

Magnetoluminescence of the two-dimensional electron-hole fluid

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Band-gap renormalization due to the presence of a dense electron-hole plasma in GaAs quantum wells is studied by time-resolved luminescence and magnetoluminescence spectroscopy. We show that heterostructures with long carrier lifetimes exhibit saturation of the band-gap renormalization in a magnetic field, in accord with previous results. However, the usual carrier-density-dependent gap shrinkage in a magnetic field is observed in multiple quantum wells with short carrier lifetimes. The strong dependence of the magneto-optical properties on the carrier lifetimes indicates that two different phases can be formed in the plasma at high magnetic field: a highly correlated phase, analogous to the condensed electronic or excitonic state at the equilibrium density, in quantum wells with sufficiently long carrier lifetimes; and a free-carrier gas in heterostructures with short lifetimes. In the former state, the band gap is found to renormalize by an amount equal to twice the exciton binding energy, independent of the actual photogeneration rate. On the other hand, condensation processes are prevented in the short-lived phase, thus resulting in the expected density-dependent band-gap renormalization.

I. INTRODUCTION

In recent years a great deal of interest has been devoted to the optical properties of semiconductor quantum wells (QW's) in high magnetic field.¹ These studies have clarified the impact of the Landau-level quantization on the two-dimensional energy dispersion of the QW. The magnetoexciton binding energy in GaAs multiple quantum wells (MQW's),²⁻⁴ the in-plane valence-band masses,^{2,3} the Landau-level broadening,^{1,5} and the oscillation of the Fermi level as a function of the filling factor⁶⁻¹¹ have been investigated both experimentally and theoretically. The investigations have been performed in the limit of a low-density electron or exciton gas. However, the physics of a dense two-dimensional electron-hole plasma (EHP) in a magnetic field has not been widely studied. Only recently has it been shown that the application of strong magnetic fields to a dense two-dimensional electron-hole gas results in a variety of interesting effects associated with the modified energy spectrum of the electronic states and with the contribution of many-body interactions.^{12,13}

In a magnetic field the continuous density of states for electrons and holes is split into discrete Landau levels separated by the cyclotron energy (which is different for electrons and holes) and with a degeneracy $2eB/h$ (which is the same for electrons and holes). The radiative-recombination processes in a magnetic field involve transitions between electron and hole subband Landau levels. The spectral shape of the luminescence therefore depends on the Landau quantization of the two-dimensional density of states and on the chemical potential μ of the electron-hole population. In addition, lifetime effects

govern the possibility of phase transitions, such as the transformation of an electron-hole plasma into an electron-hole liquid,^{14,15} and these effects may be changed drastically in a magnetic field. Up to now two limiting situations have been treated extensively: (i) the weak-magnetic-field approximation, in which many Landau levels are filled by a relatively low-density electron-hole population (i.e., $\hbar\Omega \ll \mu$, where $\Omega = eB/m$ is the cyclotron frequency); and (ii) the ultraquantum limit ($\hbar\Omega \gg \mu$), usually achieved in strong magnetic fields, where carriers are confined in their lowest Landau level. In the first case, oscillatory phenomena in the magnetoluminescence spectra are observed as the number of filled Landau levels decreases with increasing magnetic field. In the second case, many-body interactions become dominant, as in the fractional quantum Hall effect.

In this work we investigate the more complex situation of a very-high-density two-dimensional electron-hole population ($n \simeq 10^{13} \text{ cm}^{-2}$) in a strong static magnetic field ($B = 20 \text{ T}$). Under these conditions the inequality $\hbar\Omega \ll \mu$ is satisfied because of the extremely high carrier density, which results in large filling factors despite the large splitting of the Landau levels. The important consequence of this situation is that both the many-body interactions in the dense carrier gas and the magnetic-field-induced modification of the density of states must be taken into account. From the theoretical point of view, state-of-the-art calculations have shown the oscillatory behavior of the exchange and correlation contributions to the quasiparticle self-energies^{6,10} and the enhancement of the electron-hole correlation in magnetic fields.¹⁶ In addition, the phase transition of the quasi-two-dimensional neutral electron-hole plasma into an excitonic conden-

