Influence of magnetic fields on an extremely narrow exciton line in a high-carrier-density heterojunction

F. A. J. M. Driessen, S. M. Olsthoorn, T. T. J. M. Berendschot, H. F. Pen, and L. J. Giling
Department of Experimental Solid State Physics, Research Institute for Materials, University of Nijmegen, Toernooiveld, NL 6525 ED Nijmegen, The Netherlands

G. A. C. Jones, D. A. Ritchie, and J. E. F. Frost
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 OHE, United Kingdom
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High-resolution magnetophotoluminescence measurements are reported on a GaAs-$Al_xGa_{1-x}As$ heterojunction that has two occupied subbands at zero magnetic field. A very efficient, extremely narrow-band luminescence involving the two-dimensional electron gas is observed above a certain critical field strength in spite of the large built-in electric field acting on the photoexcited holes. The photoluminescence peak shows pronounced magneto-oscillations in peak energy, as well as in luminescence intensity and peak width. The luminescence is attributed to the exciton of the second subband, which hybridizes with the Landau level of the first subband in which the Fermi level resides. This many-body interaction is most efficient at odd filling factors, i.e., if the Fermi level lies in the extended states. The oscillations in photon energy and full width at half maximum originate from $n_c=1$ Landau-level crossings with the lowest $n_c=2$ Landau level. The optical data show that the exciton of the second subband is consecutively populated and depopulated. Tilted-field measurements enable the determination of the intrasubband diamagnetic shift in a parallel field $\delta E_{12}/\delta B$.

INTRODUCTION

Magneto-optical studies of two-dimensional electron gases (2DEG's) in semiconductor heterostructures have revealed a variety of information on, for example, Landau quantization,[1] electron-electron interactions,[2] and both the integer and fractional quantum hall effect.[3–5] In order to observe photoluminescence (PL) from the 2DEG a large concentration of holes is required. This has been either by realized hole confinement in a modulation-doped quantum well (MDQW)[1,6] or by growing an additional δ layer of acceptors at a well-defined distance from the heterointerface in the GaAs buffer layer.[2,7,8] In other work, structures with a thin GaAs buffer layer were chosen in order to prevent bulk luminescence from dominating the spectrum.[9,10]

Recently, Chen *et al.*[11] reported very pronounced magneto-oscillations in the PL intensity of an excitonlike interband transition in an (In,Ga)As MDQW in which the Fermi level was just below the $n_c=2$ subband. This behavior, which had $1/B$ periodicity, originated from a many-body interaction between the $n_c=2$ conduction subband and the Landau level of the $n_c=1$ conduction subband in which the Fermi level resided. The interaction was found to be largest when the Fermi level lay within the extended states, i.e., at odd-integer Landau-level filling factors. This is known as the optical Shubnikov–de Haas (OSdH) effect. Using three-band model calculations without including magnetic field, Mueller[12] showed that a reduction of the separation between an unoccupied second subband and the Fermi level caused an excitonic enhancement owing to hybridization between the $n_c=1$ Fermi-edge resonance[13] and the excitonic resonance of the $n_c=2$ subband. This led to a strong enhancement of the optical matrix element for this mixed state, even in the absence of a real $n_c=2$ population because virtual $n_c=2$ excitons are formed as a result of intrasubband scattering. Moreover, it is also the case under conditions of PL experiments because of the presence of photoexcited nonequilibrium electrons and holes.

