## Fermi-edge-induced magnetophotoluminescence in high-carrier-density single heterojunctions

F. A. J. M. Driessen and S. M. Olsthoorn

Department of Experimental Solid State Physics, Research Institute for Materials, University of Nijmegen, Toernooiveld, NL 6525 ED Nijmegen, The Netherlands

T. T.J. M. Berendschot

High Field Magnet Laboratory, University of Nijmegen, Toernooiveld, NL 6525 ED Nijmegen, The Netherlands

L.J. Giling

Department of Experimental Solid State Physics, Research Institute for Materials, University of Nijmegen, Toernooiveld, NL 6525 ED Nijmegen, The Netherlands

D. M. Frigo

Billiton Research B.V., P.O. Box 40, NL 6800 AA Arnhem, The Netherlands

G. A. C. Jones, D. A. Ritchie, and J. E. F. Frost

Cauendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 OHE, United Kingdom (Received 8 June 1992; revised manuscript received 7 August 1992)

Fermi-edge-induced excitonic magnetophotoluminescence (MPL) of second ( $n_c = 2$ )-subband electrons and photoexcited holes  $X(n_c=2,h)$  is reported in high-carrier-density GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As single heterojunctions for two-dimensional carrier densities in the range  $(6.6-16.2) \times 10^{15}$  m<sup>-2</sup>. The MPL shows magneto-oscillatory behavior of intensity, photon energy, and peak width. Furthermore,  $n_c = 2$  subband recombination with light holes  $X(n_c=2, l)$  is reported. A splitting of the  $X(n_c=2, h)$  is reported at low carrier density, which results from the separate interaction of the two spin-split states of the  $n<sub>c</sub> = 1$  Landau level in which the Fermi level resides on the second subband. The dependence of the spectra on both excitation density and temperature indicates strongly that photoluminescence (PL) from the  $n<sub>c</sub> = 3$ subband occurs. PL originating from the two-dimensional electron gas (2DEG) and that from the GaAs buffer layer could be distinguished by excitation below and above the GaAs band gap. In the highestcarrier-density sample, evidence is found for the formation of a band of localized states betwen the spin components of a Landau level. Recombination occurs between the exponentially decaying tails of the photoexcited holes, which relax to the flat-band region of the GaAs. Additional information on the 2DEG was obtained via resonant-excitation experiments. Phonon replicas of both the second-subband exciton and the *unpopulated* third subband are observed. The latter very sharp replica appears if a nonequilibrium electron concentration is created in the resonantly excited third subband. Resonant excitation also reveals the recombination of electrons in the  $n_c = 2$  subband with holes located at neutral acceptors. Strong indications were found for PL of the  $n_c = 1$  lowest Landau level upon resonantly exciting the  $X(n_c=2, h)$  transition in the lowest-carrier-density sample.

#### I. INTRODUCTION

The presence of exciton states at the Fermi edge of degenerate semiconductors, as a result of electron-hole multiple scattering, was demonstrated by calculations of Mahan.<sup>1</sup> These many-body excitons are observed as a logarithmic singularity in low-temperature absorption spectra: the Fermi-edge singularity (FES). The FES is usually not seen in luminescence spectra because conservation of momentum requires that mainly holes near  $k=0$  are involved in emission. Skolnick et al.<sup>2</sup> circumvented momentum conservation by real-space localization of the holes as a result of alloy fIuctuations in a modulation-doped  $In_{0.47}Ga_{0.53}As-InP$  quantum-well structure. Hence, luminescence from all electrons up to  $E_F$  could be observed, thereby obtaining the density of states between the Landau levels in the two-dimensional electron gas  $(2DEG)$ .<sup>3</sup>

Various other properties of 2DEG's have been studied magneto-optically, such as electron-electron interacions,<sup>4</sup> the integral and fractional quantum Hall effects,  $5^{-7}$  and Auger recombination within Landau levels.<sup>8</sup> In these studies use was made of modulation-doped quantum wells (MDQW's) because of the presence of hole subbands  $n_n$  (Refs. 3 and 9) and GaAs-Al<sub>x</sub>Ga<sub>1-x</sub>As single heterojunctions, for which photoluminescence (PL) from the GaAs buffer layer was prevented from dominating the spectrum. This was achieved either by choosing very thin buffer layers<sup>10</sup> or by placing an additional  $\delta$  layery thin buffer layers<sup>10</sup> or by placing an er of acceptors in the GaAs buffer.<sup>5,11,12</sup>

Partly motivated by recent spectroscopic observations of the fractional and integral quantum Hall effect,  $6,7,13,14$ renewed theoretical interest has arisen in the subject of a confined Fermi sea.  $15-19$  The latter references examined the inhuence of the Fermi sea on a single subband in a magnetic field.

The presence of a second subband has been shown experimentally to be of great influence. Chen et  $al$ .<sup>20</sup> showed that the hybridization between the Fermi-edge resonance and the second subband appeared to result in strong magneto-oscillations with Shubnikov —de Haas (SdH) periodicity in the intensity of an excitonlike PL transition. In their MDQW the Fermi level was located just below the  $n_c = 2$  subband. Without including the effect of a magnetic field, Mueller, Ruckenstein, and 'Schmitt-Rink<sup>21,22</sup> examined theoretically the influence of an excitonic resonance originating from a higher conduction subband on the optical spectra of an  $n$ -type degenerate semiconducto.

In a recent paper,  $2^3$  we reported the observation of the second-subband exciton  $X(n_c=2, h)$  in a high-carrier density single heterojunction. This exciton could be observed because of hybridization between the Fermi-edge resonance and the second subband. The  $X(n_e = 2, h)$  was observed if the magnetic-field strength was large enough to depopulate the second subband. In contrast to the previously mentioned magneto-optical publications on 2D systems, the holes in the heterojunction were not localized or trapped in thin buffer layers, quantum-well structures, or at additional acceptor sites. The intensity of the  $X(n_c=2, h)$  luminescence showed oscillatory behavior with SdH periodicity. The extremely small linewidth of the PL signal also permitted the observation of magneto-oscillations in both photon energy and peak width. These oscillations were caused by crossing Landau levels of the  $n_c = 1$  subband with the lowest Landau level of the  $n_c = 2$  subband. Furthermore, tilted-field measurements enabled the determination of the intrasubband diamagnetic shift in a parallel field.

