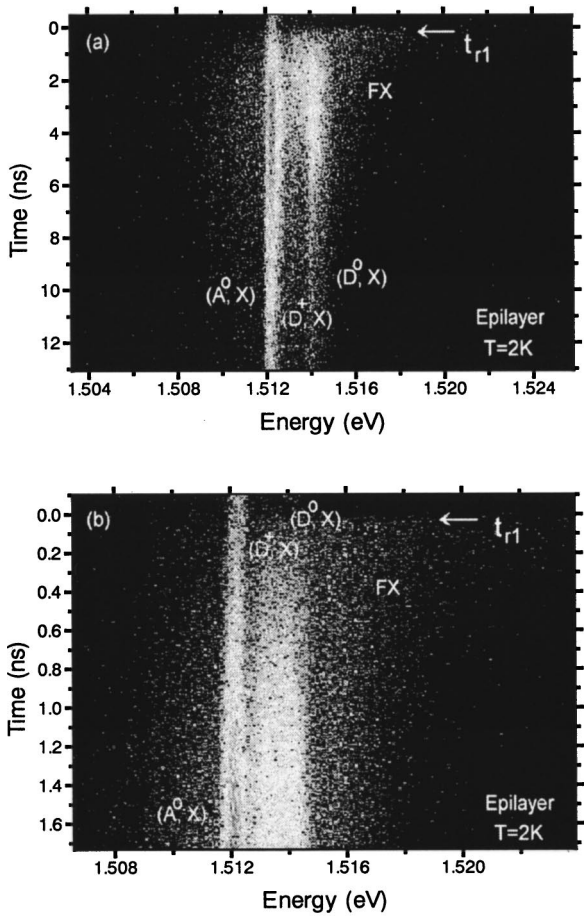

FIG. 4. (a) Streak image of the transient PL detected in an epilayer sample. (b) Enlarged transient image right after the laser pulse. The gray levels are defined as in Fig. 3. The peaks of FX, (D^0,X), (D^+,X), and (A^0,X) are identified. The rise edge of the FX is sharp. The exciton energy relaxation is responsible for a delayed PL rise edge with decreasing recombination energy.

in GaAs/$Ga_{1-x}Al_xAs$ heterojunctions.[16,17] This indicates that the nonradiative recombinations are slower processes in these samples.

For comparison, we display in Fig. 4(a) the time resolved PL spectrum of an epilayer sample. Very sharp PL peaks of FX, bound exciton (D^0,X), (D^+,X), and (A^0,X) are observed. As happens in the heterojunctions, also in the epilayer, a longer PL intensity rise time and decay time for the states with larger binding energy are observed. However, the rise time for the FX is much shorter than that detected for the heterojunctions. The PL intensity of the FX rises almost instantaneously after the laser pulse. To study the sharp rising edge of the FX, we have measured the transient PL spectrum in a much shorter time range [Fig. 4(b)]. The rise time is determined to be shorter than 50 ps without performing any spectral deconvolution of the laser pulse. Moreover, the PL rise time for the FX remains sharp with increasing laser power, shown in Fig. 5. One may have noticed that the FX decay time depends on the laser power in Fig. 5. This is attributed to the fact that the density of excitons with large momentum k increases with increasing excitation power. These high k excitons have a reduced recombination probability because they need to relax their momentum for the radiative recombination. A detailed discussion of this topic is beyond the scope of this paper and will be presented elsewhere.

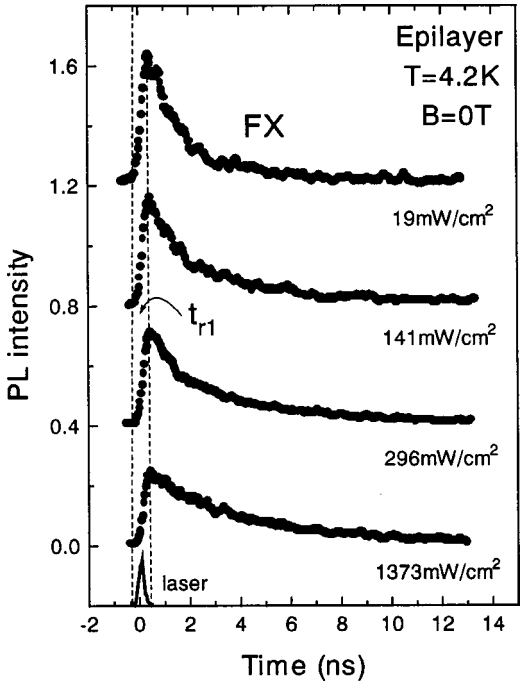

FIG. 5. Transient PL intensity of the FX for four excitation densities in the epilayer. All curves show the same PL rise time, independently on the excitation density in the epilayer.