In this paper we present magnetophotoluminescence spectra of a high-carrier-density GaAs-$Al_xGa_{1-x}As$ heterojunction with two populated subbands for which the holes were subjected to a very strong built-in electric field. This is in contrast to the previously mentioned magneto-optical publications on 2D systems, in which holes were either localized or trapped in thin buffer layers, quantum-well structures, or at additional acceptor sites. An extremely narrow PL line was observed when the strength of the magnetic field first caused depopulation of the second subband. This emission tended to dominate the spectrum at increasing fields. As with the emission reported in Ref. 11, this luminescence involves second-subband electrons and photoexcited holes with a small exciton binding energy, and is caused by the many-body interaction of the Fermi-edge electrons with the lowest Landau level of the second subband. Apart from the oscillations in PL intensity with SdH periodicity in a MDQW with one occupied subband which were previously reported by Chen *et al.*,[11] we have also been able to resolve oscillations in both energy and width of the luminescence as a result of the extreme sharpness of the PL line (≈30 times narrower than that in Ref. 11). The small width of the emission also enables us clearly to distinguish between various exciton peaks originating from the buffer layer and those from the 2D luminescence. Furthermore, we report the determination of the intrasubband diamagnetic shift, which was obtained from tilted-field measurements.

EXPERIMENTAL DETAILS

The experiments were carried out on a high-carrier-density GaAs-$Al_xGa_{1-x}As$ heterojunction grown by molecular beam epitaxy. The structure was grown on a semi-insulating GaAs substrate and consisted of a 20-period GaAs-AlAs superlattice buffer, a 1-μm-thick undoped GaAs buffer layer, an 80-nm $Al_{0.33}Ga_{0.67}As$ layer, and finally, a 10-nm GaAs capping layer. The $Al_{0.33}Ga_{0.67}As$ was undoped except for one monolayer 10 nm from the heterointerface, which was doped with 5×10^{16} m^{-2} Si atoms on Ga sites. Monolayer doping of the barrier of a GaAs-$Al_xGa_{1-x}As$ heterojunction results in higher carrier densities n and increased mobilities μ of the 2DEG.[14] Values of these quantities for the aforementioned heterojunction measured in the dark were $n_{\text{dark}}=6.4\times10^{15}$ m^{-3} and $\mu_{\text{dark}}=39$ $m^2V^{-1}s^{-1}$ for our sample;[15] after saturation of the persistent photoconductivity[15] by illumination with a red LED the values were $n_{\text{light}}=11.3\times10^{15}$ m^{-3} and $\mu_{\text{light}}=39$ $m^2V^{-1}s^{-1}$.[16] Other sample parameters can be found in the work of Kusters *et al.*[15,17]

PL measurements were taken with the sample mounted in a strain-free way in the bore of a 7 T split-pair superconducting magnet. Optical excitation was provided by a tunable cw ring dye laser using Styryl 9 as dye at typical excitation densities of 10^3 W m^{-2}. The luminescence was dispersed by a 0.6-m double monochromator with 1200-lines/mm gratings and was detected by a cooled photomultiplier tube with a GaAs photocathode. The measurements were mostly performed in a Faraday configuration with $\mathbf{k}\|\mathbf{B}\|[100]$. The persistent photoconductivity[16,18] was saturated during the measurements because the laser energy was slightly higher than that of the GaAs band gap. From this it may also be inferred that both ionization of deep donor states (*DX* centers) in the $Al_xGa_{1-x}As$ and generation of electron-hole pairs in the GaAs occurred. A reduction of the carrier density due to neutralization of *DX* centers in the $Al_xGa_{1-x}As$ under continuous photoexcitation (as reported by Kukushkin *et al.*[19]) did not influence our measurements because in our case the excitation energy was too low to neutralize the *DX* centers.

RESULTS AND DISCUSSION

Figure 1 shows the excitonic part of the PL spectrum of the sample at various magnetic-field strengths recorded at $T=1.5$ K. The zero-field spectrum is dominated by the PL of the GaAs buffer layer; it shows free excitons *FX*, as well as excitons bound to residual acceptors (A^0,X) and donors (D^0,X). The dominance of the PL signal of the GaAs buffer layer over any possible luminescence involving confined electrons at the GaAs-$Al_xGa_{1-x}As$ interface is, of course, due to the large built-in electric field near the heterointerface. Hence,