In this paper we further investigate Fermi-edgeinduced magnetophotoluminescence (MPL) in three high-carrier-density heterojunctions with total sheet carrier densities  $n_T$  ranging between 6.6 and 16.2×10<sup>15</sup>  $m.$ <sup> $-2$ </sup> New Fermi-edge-induced emissions and other information of the 2D system are reported. We discuss temperature-dependent behavior and excitation-densitydependent behavior for excitation both below and above the GaAs band gap.

Finally, we present PL measurements in which the laser was resonantly tuned to several 2D emissions. Strongly enhanced 2D-PL processes, which are related to the resonantly excited states and not observable in the nonresonant spectra, yield additional information on the 2D system.

#### II. EXPERIMENTAL DETAILS

The experiments were carried out on three monolayerdoped GaAs-Al<sub>x</sub>Ga<sub>1-x</sub>As heterojunctions with high carrier density grown by molecular-beam epitaxy at the Cavendish Laboratory, Cambridge. The sample structure has been described in our previous paper.<sup>23</sup> The distance z between the Si doping layer in the  $Al_xGa_{1-x}As$ and the GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As heterointerface differed between the samples. These spacer thicknesses z for the

TABLE I. Properties of the samples: spacer thickness z, total sheet carrier density  $(n_T)$ , and mobility  $(\mu)$  measured in the dark and after saturation of the persistent photoconductivity effect by illumination with a red LED (after Ref. 25). Separate sheet carrier densities of the two subbands  $n_{1,2}^{\text{light}}$  at zero field are specified.

Sample		2	3
Growth run	A <sub>233</sub>	A232	A 227
$z$ (nm)	20	10	
$n_T^{\text{dark}}(10^{15} \text{ m}^{-2})$	3.8	6.4	8.9
$n_T^{\text{light}}(10^{15} \text{ m}^{-2})$	6.6	11.3	16.2
$n_1^{\text{light}}(10^{15} \text{ m}^{-2})$	6.3	10.3	14.5
$n_{2}^{\text{light}}(10^{15} \text{ m}^{-2})$	0.3	1.0	1.7
$\mu_{\text{dark}}$ (m <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	55	39	9.8
$\mu_{\text{light}}$ (m <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	56	39	8.5

heterojunctions, as well as the corresponding carrier densities and mobilities measured both in the dark and after saturation of the persistent photoconductivity<sup>24</sup> by illumination with a red light-emitting diode (LED), are shown in Table I. Other sample parameters can be found in the work of Kusters et al.<sup>25,26</sup> The experimental conditions for the MPL measurements have also been described previously.<sup>23</sup>

## III. RESULTS AND DISCUSSION

#### A. Lowest carrier density

# 1.  $X(n_c = 2,h)$  and  $X(n_c = 2,l)$

The excitonic part of the spectrum of sample <sup>1</sup> at various magnetic-field strengths recorded at  $T=1.5$  K is given in Fig. 1. For this sample with the lowest carrier density a few peaks showed magneto-oscillatory behavior of photon energy, as is shown in Fig. 2. Because the most intense line has qualitatively the same magneto-optical properties as the  $\hat{X}(n_c=2, h)$  in sample 2, reported on in bur previous paper,  $23$  it has also been assigned to secondsubband luminescence, which can be observed owing to the involvement of the Fermi sea. Because the second subband of sample <sup>1</sup> has a lower electron concentration than that of sample 2, a lower magnetic field is required to depopulate the second subband, so the  $X(n_c = 2, h)$  can be resolved at fields as low as 0.8 T. The PL line of sample <sup>1</sup> is narrower than that of sample 2 because of the thicker  $Al_xGa_{1-x}As$  spacer layer that separates the 2DEG from the ionized impurities in the sheet-doping layer. The same procedure we used previously<sup>23</sup> for sample 2 was applied to construct the Landau fan diagram of the  $n_c = 1$  subband and the behavior of the Fermi energy. First, the energy of the second subband was extrapolated to zero field, and the energies of the lowest subband and that of the Fermi level were calculated using That of the Fermi level were calculated using<br> $E_2 - E_1 = 22.3$  meV and  $E_F - E_2 = 1.1$  meV, which values were taken from magneto-transport measurements<br>on this sample for  $B = 0$  T.<sup>25</sup> Second, the Landau fan diagram was constructed taking an effective mass of 0.070 $m_0$ , where  $m_0$  is the mass of a free electron. This



FIG. 1. PL spectra of sample 1 at various magnetic fields  $B$ .

value was calculated directly from the relationship besheet carrier density and the effective mass valid neto-optics.<sup>4</sup> The calculated energy of the Fermi  $= 0$ , assuming  $\delta$ -function-shaped Landau levels, by the bold line in Fig. 2 in the pertinent range of field strengths. The dotted line shows the photon ener-



FIG. 2. The photon energies of various Fermi-edge-induced 2D peaks in sample 1, parts of the Landau-level fan diagram of the  $n_c = 1$  subband, and the calculated energy of the Fermi level in the field range of interest (bold line) assuming  $\delta$ -functionshaped Landau levels. Filling factors  $v$  are shown at the top axis.

gy of the second subband if it were unaffected by the Fermi electrons. It should be noted that this photon energy shows a detectable quadratic dependence upon  $B$ , which mplies a small exciton binding energy. This is in contrast to results obtained for sample  $2,^{23}$  and can be attributed both to the smaller value for the intrinsic electric field, which acts on the photoexcited holes, and the larger extent of the  $n<sub>c</sub> = 2$  subband, which results in overlap with these holes.

Above  $B = 4$  T another remarkable peak is observed at higher energy (see the upper line in Fig. 2). The phot energy of this peak is also affected by crossing  $n_c = 1$  Lancates that 2D electrons are involved in the recombination process. Furthermore, the peak disappears above  $\cong$  20 K. These observations show that the peak is induced by the Fermi sea. Because of the strong interaction between the  $n_c = 2$  subband and the Fermi electrons, the most likely candidate for the emission is an exciton consisting of  $n_c = 2$  electrons and light holes, hereafter denoted  $X(n_c = 2, l)$ . Fermi-edge-induced emission of the  $n_c = 3$  subband is highly unlikely, because the relaxation time between the  $n_c = 3$  and  $n_c = 2$ bands (which both have nonequilibrium concentrations of electrons in them) should then exceed the typical time cale for Fermi-edge-induced luminescence. This intrasubband relaxation time can be estimated to be around  $100 \text{ ps.}^{27,28}$  The even smaller recombination times of the ents. Furthermore, larger excitonic effects induced processes would then imply lifetime broadening<br>of the peaks of at least 10 meV, which was not observed are expected for Fermi-edge-induced PL from the  $n_c = 3$ subband, owing to the greater extent of the  $n_c = 3$  wave function; these effects were not observed in our experiments either.