Integrating the PL signal of Figs. 3(d) and 4 for each type of exciton, we obtain the transient PL intensities spectrally in the epilayer (Fig. 6) and heterojunction (Fig. 7) samples, respectively. The curves are normalized to the maximum transient intensity.

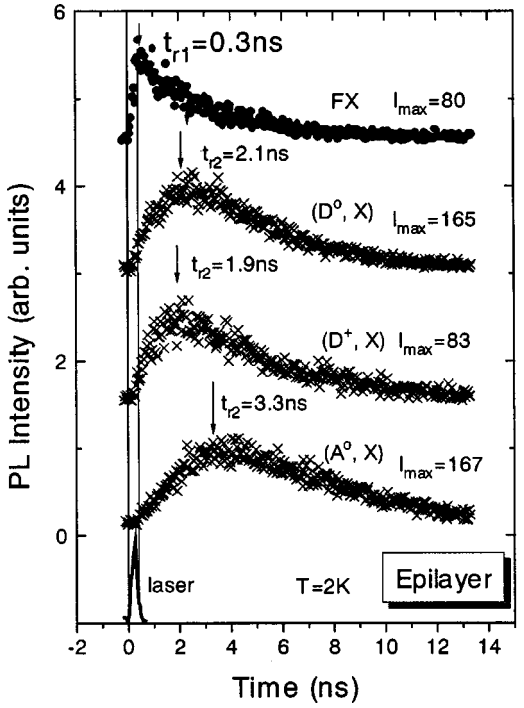

FIG. 6. Energy integrated transient PL intensity for different types of excitons in the epilayer sample. The rise time for the FX is short while that of all bound excitons are longer than 1 ns.

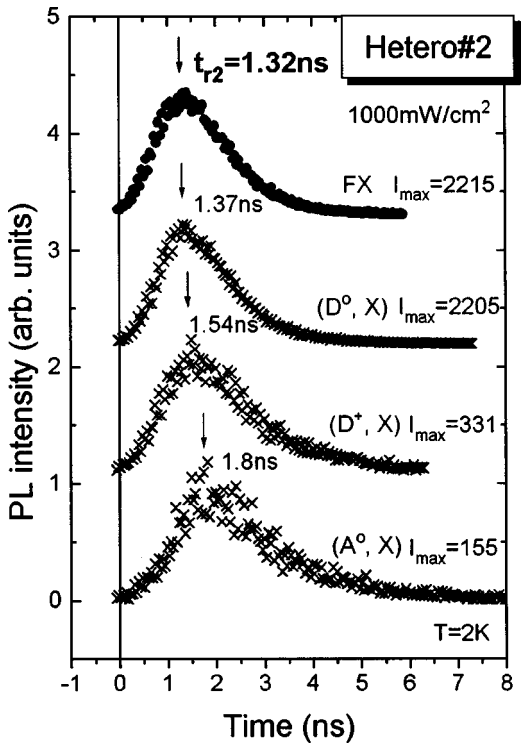

FIG. 7. Energy integrated transient PL intensity for different types of excitons in hetero No. 2. The rise time for the FX is longer than 1 ns and comparable to that of bound excitons.

In epilayers, the PL intensity of the FX rises abruptly at a time delay t_{r1} after the excitation, while the intensities for all bound excitons take over 1 ns to reach the maximum (Fig. 6). The slow rise time is denoted as t_{r2} for both Figs. 6 and 7. In Fig. 8(a), we depict the energy relaxation process occurring in epilayers. The laser pulse excites carriers at an energy far above the band gap. Then, the electron-hole pairs relax to the band edge within a time shorter than 50 ps by emitting optical phonons. After reaching the band edge, these excitons can be trapped by neutral, ionized donors and neutral acceptors, indicated in Fig. 8(a) before recombining. In high quality GaAs samples, the trapping time can be as long as 1 ns. Experimentally, (A^0,X) is found to have a longer rise time of the PL intensity than the (D^0,X) and (D^+,X). This indicates that the (D^0,X) and (D^+,X) can relax further to the lower energetic (A^0,X) [dotted arrows in Fig. 8(a)]. Such an energy relaxation can occur as long as the impurities are spatially not too far apart. The energy relaxation of the excitons prolongs the PL decay time of the low-energy states, because the decayed excitons are partially compensated by the excitons relaxing from high-energy states, see both Figs. 6 and 7. This behavior can be simulated by a relaxation ladder of the excited carriers to the ground state, similarly to what happens in self-organized quantum dots.[18]