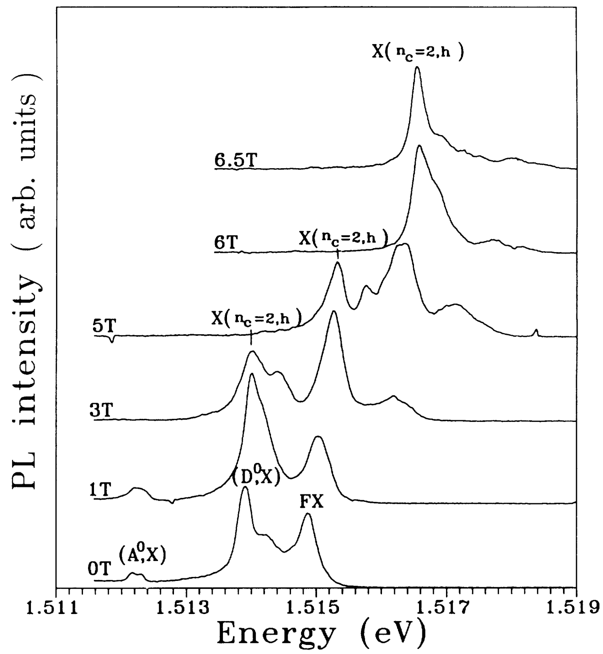

FIG. 1. Excitonic parts of the PL spectra at various magnetic-field strengths.

photoexcited holes will quickly move away from the interface to the GaAs buffer layer where recombination with photoexcited electrons can occur.

This situation changes if a magnetic field is applied. An intense and narrow PL line, which is absent in the spectrum of bulk GaAs and labeled $X(n_c=2,h)$, is observed if the magnetic field has passed a certain critical field value of about 2.5 T. As can be seen in Fig. 2, the energy of this PL peak shows oscillations which are periodic in $1/B_\perp$, where $B_\perp$ is the component of the field perpendicular to the interface. That the periodicity of

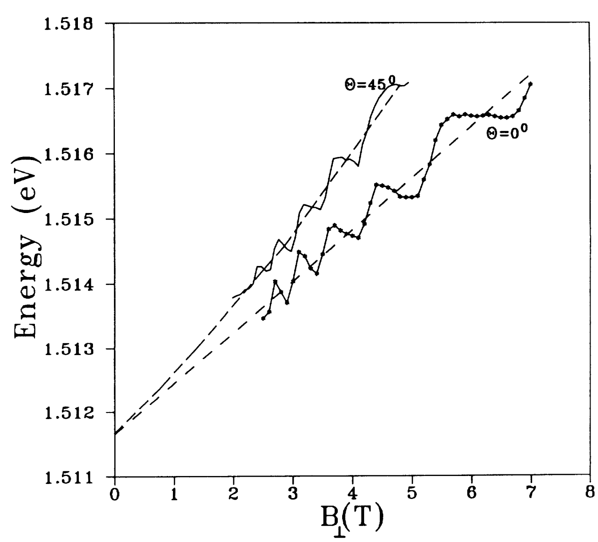

FIG. 2. Energy position of peak $X(n_c=2,h)$ versus the perpendicular component of magnetic field. The upper line shows the behavior of the peak if the field is tilted 45° with respect to the 2DEG.

these oscillations depends only on the perpendicular component is shown by the upper trace in Fig. 2 in which the magnetic field was at an angle of 45° with respect to the 2DEG. Changes in photon energy in the tilted field will be discussed later in this paper. Therefore, despite the large built-in electric field acting on the holes, luminescence of the 2DEG is observed in a magnetic field. As the field strength increases, the intensity of this line becomes so strong that it eventually dominates the entire spectrum, as is shown in Fig. 1.

Apart from the well-resolved oscillations in photon energy, magneto-oscillations are also observed both in peak intensity and in peak width (see Fig. 3). Maxima in the peak intensity occur at odd filling factors. A similar periodically enhanced PL signal at odd filling factors was observed by Chen *et al.*[11] in a MDQW with one occupied subband. This PL signal resulted from nonequilibrium $n_c=2 \rightarrow n_v=1$ exciton-like emission, which hybridized with the energetically proximate 2DEG. The strength of this many-body (Coulomb) interaction was maximal if E_F lay in the extended states of the particular Landau level (LL) of the lowest subband.