sate^{16,17} or into an electron-hole liquid¹⁵ has been predicted, which should result in significant modifications of the magneto-optical spectra. From a practical point of view, performing experiments on the emission of two-dimensional EHP in high magnetic fields offers the additional advantage that the spectrum of the density of states consists of a series of generally broadened Landau levels separated by gaps. Therefore there are more singularities in the density of states than only the band-gap edge, as in the case at zero magnetic field. It is thus possible to obtain more direct and precise information from the experiments than in the case at zero field. These facts have been exploited by Potemski *et al.*^{12,13} in experiments of luminescence under high excitation. They have observed a clear Landau-level quantization in the emission spectra, which did not show any dependence on the excitation intensity. From these data they concluded that the band-gap renormalization saturated at a value which was found to be twice the exciton binding energy. Furthermore, a clear many-body mass enhancement of the effective mass was reported. In order to investigate these fascinating properties and to provide deeper insight into the many-body interactions in the presence of a strong magnetic field we have carried out systematic magnetoluminescence and time-resolved luminescence experiments under high excitation intensity in suitably designed GaAs/Al_xGa_{1-x}As multiple-quantum-well samples. We show that band-gap renormalization (BGR) in a magnetic field does occur in heterostructures having short carrier lifetimes. Conversely, saturation of the BGR is observed in heterostructures having long carrier lifetimes, probably due to the formation of a constant-density exciton condensate or electron-hole liquid.

This paper is organized as follows. In Sec. II we describe the experimental techniques. In Sec. III we present the results of time-resolved luminescence and time-integrated magnetoluminescence. In Sec. IV we discuss our data and draw our conclusions. Details of the calculations of the exchange energy due to inter-Landau- and intra-Landau-level interactions are shown in the Appendix.

II. EXPERIMENT

The investigated samples are high-quality GaAs/Al_{0.36}Ga_{0.64}As multiple quantum wells grown by molecular-beam epitaxy on a (001) GaAs substrate. The samples consist of 25 periods with a 10.6-nm well width and 15.2-nm barrier width. The structural parameters have been determined by high-resolution double-crystal x-ray diffraction. Two different configurations have been adopted. Sample 1 consists of 25 periods directly grown on the semi-insulating GaAs buffer layer. Sample 2 comprises a 1- μ m-thick Al_{0.36}Ga_{0.64}As barrier layer next to the substrate acting as optical confiner and then the 25-period MQW. The barrier layer provides a negative refractive-index discontinuity of about 7%, resulting in a favorable optical confinement of the emitted luminescence and in high stimulated-emission efficiency. Samples 1 and 2 correspond to samples 2 and 6 of our previous work,¹⁸ respectively. The time-resolved lumines-

cence measurements are performed adopting the same backscattering geometry without a magnetic field. The excitation source is the second-harmonic beam of an amplified, actively and passively mode-locked neodymium-doped yttrium aluminum garnet (Nd:YAG) laser, providing 25-ps full width at half maximum pulses at 5-Hz repetition rate and about 100-MW cm⁻² peak power density. The luminescence is dispersed by a 25-cm spectrometer with a two-dimensional readout system consisting of a single-shot streak camera and a cooled charge-coupled-device (CCD) detector. The time resolution of the system is about 20 ps. The magnetoluminescence experiments are performed at 2 K with samples immersed in superfluid helium and in magnetic fields up to 20 T, using Bitter coils. All the measurements are performed with the magnetic field parallel to the carrier confinement direction. A pulsed N₂-pumped dye laser is used as an excitation source. The dye head provides 5-ns pulses at a 10-Hz repetition frequency with maximum power densities of the order of a few MW cm⁻² after focusing. The luminescence is collected in backscattering geometry from the sample surface and is analyzed by a 1-m single monochromator equipped with a photomultiplier tube and lock-in amplifier.

III. RESULTS

A. Zero-field high-excitation-intensity luminescence

We first summarize the results of the optical measurements carried out at zero magnetic field under the same excitation conditions as for the magneto-optic investigations described in Sec. III C. Two basically different radiative-recombination processes can be investigated depending on the actual optical confinement of the emitted luminescence within the highly excited heterostructure.¹⁸ MQW heterostructures with low optical confinement (sample 1), directly grown onto the absorbing GaAs substrate, exhibit broad emission bands due to the progressive filling of the higher quantized states as the photogeneration rate is increased. On the other hand, MQW heterostructures grown on optically confining cladding layers (such as sample 2) or consisting of a large number of wells show a sharp stimulated emission evolving on the low-energy side of the fundamental heavy-hole-exciton transition. Under this condition no band-filling emission is observed, indicating the saturation of the spontaneous emission¹⁹ and shortening of the radiative lifetime.^{20,21} These results are exemplified in Figs. 1(a) and 1(b), where we show the intensity dependence of the electron-hole-plasma emission in samples 1 and 2, respectively. The luminescence spectra of sample 1 show evidence of the filling of high-energy subbands in the well, above the second electron-to-heavy-hole transition. In addition, a sharp electron-hole plasma emission from the bulk-GaAs buffer layer is observed around 820 nm. The peaks observed at the main intersubband transitions from the quantum well are considerably smeared out when the luminescence is spatially resolved or collected through a pin hole placed in the center of the excitation spot.¹⁸ Whether the peak at the same energy of the exciton ob-