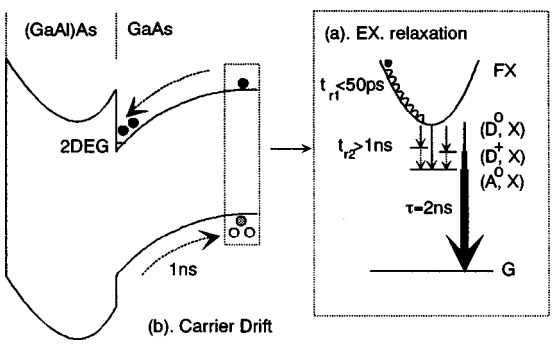

FIG. 8. Schematic diagram for (a) exciton formation and relaxation in epilayers or the flat band region of heterojunctions and for (b) carrier vertical drift in heterojunctions.

Interesting is the origin of (D^+,X), since this PL peak could be assigned as neutral donor-hole transition according to their transition energy.[15] The long rise time of this peak shows clearly that it belongs to the group of bound excitons. Otherwise, the PL rise time should be very short, since neutral donors should exist before the laser pulse and the energy relaxation to the band edge takes only a few picoseconds for the holes. Moreover, the rise time of the (D^+,X) is shorter than that of (D^0,X) and (A^0,X) in the epilayer sample (Fig. 6), which suggests that the ionized donor can polarize and attract the free excitons effectively. The relative PL intensity still depends on the concentration of the different impurities.

We point out that the PL rise time of the FX in our epilayer samples is quite different from that presented in Ref. 5, where a very long PL intensity rise time over 1 ns had been reported. We checked also the PL rise time of the FX under resonant excitation near the band edge, but we could not observe any PL rise time as long as the one reported in Ref. 5. Still, the quality of our sample is good as one recognizes from the fact that the line width of the (A^0,X) exciton is as narrow as 0.44 meV. In our samples, we observed a PL rise time over 1 ns only for the bound excitons.

Also in the heterojunction, the PL rise time of the bound excitons is longer than that for the FXs (Fig. 7) due to the energy relaxation of the excitons. However, in the heterojunctions the PL rise time strongly depends on the excitation power. This is clearly different from the epilayers and can not be explained simply by the energy relaxation in k space. The transient PL spectra shown in Fig. 3 are integrated over the energy region of the FXs and plotted in Fig. 9. We found that the rising edge of the PL signal actually contains two components, i.e., a fast and a slow part. The fast component dominates the PL spectrum of the FX for a low excitation power density, while the slow component increases quickly with increasing excitation power density and finally overwhelms the fast components. This behavior proves the existence of two types of carriers forming excitons in heterojunctions,[10] i.e., the carriers locally excited in the flat band region [gray holes in Fig. 8(b) for the fast PL rise time] and the carriers drifting vertically from the interface to the flat band region [open holes in Fig. 8(b) for the slow PL rise time]. With a low excitation power density, the density of the vertically drifting carriers accumulated in the flat band region is low. In this case, the excitons formed by carriers locally excited near the flat band region dominate the PL intensity and the rising edge for FX behaves similarly to the epilayer samples, as is shown in Figs. 4 and 5. In this case, exciton formation is mostly geminate. In contrast, for a high excitation power, many vertically drifting holes reach the flat band region where they form excitons with locally excited electrons. Then, the PL rise time of the FX is delayed by the time required for the vertical drift of the holes. Thus