# 2. Effects of spin splitting of  $n_c =1$  Landau levels on  $X(n_c = 2,h)$

Apart from these two dominating PL processes involvthe  $n_c = 2$  subband, the  $X(n_c = 2, h)$  shows a splitting at high fields (around  $v=5$ ). This splitting can be observed in this sample because the  $X(n_c = 2, h)$  emission is the narrowest. The detailed spectra in this field region in Fig. 3. The energy difference between both he  $X(n_c=2, h)$  peak increases until filli factor 5 has been reached; at higher fields it decreases again. It is well established that the effective  $g$  factor, which determines the separation between two spin-split states, is enhanced at odd filling factors. This g-factor enhancement is caused by exchange interaction among electrons, and is proportional to the population difference between two spin components of a Landau level.  $29,30$ Maximal enhancement is therefore obtained at odd filling at which one spin level is fully occupied and the other is unoccupied. The measured increase in energy difference at filling factor  $v=5$ , therefore, shows that spin splitting of the Landau level in which  $E_F$  resides influences the  $X(n_c=2, h)$ . The quantity  $g^* = \Delta E / \mu_B B$ , where  $\Delta E$  is the energy splitting between the two peaks and  $\mu_B$  is the Bohr magneton, is plotted versus the field strength in the inset of Fig. 3. It must be stressed that

this is not necessarily the real value of the enhanced g factor at  $v=5$ : it is the effect which these two spin-split states have on the photon energy of the  $X(n_c=2,h)$ . The inset, therefore, serves only to visualize the maximum at  $v=5$  ( $B = 5.4$ T).

#### 3. Deviation from linear behavior of Landau levels

In Fig. 2 it was seen that the energy of the  $X(n_c=2, h)$ is affected by the crossing of the  $n_c = 1$  Landau levels. A less pronounced, though remarkable deviation of  $\sim 0.2$ meV in photon energy was observed at  $B = 4.6$  T: exactly the field at which  $E_F$  shifts dramatically ( $v=6$ ). Skolnick et  $al$ .<sup>3</sup> observed deviations from the linear behavior of Landau levels as soon as they began to depopulate (i.e., at even  $v$ ) in a MDQW with one occupied subband. The deviations were of order 5 meV and were mainly attributed to the dependence of excitonic effects on Landau-level filling. Because the exciton binding energies are considerably greater in a MDQW than in a single heterojunction, we expect that the same effect causes the small deviations from linearity upon depopulating the lowest Landau level of the  $n_c = 2$  subband.



## 1. Low-field data: Crossing behavior of  $(A<sup>0</sup>, X)$  and  $X(n<sub>c</sub> = 2, h)$

Earlier data on sample 2 (Ref. 23) reported the situation in which the Fermi level  $E_F$  lay either below or in the lowest Landau level of the  $n_c = 2$  subband. Below  $B \approx 2.5$  T this is no longer the case and the intensity of the second-subband exciton  $X(n_c=2, h)$  is greatly reduced because then the transition probability for the  $X(n_c=2, h)$  emission is no longer enhanced. Very lowintensity  $X(n_c = 2, h)$  luminescence can, however, still be seen in the spectra at low magnetic fields because a nonvanishing wave-function overlap with the photoexcited holes exists as a result of the relatively large mean extent  $\langle z \rangle$ <sub>2</sub> of the  $n_c = 2$  subband electrons from the interface. This  $\langle z \rangle_2$  is especially large if the depletion charge is neutralized as a result of electron-hole pair generation,  $31$ as is the case under our experimental conditions. Figure 4 shows portions of the spectra for fields between  $B = 0.3$ and 1.9 T. Below  $B = 0.8$  T the  $X(n_c = 2, h)$  has clearly ower energy than the two  $(A^0, X)_{J=5/2,3/2}$  states. The  $X(n_c=2, h)$  follows the lowest Landau level of the second subband and the photon energy of  $(A^0, X)$  shows a less pronounced dependence upon  $B$ , which is a characteristic



FIG. 3. The detailed spectra of sample 1 for  $5.1 < B < 6.0$  T. The inset shows the quantity  $g^* = \Delta E / (\mu_B B)$ , with  $\Delta E$  the energy difference between the two  $X(n_c=2, h)$  peaks,  $\mu_B$  the Bohr magneton, and  $B$  the magnetic field.



FIG. 4. The low-field crossing behavior of the second subband exciton  $X(n_c=2, h)$  and "bulk" acceptor-bound excitons  $(A<sup>0</sup>, X)$  in sample 2.

behavior of a bound exciton. The spectra show that the two ( $A^0$ ,X) states are indistinguishable from the secondsubband exciton  $X(n_c=2, h)$  between  $B = 0.8$  and 1.3 T. Above  $B=1.3$  T the  $X(n_c=2, h)$  has a higher energy than  $(A^0, X)$  and, as shown in Fig. 4 of Ref. 23, at higher field strengths the  $X(n_c=2, h)$  shows its characteristic magneto-oscillatory properties. This crossing of the  $(A<sup>0</sup>, X)$  indicates that the interaction between the "bulk excitons" and the "2D excitons" is negligible, otherwise the noncrossing rule of quantum mechanics would have applied,  $32$  resulting in mixed charaeter of the two transitions.