served under low-excitation conditions witnesses the coexistence of excitons and electron-hole plasma, or can purely be attributed to the spatial inhomogeneity of the carrier distribution, is still a matter of controversy. Potemski *et al.*¹² have claimed that the saturation of this peak in a magnetic field strongly indicates its intrinsic origin. Furthermore, at certain high-magnetic-field values the excitonic peak is at very different energies than the expected lowest Landau level in the EHP. Nevertheless, they never observed a peak at energies corresponding to this lowest Landau level. Cingolani *et al.*¹⁸ have claimed that the peak at the energy of the exciton can entirely be explained on the basis of spatial inhomogeneity of the carrier-density profile. However, for the following,

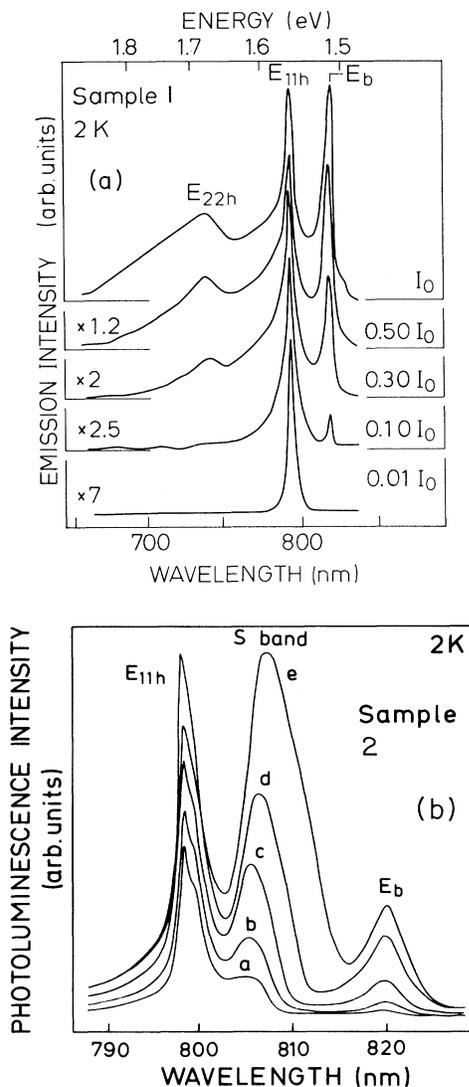


FIG. 1. (a) High-excitation-intensity photoluminescence spectra of sample 1 at different photogeneration rates ($I_0 \approx 2 \text{ MW cm}^{-2}$). The temperature is 2 K. (b) Same as (a), but for sample 2. The excitation intensities are I_0 (curve e), $0.5I_0$ (curve d), $0.3I_0$ (curve c), $0.1I_0$ (curve b), and $0.05I_0$ (curve a).

this different interpretation does not affect the main conclusions of this work.

The energy position of the observed emission bands in Fig. 1(a) agrees well with the low-intensity photoluminescence excitation spectrum. From the line shape of the electron-hole-plasma luminescence²² we estimate at the highest excitation intensity a total photogenerated carrier density of about $1.5 \times 10^{13} \text{ cm}^{-2}$ (corresponding to a partial filling of the $n=2$ light-hole subband) and a band-gap renormalization of the order of 60 meV (related to the spectral position of the luminescence tail in the low-energy range).

Although the two samples have identical MQW configurations, the experimental spectra of sample 2 clearly exhibit a different behavior [Fig. 1(b)]. The optical confinement provided by the thick $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layer results in a sharp emission band (S band) peaked in the low-energy tail of the heavy-hole exciton and growing superlinearly with the excitation intensity. In the same excitation-intensity range adopted for sample 1, this S band redshifts by about 10 meV with increasing optical pumping. Our previous study of the optical gain connected to this radiative-recombination channel¹⁸ demonstrated that this emission band is due to the optical amplification of the spontaneous luminescence of the EHP.