In our sample the electron concentration of the second subband is $n_2=1.0\times10^{15}$ m^{-2} at zero field, and the zero-field Fermi level is located, therefore, approximately 3.5 meV above $n_c=2$.[15] In a magnetic field the position of E_F oscillates, owing to the highly singular density of states and the field proportionality of the Landau-level degeneracy.[20,21] Using the relationship between magnetic field and Fermi level in Ref. 21, we estimate that E_F shifts below the lowest spin split LL of the second subband at a field of about 2.5 T. Consequently, above this field strength hybridization can be observed between the exciton of the lowest LL of the second subband and the particular LL of the $n_c=1$ subband in which E_F resides. Because the luminescence is first observed (or at least gains enormously in strength) above the field strength at which the second subband initially depopulates and, furthermore, because maxima in PL intensity are observed at odd ν, we expect the origin of the PL signal to be similar to that described by Chen *et al.*[11] Apart from the fact that our sample has two populated subbands, the most important physical difference from the system of Chen *et al.*[11] is that in our system the photoexcited holes are not located close to the interface in a hole subband and, as a result, the magneto-optical characterization of single heterojunctions with thick buffer layers is possible. The exciton binding energy between the $n_c=2$ subband electrons and the holes is expected to be considerably smaller than that in the work of Chen *et al.*[11] due to the lack of hole confinement.

Assuming that the luminescence involves the exciton of the lowest LL of the second subband, hereafter denoted $X(n_c=2,h)$, then the Landau fan diagram of the sample can be calculated as follows. It is shown below that the exciton binding energy is negligible and, therefore, that the dashed line in Fig. 4 would reflect the photon energy of the $n_c=2$ subband were it not influenced by the LL's of the $n_c=1$ subband. A second subband effective electron mass of $m_2^*=0.069m_0$ (where m_0 is the free-

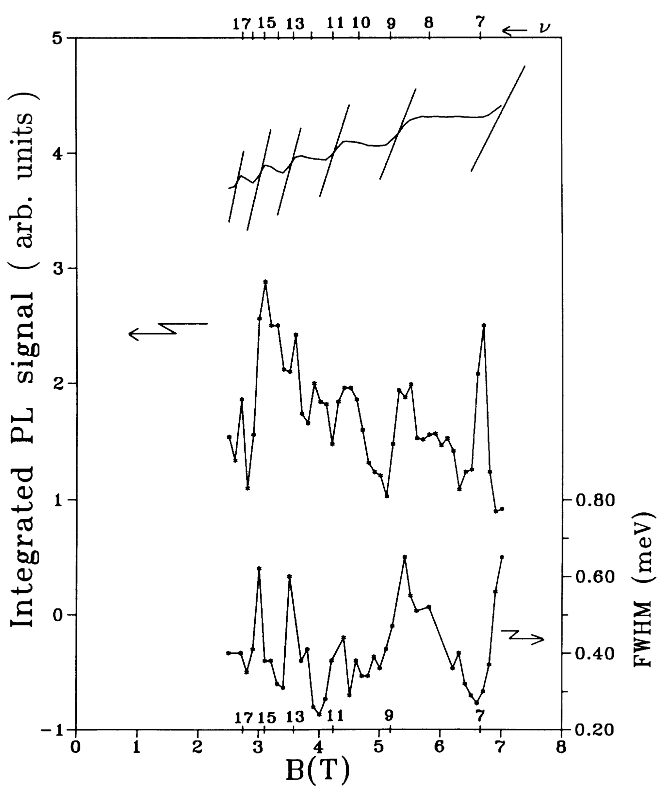

FIG. 3. Integrated PL intensity (left axis) and full width at half maximum (right axis) of peak E_f as a function of magnetic field. For clarity, the photon-energy data of Fig. 2 are also shown in this figure. The top axis shows the filling factor ν; at the bottom axis odd ν values are repeated.