## 2. Excitation density dependence of the emissions

The efficiency of the Fermi-edge-induced  $X(n_c=2, h)$ transition in magnetic field was found experimentally to<br>be very large, both by Chen et al.<sup>33</sup> in MDOW's and in our work on single heterojunctions.<sup>23</sup> Here we present the behavior of the PL spectrum of sample 2 as a function of excitation density  $\overline{P}$  for laser excitation at  $B = 5.4$  T  $(v=7)$  at energies both below and above the band gap. Figure  $5(a)$  shows the spectra for above-band-gap excita-Figure 5(a) shows the spectra for above-band-gap excita-<br>tion with  $E_l = 1.537$  eV, whereas Fig. 5(b) shows the P dependence for  $E<sub>l</sub>=1.52$  eV, which is below the band gap at  $B = 5.4$  T. The  $X(n_c = 2, h)$  peak is seen to be considerably broader when  $E_1 > E_{\text{gap}}$ . This is presumably



FIG. 5. The PL spectrum of sample 2 vs excitation density at  $B = 5.4$  T ( $v=7$ ) for (a) above and (b) below band-gap laser excitation. The energy of  $FX$  decay is also shown in (b). Energies of the  $X(n_c=2, l)$  and the  $X(n_c=2, h)$  are shown in (c) with parts of the Landau fan diagram.

caused by exciton scattering by "hot" electrons. This scattering mechanism is known to have a profound effect exciton kinetics.<sup>34</sup> Furthermore, luminescence from the GaAs buffer layer ( $A^0$ , X) and especially FX is seen in Fig. 5(a) to increase faster than that from the  $X(n, =2, h)$ upon increasing P. The intensity rise of  $(A^0, X)$  and  $FX$ s considerably less pronounced for the below-band-gap is considerably less pronounced for the below-band-gap excitation in Fig. 5(b). If  $E_l > E_{\text{gap}}$ , excitons in the GaAs buffer layer are formed from free electrons and holes; whereas for  $E_l < E_{\text{gap}}$ , no free carriers are present. Obviously, the exciton formation probability for below-bandgap excitation cannot compete with the probability of exciton formation via free carriers in this heterojunction. The relative suppression of  $3D$  excitons observed in Fig. d by the appearance of a shoulder at higher energy which appears at high P. This shoulder is absent in Fig. 5(a), which indicates that its origin lies in the 2D system and not in the GaAs buffer layer. In view of its properties, discussed below, the shoulder has been labeled  $(n_c = 3, h)$ . The peak labeled  $X(n_c = 2, l)$  in Fig. 5(b) has slightly higher energy than that of the  $FX$ , which is more clearly observed in Fig.  $5(a)$ . The FX energy is also indicated in Fig. 5(b). As with the previously described light exciton of the  $n_c = 2$  subband in sample 1, upon changing B the energy of this  $X(n_c = 2, l)$  peak also appears to be affected by crossing  $n_c = 1$  Landau levels. This can be seen in Fig.  $5(c)$ , where the energy of the  $X(n_c=2, l)$  is shown together with that of the  $X(n_c=2, h)$  and parts of the Landau-level fan diagram. Therefore, the Fermi-edge-induced light exciton of the  $n_c = 2$  subband has also been observed in sample 2 by studying the excitation density and excitation energy dependence of the spectra.

## 3. Temperature dependence of the spectra

Figure 6 shows the temperature dependence of sample 2 at  $B = 7.0$  T. At this field the  $n_c = 2$  subband is not populated by equilibrium electrons, as can be seen in Fig.



FIG. 6. The temperature dependence of the PL spectrum of sample 2 at  $B = 7$  T.

4 of our previous paper.<sup>23</sup> Two subpeaks appear on the high-energy side of the  $X(n_e=2, h)$  upon increasing temperature; their origin is uncertain. A possibility is that thermal population of holes in higher  $J_z$  states may result in additional  $PL$  peaks. Another possibility for the origin of one subpeak concerns a separate many-body interaction on the second subband of two spin-split states of the particular  $n_c = 1$  Landau level in which  $E_F$  resides, as was also found above for sample <sup>1</sup> through the observation of g-factor enhancement. This can occur here because the experiment was performed at  $B = 7$  T, just above the  $v=7$  field. Therefore, thermal depopulation of the slightly populated energetically lowest spin component of Landau level  $N=3$  of the  $n_c=1$  subband may lead to the same effect described above: separate influence of both spin components on the  $X(n<sub>c</sub> = 2, h)$ , resulting in the observation of a splitting of the  $X(n<sub>c</sub>=2, h)$ . The entire disappearance at  $\sim$  20 K of the broadened complex of peaks is not surprising because the interaction between the Fermi electrons and the second subband is highly sensitive to temperature.  $3,21$ 

Free-exciton PL of the buffer layer was observed around 1.518 eV and disappeared at about 50 K. At higher temperatures, a peak appeared which at low  $T$  was observed around 1.5184 eV. The luminescence of this peak could still be clearly observed at temperatures up to 320 K, which is the upper limit in our sample cell at  $B\neq0$ . The energy of this peak followed that of the GaAs band gap as a function of temperature. At  $T = 320$  K the PL intensity depended quadratically on the excitation density:  $I \propto P^2$ . This behavior is not expected for bufferlayer excitons because of screening of ionized impurities. In quantum wells it is well known that electrons and holes exhibit a strong exciton binding owing to their large wave-function overlap,  $35,36$  and exciton PL can still be observed at higher temperatures compared to bulk exciton PL.<sup>37</sup> In our system the  $n_c = 3$  subband has similar properties. First, this subband has a large spatial extension into the flat-band region, which ensures a considerable overlap with photoexcited holes; second, ionized impurities are spatially separated in  $Al_xGa_{1-x}As$ . Therefore we believe that the highest-energy peak corresponds to the  $(n_e=3,h)$  recombination, which possibly has an exciton binding energy. Temperature-dependent measurements on samples <sup>1</sup> and 3 showed qualitatively the same behavior. At low temperature, an  $X(n_c = 2, h)$  peak was observed, and at high temperature a new peak at higher energy appeared. The energy difference between these two peaks increased upon increasing sheet carrier density: they were 0.9, 1.7, and 3.<sup>5</sup> meV for samples 1, 2, and 3, respectively. This supports the assignment to  $(n_c = 3, h)$  because all subband separations increase if the intrinsic electric field increases.<sup>38</sup> Further experimental data supporting the  $(n_c=3,h)$  assignment are reported below in the resonant excitation experiments.