These results show evidence of the difference between the optical properties of a dense electron-hole population in quantum wells under quasistationary conditions or above the threshold for lasing action.

B. Time-resolved high-excitation-intensity luminescence

Above the threshold for stimulated emission the carrier lifetime decreases down to several tens of picoseconds with increasing photogeneration rate. It is important to determine this lifetime shortening in order to evaluate the corresponding carrier-density reduction. To provide a quantitative estimate of this effect, we have measured the luminescence decay times of samples 1 and 2 under high excitation. The results are displayed in Figs. 2(a) and 2(b), where we show the time evolution of the luminescence. The decay time of the band-filling luminescence emitted by the higher-energy states in the well of sample 1 is $\tau_1 \approx 500 \text{ ps}$. At shorter times a broad electron-hole-plasma distribution is formed in the absorption continuum of the well. This results in a flat and broad luminescence spectrum in the first 100 ps [Fig. 2(a)]. At longer times carriers relax to the lowest subband and the carrier distribution narrows due to the decrease of the electron temperature. After about 500 ps, the high-energy tail of the band-filling luminescence disappears and the luminescence arises from a thermalized electron-hole plasma.

A completely different behavior is observed in the case of sample 2, as shown in Fig. 2(b). The stimulated-emission band rises following the time evolution of the exciting pulse, and decays with a time constant $\tau_2 = 32 \text{ ps}$. This extremely efficient decay channel considerably reduces the effective photogenerated carrier density and prevents the observation of a spontaneous radiative decay

from higher-energy states. For longer times, the emission spectrum shows again the broad spontaneous EHP emission around the energy of the fundamental interband transition.

According to the measured values of τ we expect that the carrier density in sample 2 reduces by a factor τ_1/τ_2 with respect to sample 1 under the same excitation conditions. Therefore the effective photogenerated carrier density in the spectra of Fig. 2(b) is of the order of 10^{12}cm^{-2} at the maximum excitation intensity. Detailed information on the transient band-gap renormalization has been obtained recently by studying the time evolution of the stimulated-emission spectra of sample 2.²¹

C. High-excitation-intensity luminescence in a magnetic field

In light of these results we now discuss the magneto-optic spectra of samples 1 and 2 under the same excitation conditions as in Fig. 1 as a function of the magnetic field. In Fig. 3 we show the luminescence spectra of sample 1 under the maximum excitation flux and at different magnetic-field values. The main feature of these spectra is the appearance of sharp peaks related to inter-Landau-level transitions superimposed on the band-filling luminescence. The effect of the quantizing magnetic field becomes evident in the spectra taken at fields larger than

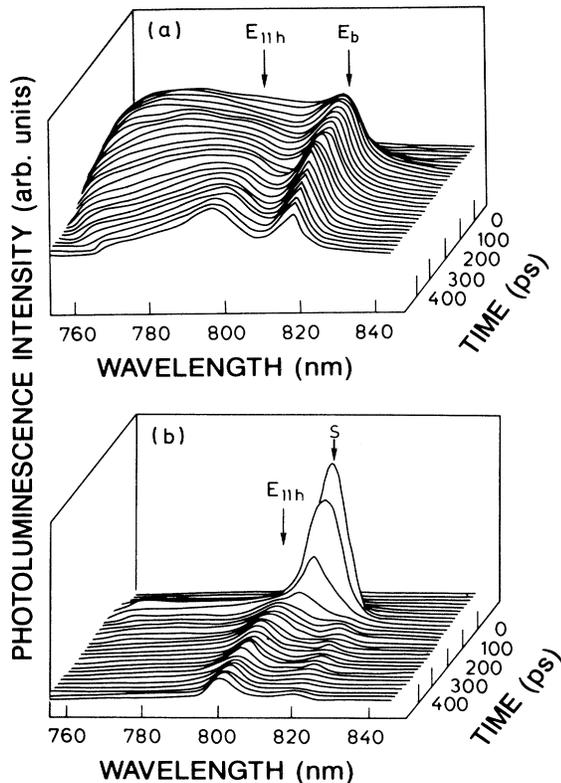


FIG. 2. (a) Time-resolved high-excitation-intensity photoluminescence spectra of sample 1. The temperature is 5 K and the exciting power density is $I_0 \approx 2 \text{ MW cm}^{-2}$. (b) Same as (a), but for sample 2.