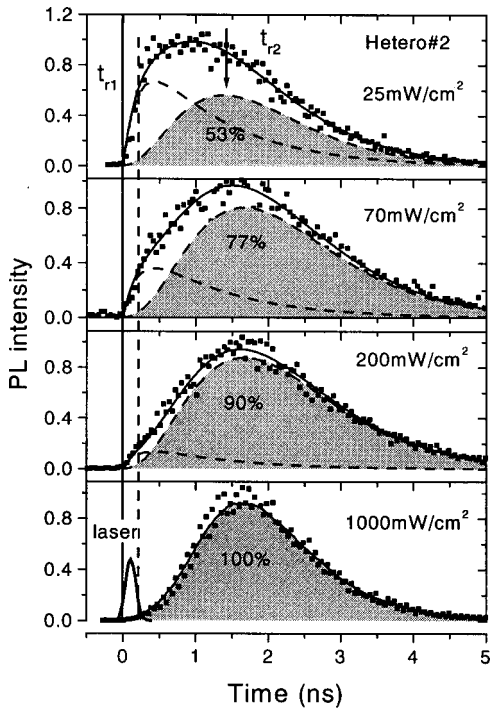

FIG. 9. Transient PL intensity of the FX for four excitation densities in the hetero No. 2 sample. The line shapes are separated into two contributions (dashed lines) corresponding to the recombinations of two types of FX (geminate and bimolecularly formed excitons) in the flat band region. The PL signal of the vertically drifting carriers is shadowed and marked with the percentage contribution for each excitation density.

in high excitation power, exciton formation is mostly bimolecular.

V. NUMERICAL EVALUATION AND DISCUSSION

We determine now the relative contribution of the vertical carrier transport to the PL signal in heterojunctions. In Fig. 9, the PL contribution with a fast rise time corresponds to the recombination of excitons locally excited and takes the following form:

$$I_{\text{fast}}(t)=I_{f0}(e^{-t/\tau}-e^{-t/\tau_r}), \tag{1}$$

where τ is the PL decay time, τ_r is the PL rise time and I_{f0} is proportional to laser excitation power. The linear power dependence of I_{f0} is understood to be connected with the geminate exciton formation. In such a case, each exciton is formed by one electron-hole pair generated exactly by one photon. This is observed only when the coupled electron-hole pairs do not dissolve (due to scattering processes, drift, or diffusion) until they relax their energy to the band edge and finally recombine. Instead, the PL contribution with a slow rise time is connected with the carriers (holes for *n*-doped heterojunctions) created near the heterointerface and reaching the flat band region after a vertical drift. Unlike the fast PL component, the slow PL contribution has a quadratic dependence on the excitation power. For this part of the PL signal, the exciton formation takes place bimolecularly after the holes drift vertically from the heterointerface to the flat band region of GaAs (this takes about 1 ns) and finally couple with the long-living electrons excited locally and still present there. The convex curvature of the rise edge of the slow PL component depends sensitively on the details of the vertical motion of the photoexcited holes.[19] One shall remark that it would be inappropriate to call the slow PL component a nonlinear optical effect or a ''nonlinear PL signal'' [20–22] in spite of its quadratic power dependence. In fact, there is no evidence of a higher-order polarization. Assuming an exponential decrease of the interface electric field from the heterointerface to the flat band region of the form

$$|\vec{\mathbf{E}}(z)|=E_0e^{-z/d}, \tag{2}$$

where $z=0$ corresponds to the position of the heterointerface, the vertical drift of the holes until the flat band region is described by a nonlinear differential equation. Including in the equation of motion also the scattering of the carriers (holes in *n*-doped heterojunctions) off impurities and taking into account the distribution of the photocreated carriers along the vertical direction z (in connection with the light intensity variation along z), one obtains the following transient PL intensity:

$$I_{\text{slow}}(t)=I_{s0}e^{-t/\tau}((1+\gamma t)^{\beta}-1) \tag{3}$$

for the slow component of the transient PL signal.[19] Here τ is the decay time of the excitons in the flat band region. I_{s0} is proportional to the densities of the electrons and the holes present in the flat band region. These electrons and holes have a different ''history'' (as explained above). Therefore, they cannot be compared directly. But the densities of these electrons and holes are both proportional to the excitation power. Therefore, I_{s0} has a square power dependence. The dimensionless exponent β depends on the scattering rate of the carriers during the vertical drift to the flat band region, as well as on the absorption coefficient of GaAs, and the decay length and amplitude of the interface field. The unusual convex form of the rising edge of I_{slow} observed experimentally at high excitation powers (Fig. 9) can be observed only for good samples and at low temperatures, i.e., for $\beta\gg 1$. The maximum of the slow component of the transient PL signal is found at

$$t_{\max}=\beta\tau-\frac{1}{\gamma}. \tag{4}$$

This maximum is closely related to the average drift time of the carriers from the interface to the flat band region. The form (3) fits the lineshape of the slow PL component well and enables a separation of the total transient PL signal of the FX in the heterojunction into the fast and slow components. The relative intensity of the slow component is indicated in Fig. 9 for four excitation power densities. The maximum of the slow PL component is reached at 1.6 ns after the excitation and does not change significantly with the excitation power density (Fig. 9). From the fit of the slow PL component, we found a mean fly time of 64 ps between two scattering processes off impurities during the vertical transport and an average decay time of 0.51 ns for the excitons formed bimolecularly in the flat band region. The decay time differs slightly for the excitation density of 70 mW/cm^2 and 200 mW/cm^2, which might be related to the variation of the interface electric field, and therefore of the carrier dynamics,