#### C. Highest carrier density

For sample 3 (that with the highest carrier density) luminescence was observed with the characteristic 2D magneto-oscillatory properties above  $\sim$  4 T. This field is stronger than the critical values of the other two samples because this is required to depopulate the second subband (cf. our previous paper<sup>23</sup>). Figure 7 shows the very rapid rise in the 2D-luminescence efficiency for this sample: below  $\sim$  4 T the PL is unobservable, whereas at  $B = 6.8$ T its integrated intensity is about seven times larger than the sum of integrated  $(D^0, X)$  and FX buffer layer PL. As an illustration, the 6.8-T spectrum of this sample is shown in the inset of Fig. 7. Maxima in the PL intensity are observed at the odd filling factors 11, 13, 15, and 17. Furthermore, a pronounced maximum is observed at  $v=10$ . In our previous paper<sup>23</sup> we also observed maxima at even  $\nu$ , which were less pronounced than those at odd integers. We argued that, in spite of population fluctuations in the second subband, the Fermi sea was also of influence in observing even  $\nu$  intensity maxima. The present observation of a maximum at  $v=10$  and the absence of maxima at higher even integer filling factors shows that merely the appearance of population fluctuations, which occur at all  $\nu$ , is not sufficient to account for the observed even  $\nu$  maxima in our single heterojunctions. Moreover, the observation of a maximum at  $v=10$ points to the formation of a band of localized states between the spin components of the respective Landau level of the  $n_c = 1$  subband. This occurs if the spin splitting  $g^*\mu_B B$  is large compared with both the thermal broadening  $kT$  and the broadening of the Landau levels  $\Gamma$ . The



FIG. 7. The ratio of the total integrated  $X(n_c=2, h)$  PL to that of the bulk lines  $(D^0, X)$  and FX, vs magnetic field for sample 3. The inset shows the 6.8-T spectrum; the relative gain for the  $(D^0, X)$ -FX PL is 10.

effective  $g$  factors  $g^*$  of both subbands are known to be fairly large from magnetotransport measurements: typical values are 3 for the  $n_c = 1$  subband and 8 for the  $n_c = 2$  subband.<sup>26</sup> Using  $g^* = 3$ , we find at  $B = 6.7$  T that  $g^* \mu_B B = 1.1$  meV,  $kT = 0.12$  meV, and  $\Gamma(n_c = 1) \approx 0.4$ meV for this sample. Hence, the above spin-splitting criterion is fulfilled and Fermi-induced PL at even filling factors is possible in the high-field regime. The previously observed presence of less pronounced even  $\nu$  maxima [shoulders in sample 2 (Ref. 23)] can also be ascribed in the same way.

Magneto-oscillatory behavior of the  $X(n_e=2, h)$  photon energy and its width were also observed for sample 3. Owing to the thin spacer layer in this sample, an overall larger width was measured that oscillated between 0.4 and 0.8 meV. For constructing the Landau fan diagram the magneto-optical effective mass of the  $n_c = 1$  subband electrons at  $n = 16.2 \times 10^{15} \text{m}^{-2}$  was obtained by linear extrapolation of the relationship between  $m^*$  and n (reported by Plaut et  $al.$ <sup>4</sup>), as was previously performed for sample 2.<sup>23</sup> Because of this large value of  $n$ , the extrapolation gives rise to an uncertainty in the effective mass, namely  $0.084 < m^*/m < 0.087$ . Using  $m^* = 0.085m$ , a remarkably good match was obtained between the fan diagram and the field intervals at which the photon energy increases relatively quickly.

#### D. PL mechanism

A new phenomenon was observed by comparing the three similar heterojunctions with variable sheet carrier densities. To wit, Table II summarizes the zero-field energies of the  $n_c = 2$  subband, the calculated zero-field energy separation between the Fermi level and the  $n_c = 2$ subband  $E_{f2}$  and that between the  $n_c = 2$  and  $n_c = 1$  subbands  $E_{21}$ , and finally the calculated zero-field energy of the Fermi level  $E_f$ , for all three samples investigated in this paper. It is seen that within experimental error this zero-field Fermi energy is found to be 1.515 eV in all sam-

TABLE II. Energy of the  $n_c = 2$  subband at zero field  $E_2$ , the zero-field energy separation between the Fermi level and the  $n_c = 2$  subband  $E_{f2}$  and that between the  $n_c = 2$  and  $n_c = 1$  subbands  $E_{21}$ , and the calculated energy of the Fermi level  $E_f$  at zero field.

Sample	$E_2$ (eV)	$E_{f2}$ (meV)	$E_{21}$ (meV)	$E_f$ (eV)
	1.5137	1.1	22.3	1.5148
	1.5116	3.6	30.3	1.5152
	1.5087	6. 1	40.0	1.5148

ples. To a good approximation this Fermi energy equals the Fermi energy in the n-type GaAs buffer layer, which lies  $\sim 0.6 \times E_D$  below the band gap, <sup>39</sup> where  $E_D$ , the donor ionization energy in GaAs, is 5.715 meV. The fact that the inferred Fermi energy in Table II equals 1.515 eV implies that the photoexcited holes, which participate in the various Fermi-induced 2D excitons, are situated in the Hat-band region of GaAs: recombination takes place between their exponentially decaying tails and the 2D electrons. In our PL experiments, therefore, the energies of the subbands decrease if the sheet carrier density increases.

## E. Resonant excitation

So far we have studied the various Fermi-edge-induced emissions of the 2D system by using excitation just above the band gap of GaAs. In the following we used exact tuning of the laser energy to certain PL peaks. This resonant excitation on a PL peak enhances the probability of formation of the particular luminescent process.  $40-42$ The intensities of all peaks that are related to the resonantly excited peak then increase considerably. We used resonant excitation (RE) of 2D luminescence to obtain additional information on the 2D system, which remained hidden in nonresonant measurements; most data were obtained on the sample with the highest carrier



FIG. 8. The PL spectra obtained via resonant excitation experiments on sample 3 at  $B=7$  T. The laser was tuned: well above the exciton energies for spectra (a) and (f); to the  $X(n_c=2,h)$  peak (abbreviated to  $X_2$  in the figure) for (b), (d), and (e); and to the  $n_c=3$ shoulder [denoted  $(e_3, h)$ ] for (c).