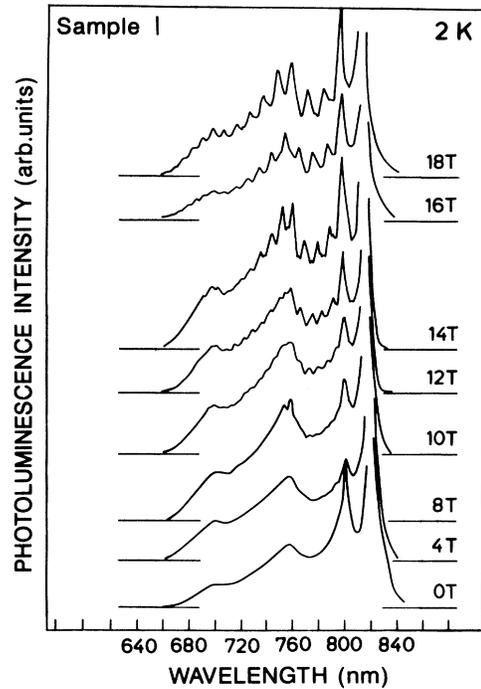


FIG. 3. Magnetoluminescence spectra of sample 1 at different magnetic fields and under the excitation intensity I_0 . The temperature is 2 K.

8 T. The Landau-level structures blueshift linearly with increasing field and 14 inter-Landau-level transitions can be observed at 18 T.

The magnetic-field dependence of the strong and well-resolved lines of sample 1 is exemplified in the fan-chart diagram of Fig. 4, which shows that all the lines are associated with Landau levels from the first electron-heavy-hole subband transition. Taking into account the degeneracy of each Landau level ($n_L = 2eB/h = 4.8 \times 10^{10} \text{ T cm}^{-2}$), the partial filling of the 14 Landau levels in the 18-T spectrum of Fig. 3 is consistent with the estimated carrier density at zero field. In this particular experiment no Landau levels related to the higher subbands are resolved. In fact, such transitions are always broader than the ones related to the lower subbands for the same reason as that also at zero field the excitonic transitions related to higher subbands are broader. In other samples higher subband Landau levels are clearly visible; however, these transitions are not our main concern here. In fact, only a few weak and barely resolved lines related to higher subband Landau levels can be observed among the sharp Landau peaks of Fig. 3.

In discussing the magnetic-field dependence of the peak energy in Fig. 4, we first note that the Landau levels shift linearly with the magnetic field, and that only the lowest energy transition exhibits the excitonic diamagnetic shift at low fields. As we discussed before in the context of the zero-field luminescence, this lowest peak remains a matter of controversy. In fact, at fields around 6–10 T the energy of the excitonic transition is rather different from the expected zeroth-Landau-level transi-

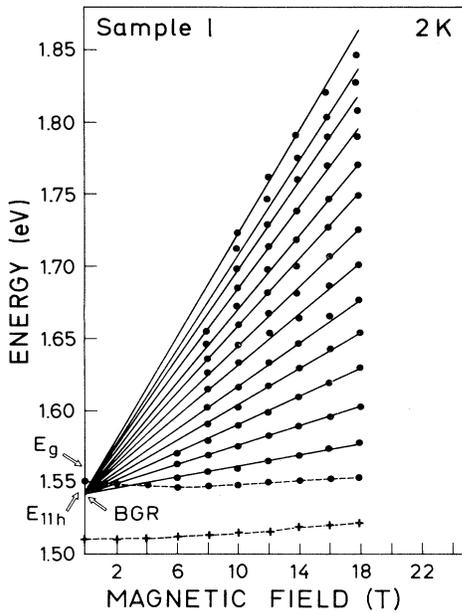


FIG. 4. Measured luminescence peak positions (dots) as a function of the magnetic field at the excitation intensity I_0 for sample 1. The straight lines are the linear interpolation of the Landau-level shift with slope ≈ 1.34 meV/T. The overall plot does not change with the excitation intensity. The arrows indicate the extrapolation of the band-gap reduction (BGR) at zero field, the heavy-hole-exciton ground state measured in absorption ($E_{11h} = 1.522$ eV), and the $n = 1$ conduction-to-heavy-hole subband gap of the quantum well ($E_g = 1.561$ eV).

tion, and in the spectra one would expect both to be visible. However, this is not the case and has constituted one of the arguments of Potemski *et al.* that these data indeed show the coexistence of the exciton inside the EHP.^{12,13}

The extrapolation of the fan curves at zero field crosses the energy axis at a lower energy than the unperturbed band gap of the investigated MQW, indicating a shrinkage of the band gap due to the single-particle energy renormalization in the electron-hole plasma.