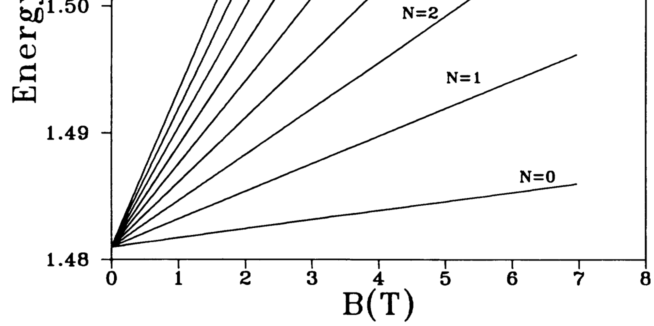

FIG. 4. Measured position of $X(n_c=2,h)$, the Landau-level fan diagrams, and the calculated position of E_F versus the magnetic field in the field region of interest (bold line). The dashed line is the lowest Landau level of the $n_c=2$ subband. The top axis shows the filling factor ν.

electron mass) was calculated from the slope of the $n_c=2$ LL. Linear extrapolation of this lowest $n_c=2$ LL yields a zero-field energy of 1.511 65 eV for $X(n_c=2,h)$.

The zero-field energies of the Fermi level E_F and the subband $n_c=1$ follow from the relationships $E(n_c=2)-E(n_c=1)=30.3$ meV and $E_F-E(n_c=2)=3.5$ meV, as determined by magnetotransport measurements.[15]

Cyclotron-resonance (CR) measurements revealed an effective electron mass $m_1^*=0.074m_0$ for this sample. This value cannot directly be used because it is known that effective masses as measured by magneto-optics are generally higher than those obtained by CR owing to the sensitivity of magneto-optics to electron-electron interactions.[3] In contrast, the energy separation between a full and empty LL as measured by CR is not affected by electron-electron interactions.[22] Plaut *et al.*[3] measured effective masses as a function of electron concentration n (up to $n=7\times10^{15}$ m^{-2}) by magneto-optics. At high carrier concentrations the effective mass increased owing to effects of nonparabolicity. The energy dependence of the effective mass is described by the relationship[23,24] $m^*=m_0/(1+2\kappa_2\varepsilon/E_g)$, where the relative Fermi level $\varepsilon=E_F-E_g$ is proportional to n, and the coefficient $\kappa_2\approx-1.4$ (Ref. 25) is a measure of the low-energy nonparabolicity. Because the second term in the denominator is much less than unity, the equation can be written as $m^*\approx m_0(1-2\kappa_2\varepsilon/E_g)$. We used this relationship to extrapolate the magneto-optical effective mass $0.079m_0$ for our sample with $n=11.3\times10^{15}$ m^{-2}. This value for the effective electron mass was used to construct the Landau fan diagram of the $n_c=1$ subband.

The calculated energy of E_F at $T=0$ assuming δ-function-shaped Landau levels is now shown by the bold line in Fig. 3 in the field range of interest. It is seen that field intervals in which the Fermi level follows a Landau level of $n_c=1$ (i.e., regions with a depopulated second subband) are consecutively followed by field intervals in which E_F populates the lowest LL of $n_c=2$.

The plot shows that at filling factors $\nu=7$ and 9 the second subband is not populated. Hence, effective coupling occurs between the extended states of the uppermost occupied $n_c=1$ LL and $X(n_c=2,h)$, thus resulting in the sharp intensity maxima at $\nu=7$ and 9 shown in Fig. 3. At slightly higher values of the magnetic field, maxima in the width of the PL line are observed, namely at $B=5.4$ T and $B\geq7.0$ T for $\nu=9$ and 7, respectively. These field values correspond exactly to the crossing between the $N=4$ and 3 Landau levels of $n_c=1$ with the $X(n_c=2,h)$. At the crossing points of these levels both the energy and width of the $X(n_c=2,h)$ are most influenced by the $n_c=1$ LL's, the latter having a much larger width than the narrow exciton emission. The photon energy of $X(n_c=2,h)$ is influenced least at field values in between two successive crossings. The field values of minimum influence were used to construct the dashed line in Fig. 4. This leads to a perfectly linear dependence of the photon energy upon magnetic field with no detectable quadratic behavior. From this it can be concluded (as was claimed above) that the exciton binding energy is very small.