density. A compilation of PL spectra, which were obtained by use of resonant excitation (RE) experiments on sample 3 at a field of  $B = 7$  T, is given in Fig. 8. Figure 8(a) was obtained with the laser tuned well above the exciton emissions; it shows the  $X(n_c=2,h)$  and residual  $(D^0, X)$  and FX PL of the GaAs buffer layer. In Fig. 8(b) resonant tuning of the laser to the  $X(n_c=2, h)$  lumines cence resulted in a shoulder  $(e_2, A^0)$  at the high-energ side of the  $(D^0, A^0)$  buffer PL, where  $A^0$  refers to the carbon acceptor. This  $(e_2, A^0)$  peak is separated 19 meV from the  $X(n_c=2, h)$ , which is in excellent agreement with a result of Kukushkin, von Klitzing, and Ploog, 43 who attributed this PL to recombination of a confined  $n_c = 2$  electron with a hole located at an acceptor. This neutral acceptor is located in the vicinity of the 2DEG where a nonvanishing wave-function overlap exists between the acceptor and the exponentially decaying  $n<sub>c</sub> = 2$ wave function. The small separation between acceptor and electron leads to a final-state Coulomb energy, which is why the separation between  $(e_2, A^0)$  and the  $X(n, =2, h)$  is considerably smaller than the carbon acceptor ionization energy of 26.4 meV.

Peak  $(e_2, A^0)$  was accompanied by its phonon replica, which is shown in Fig. 8(e). Both  $(e_2, A^0)$  and this phonon replica were absent if the laser was not tuned to the  $X(n_c=2, h)$ , as can be seen in Fig. 8(f), obtained with the laser energy tuned above that of the exciton emissions. A further indication that  $n_c = 2$  electrons are involved here is that the  $(e_2, A^0)$  transition shifted within experimental error equivalently to the lowest  $n_c = 2$  Landau level if the RE experiment was repeated on the  $X(n_c=2, h)$  at lower fields.

Apart from resonant excitation of the second-subband exciton, we also investigated the PL that we ascribed to



FIG. 9. The temperature dependence of the 7-T PL spectrum of sample 3. The  $X(n_c=2h)$  has been abbreviated to  $X_2$ ; the GaAs buffer layer PL involving rotational states of the donorbound exciton  $(D^0, X)_{\text{rot}}$  and  $FX$  can be clearly distinguished.

the  $n_c = 3$  subband in sample 3. To this end the temperature dependence of the PL spectrum was required; that at  $B = 7$  T is shown in Fig. 9. This figure and some spectra for temperatures between 1.5 and 4.2 K allowed an accurate determination of the energy of the unoccupied third subband  $E(n_c=3,h)$  of 1.518 79±0.000 04 eV at  $T=1.5$ K. In the 1.5-K spectrum a small change in slope could also be observed at this energy, as can be seen in Fig. 9. Resonant excitation was also performed at the energy of this third subband. Although there was no equilibrium electron population in the  $n_c = 3$  subband at 1.5 K, a quasipopulation was created by the RE because of the enhanced probability of exciting a valence electron to the  $n_c = 3$  subband. An extremely sharp phonon replica of this quasipopulated  $(n_c = 3, h)$  subband was then observed at 1.481 92 eV, as can be seen in Fig. 8(c), for which the laser was resonant with the  $n_c = 3$  shoulder. The phonon energy of  $36.86\pm0.04$  meV appeared to deviate slightly from the phonon energy of  $36.75\pm0.04$  meV reported for bulk GaAs.<sup>45</sup> Stroscio, Kim, and Hall<sup>46</sup> demonstrated with a strain-dependent nearest-neighbor linear-chain model that the frequencies of LO-like phonon modes were shifted by a few percent in a superlattice structure. We expect that, in the case of a single heterojunction and, moreover, one in which the mean distance of  $n_c = 3$  electrons to the interface is fairly large (of order 50 nm), this shift will be considerably smaller. Therefore, the measured increase in phonon energy may be tentatively ascribed to the presence of the heterointerface. These addi-



FIG. 10. The resonant excitation spectra of sample 1 at  $6-7$ T. Uppermost is the nonresonant spectrum at  $B = 7$  T with the laser tuned well above the excitonic emissions.

Apart from the information obtained by the RE experiments on sample 3, we obtained additional results that could only be obtained for the sample with the lowest carrier density. Figure 10 shows PL spectra from sample <sup>1</sup> that were obtained by exact tuning of the laser to the energy of the  $X(n_c=2,h)$ . The uppermost spectrum was recorded at  $B = 7$  T with the laser tuned well above the  $X(n_c=2, h)$  emission and showed only  $(D^0, A^0)$  PL of the GaAs buffer layer. Resonant excitation of the  $X(n<sub>c</sub>=2, h)$  at 7 T led to the observation of additional peaks, inter alia the broad (e<sub>2</sub>,  $A^{0}$ ) peak, separated 19.4 meV from the  $X(n_c=2, h)$ , which can again be attributed to recombination of electrons in the  $n_c = 2$  subband with holes located at acceptors. Upon repeating the RE experiment at lower fields, this PL line shifted within experimental error equivalently to the lowest Landau level of the  $n_c = 2$  subband.

An additional peak with an energy between those of  $(D^0, A^0)$  and  $(e_2, A^0)$  appeared if the  $X(n_c = 2, h)$  was resonantly excited. This peak cannot be attributed to  $(e, A^0)$  recombination in the GaAs buffer layer because the latter PL shifts linearly in magnetic field toward 1.4991 eV at  $B = 7$  T.<sup>47</sup> Furthermore, the attribution to excited acceptor states as a result of selective  $(D^0, A^0)$ pair luminescence (SPL) can also be refuted for two reasons. First, SPL peaks would disappear if the laser were tuned above energies of bound excitons, such as is the case here; second, SPL peaks should be present in the spectra of all samples, especially in that of sample 3 for which the energy of the  $X(n_c=2, h)$  (to which the laser was tuned) lay well below all GaAs buffer layer bound excitons. Calculation of the energy of the  $N = 0$  Landau level of the  $n_c = 1$  subband yielded 1.4972 eV (from 1.4914 +  $7\mu_B/0.070$ , very close to that of the observed peak 1.4964 eV. Therefore, we believe that this peak involves recombination of  $N = 0$   $n_c = 1$  electrons with photoexcited holes. The shift of this peak in the lower-field RE spectra yields  $m^* = 0.070m$ , which corresponds to the effective mass of the  $n_c = 1$  subband in this sample. Because no increase in the fully occupied  $n_c = 1$  electron concentration is possible, an enhanced  $(n_c = 1, h)$  recombination can be observed here because of the enhanced density of photoexcited holes in the 2DEG region upon resonant excitation of the  $X(n<sub>c</sub>=2, h)$ . This enhanced hole density results from the increased probability to excite a valence electron to the  $n<sub>c</sub> = 2$  subband in the RE experiment. To our knowledge, this is the first time that PL from the  $n_c = 1$  subband has been observed in a highcarrier-density single heterojunction without hole confinement. This  $n_c = 1$  PL could not be observed in the two samples with higher carrier densities, presumably because of their greater intrinsic electric field, which acts on the photoexcited hole density.