The least-squares linear interpolation of the experimental high-field transitions shown in Fig. 4 gives a slope of 1.37 meV/T for the Landau-level shift and a common intercept at zero field at the energy of 1.541 eV, in agreement with previous studies.^{12,13} This Landau shift is significantly smaller than expected. In fact, even assuming an infinite heavy-hole mass and accounting for the conduction-band nonparabolicity with an energy-dependent mass of the form $m_e(E) = m_e(1 + AE)$ [where E is the electron energy with respect to the bottom of the conduction band in the bulk and A is a parameter that in GaAs is about 0.64 eV⁻¹ (Ref. 23)], one gets a Landau shift of about 1.56 meV/T using $m_e = 0.067$ and $E = 150$ meV. Most important is that from low-excitation magneto-optical experiments, a Landau-level shift of about 1.7 meV/T is obtained for GaAs QW's. Therefore the observed slope of 1.37 meV/T corresponds to an electron mass about 20% heavier than in the low-density

case. As already reported in Refs. 12 and 13, the origin of this discrepancy lies in the many-body renormalization expected at these carrier densities.^{14,24}

The energy obtained by extrapolation of the straight lines in the fan-chart diagram of Fig. 4 deserves more attention. The obtained value of 1.541 eV is 18 meV lower than the fundamental intersubband transition energy deduced from the low-intensity photoluminescence excitation, and this corresponds to a BGR of about 2 Ry.^{12,13} The experimental value of 18 meV is by far smaller than the BGR value expected at the electron-hole density of our experiment ($n \approx 10^{13}$ cm⁻²). The density dependence of the BGR in a 10-nm-wide GaAs well can be approximated by²⁵

$$3.1 \times 10^{-3}(n)^{1/3} \text{ meV}, \quad (1)$$

which would lead to a 66-meV gap shrinkage at $n = 10^{13}$ cm⁻² carrier density. Moreover, the observed gap shrinkage does not agree with the BGR measured in similar samples by means of luminescence and optical-gain spectroscopy at high carrier densities.^{18,21,26} It is important to note that the BGR is found to be independent of the excitation intensity. All the fan charts obtained by changing the excitation intensity over three orders of magnitude (from kW cm⁻² to MW cm⁻²) extrapolate at a zero-field energy which is about 2 Ry smaller than the fundamental interband transition of the investigated MQW.¹² We stress that the entire fan-chart plot, not only the zero-field intercept, shows no dependence on excitation intensity. Therefore any deviation from linearity of the magnetic-field dependence of the transitions at low fields would not explain the observed behavior of the band-gap reduction.

This unexpected saturation of the BGR at the value $E_g - 2$ Ry is, however, not a general property of the two-dimensional electron-hole gas in a magnetic field. This is clearly demonstrated by the magnetoluminescence spectra of sample 2 shown in Fig. 5. At the highest excitation intensity the emission spectra exhibit the S band, which blueshifts with increasing magnetic field, and the heavy-hole-exciton line, collected from the external region of the excitation spot, splits into a well-resolved doublet structure.²⁷ As discussed in Sec. III B, the rising of the stimulated-emission band leads to the saturation of the spontaneous emission and prevents the observation of several inter-Landau-level transitions superimposed on the band-filling luminescence. In fact, although the condition $\hbar\Omega \ll \mu$ is still fulfilled under the high photogeneration rate, the electron-hole gas does not reach a quasistationary condition as in sample 1. Lifetime effects dominate the radiative-recombination processes in sample 2. The electron-hole population rapidly decays through the stimulated-emission channel from the lowest Landau level and the magnetoluminescence spectra only exhibit that strong feature.