It is seen in Fig. 4 that below $B\approx4$ T the second subband is again populated at odd filling factors (cf. $\nu=13$, 15, and 17). This follows both from the crossing levels in the fan diagram, as well as from the lower field values at which maxima in line width occur compared to the field values of maximal intensity. The origin of these maxima in PL intensity, which are less pronounced at $\nu=13$, 15, and 17, are partly caused by oscillations in the population of the $n_c=2$ subband with magnetic field, as was pointed out by Skolnick, Simmonds, and Fisher[26] for an MDQW. An enhancement of the PL intensity is expected if this state is populated as a result of the greater spatial extension of the $n_c=2$ subband wave function compared to that of $n_c=1$. These intensity oscillations due to population of the second subband are also expected for even filling factors.[5] In fact, we also observe less pronounced maxima in the intensity at even filling factors ($\nu=8,10,12,14$); however, since the maxima at odd ν are considerably more intense, we conclude that in spite of the slight population of $n_c=2$ at $\nu=13$, 15, and 17, coupling with the delocalized electrons still occurs.

It was already seen in Fig. 2 that the oscillatory behavior in photon energy of $X(n_c=2,h)$ depended only on the magnetic-field component which was perpendicular to the 2DEG. It is also seen in Fig. 2 that the photon energy of the second subband exciton is higher in the tilted-field measurement. This additional shift in parallel magnetic field is obtained from the difference between the two dotted lines because $B_\perp=B_\parallel$ here. If we assume a quadratic relationship between $B_\parallel$ and $E_{21}(B_\parallel)$, where E_{21} is the difference between the two subbands,[20] namely,

$$E_{21}(B_\parallel)=E_{21}+\alpha B_\parallel^2\ ,$$

then from our data α is found to be $(8.0\pm0.5)\times10^{-5}$ eV T^{-2}. This value corresponds within experimental error with the value $\alpha=(8.6\pm0.4)\times10^{-5}$ eV T^{-2} which we calculated using the experimental data of Kusters,[15] who performed parallel-field magnetoresistance measurements on the same sample.[27] This correspondence for the intrasubband diamagnetic shift of $E_{21}(B_\parallel)$ is a further strong indication that the luminescence involves the second subband and that exciton binding energies are small.

We have already mentioned above that the extrapolation of the photon energy of $X(n_c=2,h)$ to zero field reveals a value 1.511 65 eV. At high power densities P, a peak of low intensity is observed at the slightly lower energy of 1.5112 eV in the zero-field PL spectrum (Fig. 5). It is possible that this peak is the $X(n_c=2)$ peak shifted to lower energy owing to changes in the confining potential. The electric field in the GaAs buffer layer is vanishingly small at higher P as a result of the neutralization of the depletion charge, and this results both in a greater spatial extension of the $n_c=2$ subband and lower E_{21}. This "flattening" of the bands in the GaAs layer also causes an enhanced hole density near the 2DEG, which results in an increased wave-function overlap with the $n_c=2$ electrons. The reduction of depletion charge is also reflected by the relative enhancement of the (A^0,X)