# IV. CONCLUSIONS

We have presented PL measurements on three single heterojunctions which have two occupied subbands at zero magnetic field. Despite the lack of hole confinement, we observed the  $n_c = 2$  subband excitons  $X(n_c=2, h)$  in all samples, owing to the hybridization between this subband and the Landau level of the  $n_c = 1$ subband in which the Fermi level resides. This manybody interaction, which is maximal at odd filling factors, led to the observation of oscillations with Shubnikov —de Haas periodicity in the intensity of the  $X(n<sub>c</sub>=2,h)$ . Furthermore, we reported magneto-oscillations in both peak energy and peak width which are caused by the crossing of  $n_c = 1$  Landau levels. Apart from the Fermi-edgeinduced peaks  $X(n_c=2, h)$ , which appeared in all samples, a second Fermi-edge-induced peak involving the 2DEG was most clearly observed in the sample with lowest carrier density, most likely originating from the recombination of  $n_c = 2$  subband electrons with the photoexcited light holes  $X(n_c = 2, l)$ . A second-subband exciton splitting, which was maximal at  $v=5$ , was also observed in this sample. This observation points to separate interaction of the two spin states of Landau level  $N = 2$  of the  $n_c = 1$  subband with the second-subband exciton.

Excitation below and above the GaAs band gap was found useful to distinguish PL from the 2DEG and that from the GaAs buffer layer. From this behavior and temperature-dependent measurements, strong indications were found for luminescence of the  $n_c = 3$  subband. Evidence for the formation of a band of localized states between the spin components of a Landau level comes from a maximum intensity at even filling factor in the sample with the highest carrier density.

The zero-field energy of the Fermi level has been found to be 1.515 eV for all samples. This pinning to the energy of the Fermi level in the GaAs buffer layer shows that the photoexcited holes, which participate in the recombination process, are essentially located in the flat-band region of the GaAs. Therefore, the PL energies of the subbands decrease if the electron concentration of the 2DEG increases.

Resonant excitation (RE) was shown to be a useful method for providing additional information on the 2D system. First, RE on the  $X(n<sub>c</sub>=2, h)$  resulted in the appearance of various peaks that were related to the  $X(n_c=2, h)$ . Apart from the direct phonon replica of the  $X(n_c=2, h)$ , recombination was observed of  $n_c=2$  electrons with holes located at acceptors and the corresponding phonon replica therefrom. Second, resonant excitation of the third subband was used to create a nonequilibrium electron concentration in this subband. This led to the observation of an extremely sharp phonon replica of the  $n_c = 3$  subband. The sample with the lowest carrier density showed unprecedented luminescence of the lowest Landau level of the  $n_c = 1$  subband upon resonant excitation of the second-subband excitation.

#### ACKNOWLEDGMENTS

The authors wish to thank H. F. Pen for assistance. This work was supported by the Nederlandse Qrganisatie voor Energie en Milieu (NOVEM). The project was also sponsored by the SCIENCE program of the European Community.

- <sup>1</sup>G. D. Mahan, Phys. Rev. 153, 882 (1967).
- <sup>2</sup>M. S. Skolnick, J. M. Rorison, D. J. Mowbray, P. R. Tapster, S.J. Bass, and A. D. Pitt, Phys. Rev. Lett. 58, 2130 (1987).
- <sup>3</sup>M. S. Skolnick, K. J. Nash, S. J. Bass, P. E. Simmonds, and M. J. Kane, Solid State Commun. 67, 637 (1988).
- 4A. S. Plaut, K. von Klitzing, I. V. Kukushkin, and K. Ploog, in Proceedings of the 20th International Conference on the Phys ics of Semiconductors, edited by E. M. Anastassakis and J. D. Joannopoulos (World Scientific, Singapore, 1990), p. 1529.
- 5A. S. Plaut, I. V. Kukushkin, K. von Klitzing, and K. Ploog, Phys. Rev. B42, 5744 (1990).
- <sup>6</sup>H. Buhmann, W. Joss, K. von Klitzing, I. V. Kukushkin, A. S. Plaut, G. Martinez, K. Ploog, and V. B.Timofeev, Phys. Rev. Lett. 66, 926 (1991).
- 7A. J. Turberfield, S. R. Haynes, P. A. Wright, R. A. Ford, R. G. Clark, J. F. Ryan, J. J. Harris, and C. T. Foxon, Phys. Rev. Lett. 65, 637 (1990); A. J. Turberfield, S. R. Haynes, P. A. Wright, R. A. Ford, R. G. Clark, J. F. Ryan, J.J. Harris, and C. T. Foxon, in Proceedings of the 20th International Conference on the Physics of Semiconductors (Ref. 4), p. 805.
- 8M. Potemski, R. Stepniewski, J. C. Maan, G. Martinez, P. Wyder, and B. Etienne, Phys. Rev. Lett. 66, 2239 (1991).
- <sup>9</sup>D. Heiman, B. B. Goldberg, A. Pinczuk, C. W. Tu, A. C. Gossard, and J. H. English, Phys. Rev. Lett. 61, 605 (1988).
- <sup>10</sup>I. V. Kukushkin, K. von Klitzing, and K. Ploog, Pis'ma Zh. Eksp. Teor. Fiz. 47, 511 (1988) [JETP Lett. 47, 598 (1988)].
- <sup>11</sup>I. V. Kukushkin, A. S. Plaut, K. von Klitzing, and K. Ploog, Surf. Sci. 229, 447 (1990).
- <sup>12</sup>I. V. Kukushkin, K. von Klitzing, K. Ploog, and V. B. Timofeev, Phys. Rev. B40, 7788 (1989).
- <sup>13</sup>H. Buhmann, W. Joss, K. von Klitzing, I. V. Kukushkin, G. Martinez, A. S. Plaut, K. Ploog, and V. B. Timofeev, in Proceedings of the 20th International Conference on the Phys ics of Semiconductors {Ref.4), p. 1585.
- <sup>14</sup>B. B. Goldberg, D. Heiman, M. Dahl, A. Pinczuk, L. Pfeiffer, and K. West, Phys. Rev. B 44, 4006 (1991).
- <sup>15</sup>G. E. W. Bauer, Solid State Commun. 78, 163 (1991).
- <sup>16</sup>V. M. Apal'kov and E. I. Rashba, Pis'ma Zh. Eksp. Teor. Fiz. 53, 420 (1991) [JETP Lett. 53, 442 (1991)].
- $17V$ . M. Apal'kov and É. I. Rashba, Pis'ma Zh. Eksp. Teor. Fiz. 53, 46 (1991) [JETP Lett. 53, 49 (1991)].
- <sup>18</sup>I. E. Perakis and Yia-Chung Chang, Phys. Rev. B 43, 12566 (1991).
- <sup>19</sup>T. Uenoyama and L. J. Sham, Phys. Rev. Lett. 65, 1048 (1990).
- W. Chen, M. Fritze, A. V. Nurmikko, D. Ackley, C. Colvard, and H. Lee, Phys. Rev. Lett. 64, 2434 (1990); M. Fritze, W. Chen, A. V. Nurmikko, and D. Ackley, in Proceedings of the 20th International Conference on the Physics of Semiconduc tors (Ref. 4), p. 825.
- <sup>21</sup>J. F. Mueller, *Phys. Rev. B* 42, 11 189 (1990).
- <sup>22</sup>J. F. Mueller, A. Ruckenstein, and S. Schmitt-Rink, Mod. Phys. Lett. 5, 135 (1991).
- $23F.$  A. J. M. Driessen, S. M. Olsthoorn, T. T. J. M.