with the laser excitation power density. For the locally excited carriers (fast PL component) we found a PL decay time and rise time of 0.8 and 0.3 ns, respectively, for all transient line shapes. In reality, the PL rise time of 0.3 ns does not correspond to the real exciton formation time because of the corresponding spectral resolution described in Sec. II. The fitted exciton decay time depends on both, the exciton recombination rate and the capture rate of FXs by the impurities. Whereas the mean fly time characterizes the scattering process of the photoexcited carriers when no other exciton build up process is involved in the PL rise time. Therefore, we emphasize that a similar evaluation could not be performed for (D^0,X) and (A^0,X), because their intensity rise edge involve the information of FX relaxation.

Finally we discuss the reliability of our data analysis. In Fig. 3, one notices that the (D^0,X) and the FX overlap. Therefore, one could argue that the long PL rise time observed for the FX is due to the spectral overlap of the (D^0,X) and the FX. We integrated the transient PL intensity over narrow energy intervals and evaluated the PL rise time for each energy stripe. The results are plotted in Fig. 10(a) for both, the epilayer and the heterojunction samples together with the time-integrated PL spectra [Fig. 10(b)]. Two features are noticed. Firstly, the rise time reaches a local maximum for each exciton peak. Secondly, in the epilayer the rise time drops drastically when the recombination energy increases from the bound excitons to the FX, while such a strong drop is not observed in the heterojunctions. A long PL rise time is observed for the FX over a broad energy region in the heterojunctions. This indicates that the spectral overlap between the (D^0,X) and the FX is not the origin of the long PL rise time observed for the FX in heterojunctions. In fact, the halfwidth of the (D^0,X) is expected to be similar to that of the (A^0,X) and the (D^+,X). Therefore, the long PL rise time of the FX is an intrinsic property of heterojunctions and can only be explained by taking into account the delay time connected with the vertical drift of the carriers from the heterointerface to the flat band region.

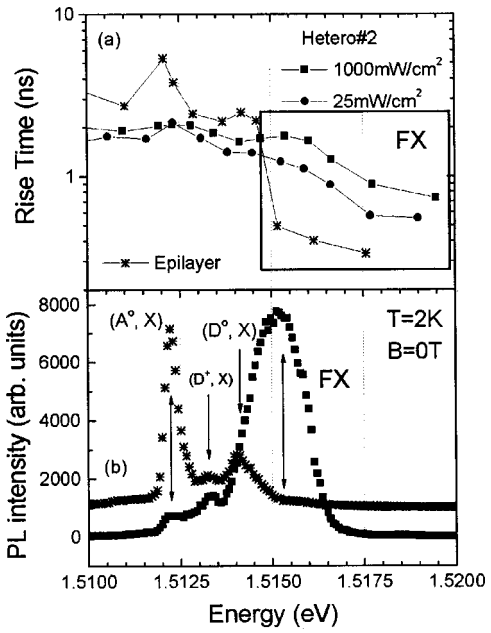

FIG. 10. (a) Energy dependence of the PL rise time in the heterojunction and epilayer samples. One shall notice the different rise times for the FX in the epilayer and heterojunctions up to a very large kinetic energy of the FX. (b). Time integrated PL spectra for hetero No. 2 and the epilayer sample, corresponding well to the PL spectra taken under CW excitation (Fig. 1).

VI. CONCLUSIONS

We have evaluated the transient behavior of different types of excitons in heterojunctions and epilayers. We have focused on the PL rise time which is found to increase with increasing exciton binding energy. Particular are the FX's in heterojunctions which exhibit a remarkably longer rise time than the FX's in epilayers. This is understood by considering the vertical drift and energy relaxation of the photoexcited carriers driven by the interface electric field before they form excitons in the flat band region. This phenomenon is observed as a PL contribution with a slow rise time (connected with the drift time required to reach the flat band region) in the transient PL signal of the FX in heterojunctions.

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[1] For recent research activities on excitons, see *Excitonic Processes in Condensed Matter, EXCON'98, Boston,* edited by R. T. Williams and W. M. Yen (Electrochemical Society, 1998).

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