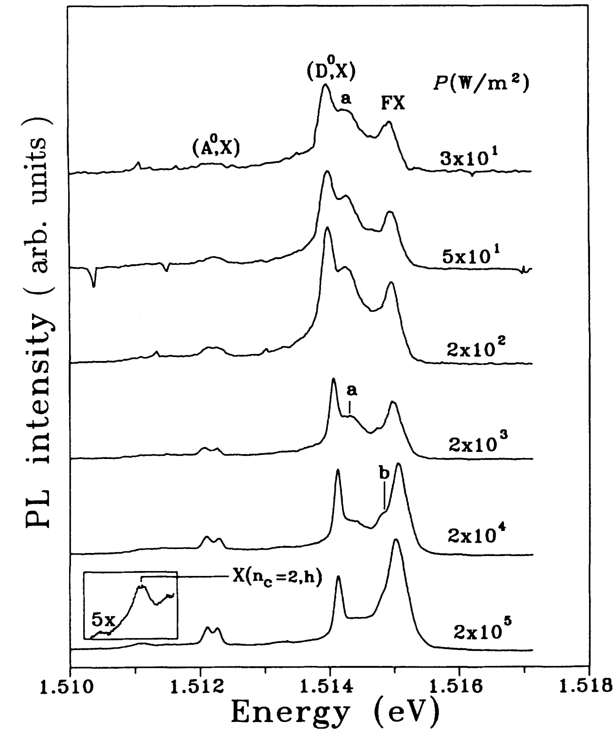

FIG. 5. Zero-field spectrum as a function of laser power density; the enlarged part of the spectrum at the bottom shows the exciton luminescence of the populated $n_c = 2$ subband.

peak from the GaAs buffer layer compared to other buffer-layer PL emission. The ionized acceptors in the depletion region at low P are neutralized at higher excitation densities of the laser light, so that a bound state between a neutral acceptor and an exciton becomes possible. Further 3D peaks correspond to donor-bound excitons (D^0,X) in the buffer layer, which show saturation effects at high P owing to the limited amount of donors present in the buffer layer. In contrast, no such saturation occurs for the FX emission.

Two other emissions, which are labeled a and b in Fig. 5, show interesting behavior as a function of excitation density. With increasing P, peak a shifts to slightly higher energy and shows a less pronounced increase in intensity than either (A^0,X), (D^0,X), or FX. A possible explanation for the origin of this peak is that it directly reflects the observation of PL from electrons at the Fermi level. Mueller[12] showed that PL from Fermi electrons could be observed if the Fermi level was located just below the second subband. In our sample at zero field the Fermi level is located below the third subband, so a many-body interaction is possible between the Fermi electrons and the virtual exciton of the third subband. At the highest excitation densities used the third subband is likely to be populated, thereby leading to a suppression of the E_F peak and the appearance of the $n_c = 3$ exciton. In comparison with the $n_c = 1$ and 2 subbands, the spatially extended $n_c = 3$ subband has a larger wave-function overlap with the photoexcited holes. Provided that the oscillator strength that results from this overlap is large enough, peak b at 1.5147 eV in Fig. 5 can be tentatively assigned to the transition between $n_c = 3$ subband electrons and photoexcited holes.

An alternative origin for peak b is via the formation of molecular excitons (or biexcitons: a bound state between a pair of free excitons) in the GaAs buffer. This tentative attribution is based on the appearance of peak b at high excitation density and the photon energy of 1.5147 eV which correspond to an additional binding energy of 0.4 meV between the two excitons. This difference is close to the calculated 0.35 meV for molecular excitons in GaAs.[28]

CONCLUSIONS

We have investigated the magneto-optical behavior of a single heterojunction with two populated subbands. Extremely narrow exciton luminescence involving $n_c = 2$ electrons and photoexcited holes is reported above field values at which the Fermi level shifts for the first time below the lowest Landau level of the $n_c = 2$ subband. Magneto-oscillations in intensity of the exciton peak are explained by a combination of population effects and, more importantly, a many-body interaction between the $n_c = 2$ exciton and the extended states of the $n_c = 1$ Landau level in which E_F resides. Oscillations in photon energy and width of the exciton peak result from crossing of $n_c = 1$ Landau levels with the exciton of the lowest Landau level of the $n_c = 2$ subband. Finally, the intrasubband diamagnetic shift has been determined from tilted-field measurements.

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$$\alpha B^2_{\parallel,\mathrm{dep}} = 2[E_F - E(n_c=2)]_{B_\parallel=0}$$

was used. With $B_{\parallel,\mathrm{dep}} = 9.0\pm0.1$ T and $E_F - E(n_c=2) = 3.5\pm0.5$ meV, this yields $\alpha = (8.6\pm0.4)\times10^{-5}$ eV T^{-2}.

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