Berendschot, H. F. Pen, L. J. Giling, G. A. C. Jones, D. A. Ritchie, and J. E. F. Frost, Phys. Rev. B45, 11 823 (1992).

- 24J. J. Harris, D. E. Lacklison, C. T. Foxon, F. M. Selten, A. M. Suckling, R. J. Nicholas, and K. W. J. Barnham, Semicond. Sci. Technol. 2, 783 (1987).
- 25R. M. Kusters, Phys. Rev. B46, 10207 (1992).
- <sup>26</sup>R. M. Kusters, J. Singleton, G. Gobsch, G. Paasch, D. Schulze, F. A. Wittekamp, G. A. C. Jones, J. E. F. Frost, D. C. Peacock, and D. A. Ritchie, Superlatt. Microstruct. 9, 55 (1991).
- 27W. H. Knox, D. S. Chemla, and G. Livescu, Solid-State Electron. 31, 425 (1988).
- <sup>28</sup>B. K. Ridley, Rep. Prog. Phys. 54, 169 (1991).
- <sup>29</sup>T. Ando, J. Phys. Soc. Jpn. 43, 1616 (1977).
- 30Th. Englert, D. C. Tsui, A. C. Gossard, and Ch. Uihlein, Surf. Sci. 113,295 (1982).
- <sup>31</sup>R. Fletcher, E. Zaremba, M. D'Iorio, C. T. Foxon, and J. J. Harris, Phys. Rev. 8 41, 10649 (1990).
- 32D. M. Larsen, in Polarons in Ionic Crystals and Polar Semiconductors, edited by J. T. DeVreese (North-Holland, Amsterdam, 1972).
- 3W. Chen, M. Fritze, A. V. Nurmikko, M. Hong, and L. L. Chang, Phys. Rev. B 43, 14738 (1991).
- <sup>34</sup>J. Aaviksoo, I. Reimand, V. V. Rossin, and V. V. Travnikov, Phys. Rev. B45, 1473 (1992).
- 35M. Shinada and S. Sugano, J. Phys. Soc.Jpn. 21, 1936 (1966).
- <sup>36</sup>A. M. Kazaryan, and E. M. Kazaryan, Fiz. Tekh. Poluprovodn. 11, 1383 (1977) [Sov. Phys. Semicond. 11, 813 (1977)].
- 37F.-Y. Juang, Yasunobu Nashimoto, and Pallab K. Bhattacharya, J. Appl. Phys. 58, 1986 (1985).
- $38$ The illustrative example of the simple triangular potential well was treated by W. Zawadzki in Two-dimensional Systems, Heterostructures, and Superlattices, edited by G. Bauer, F. Kuchar, and H. Heinrich, Solid State Sciences Vol. 53 (Springer, Berlin, 1984), p. 2.
- $39B.$  I. Shklovskii and A. L. Efros, *Electronic Properties of Doped* Semiconductors (Springer, Berlin, 1984).
- 40P. J. Dean and M. S. Skolnick, J. Appl. Phys. 54, 346 (1983).
- $41$ This method is used (inter alia) for identifying residual donor species. The intensities of so-called "two-electron" satellite peaks are increased if the  $(D^0, X)$  peak is resonantly excited (Refs. 40 and 42).
- <sup>42</sup>F. A. J. M. Driessen, H. G. M. Lochs, S. M. Olsthoorn, and L. J. Giling, J. Appl. Phys. 69, 906 (1991).
- <sup>43</sup>I. V. Kukushkin, K. von Klitzing, and K. Ploog, Phys. Rev. B 37, 8509 (1988).
- <sup>44</sup>B. J. Skromme, S. S. Bose, and G. E. Stillman, J. Electron. Mater. 15, 345 (1986).
- 45J. S. Blakemore, J. Appl. Phys. 53, R123 (1982).
- <sup>46</sup>M. A. Stroscio, K. W. Kim, and J. C. Hall, Superlatt. Microstruct. 7, 115 (1990).
- 47S. Zemon, P. Norris, E. S. Koteles, and G. Lambert, J. Appl. Phys. 59, 2828 (1986).