The magnetic-field dependence of the S band is shown in Fig. 6. The Landau-level shift is clearly linear with the magnetic field at high fields. From the linear interpolation of the fan chart we again find a 20% enhancement of the electron mass. The most striking result is that, in contrast to the case of sample 1, the renormalized band

gap obtained from the zero-field extrapolation of the fan-chart diagram clearly depends on the excitation intensity. The absolute BGR value of 32 meV obtained at the maximum excitation intensity (I_0 curve in Fig. 6) corresponds to a carrier density of $1.1 \times 10^{12} \text{ cm}^{-2}$ in Eq. (1), in good agreement with the carrier density evaluated in Sec. III B. We stress that not only the zero-field intercept but the entire fan-chart plot redshifts with increasing excitation intensity. On the contrary, the doublet structure of the exciton peak does not show any excitation intensity dependence and it shifts quadratically with the magnetic field. Other recombination processes responsible for the S-band emission, such as inelastic exciton-exciton or exciton-electron scattering,²⁸ can be excluded on the basis of the theoretically predicted magnetic-field dependence of their luminescence.²⁹ Energy and momentum conservation for these scattering processes would lead to characteristic luminescence lines centered at $\hbar\omega_{\text{ex-ex}} \simeq 2E_x - E_g = E_g - 2 \text{ Ry}$ for the exciton-exciton scattering (where E_x is the exciton energy and E_g is the gap), and at $\hbar\omega_{\text{ex-e}} \simeq E_x - E_e^{\text{kin}}$ for the exciton-electron scattering (where E_e^{kin} is the kinetic energy of the diffused electron). Therefore the magnetic-field-induced shift of these recombination lines should be²⁹ $\hbar\omega_{\text{ex-ex}}(B) \simeq 2E_x(B) - 1/2\hbar\Omega$ and $\hbar\omega_{\text{ex-e}}(B) \simeq E_x(B)$, where $E_x(B)$ is the diamagnetic shift of the exciton. Neither of these two field dependences accounts for the observed Landau-level shift of the S band. Only at small fields ($B < 1.5 \text{ T}$) does the fan-chart diagram of the S band exhibit a devia-

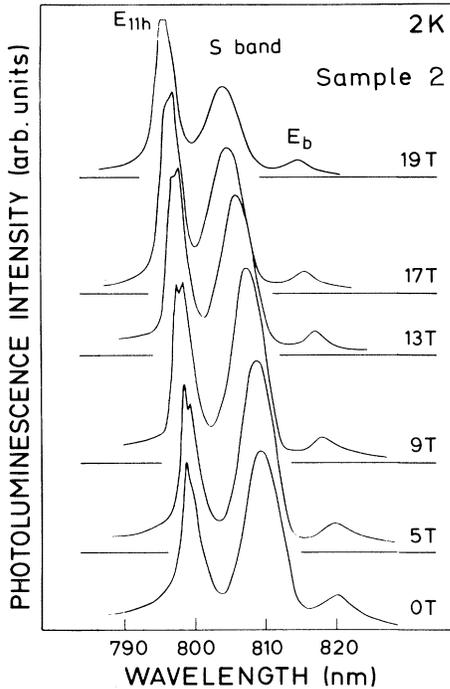


FIG. 5. Magnetoluminescence spectra of sample 2 at different magnetic fields and under the excitation intensity I_0 . The temperature is 2 K.

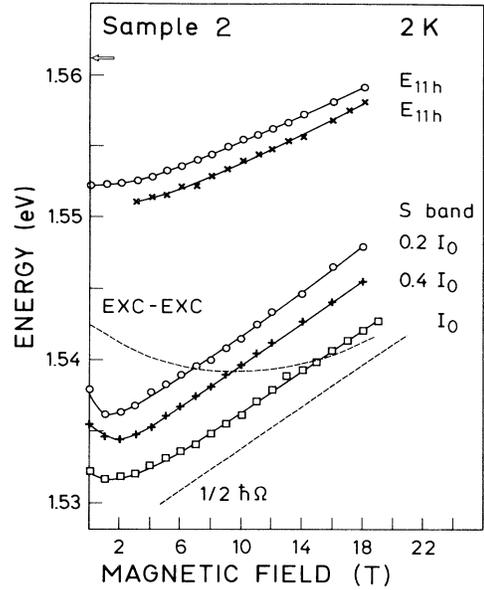


FIG. 6. Measured luminescence peak positions (symbols) as a function of the magnetic field and at different excitation intensities ($I_0 \simeq 2 \text{ MW cm}^{-2}$) for sample 2. The E_{11h} curves are related to the splitted doublet of the heavy-hole-exciton luminescence, while the lower plots are related to the S-band luminescence. The solid curves are guides for the eye. The dashed curves represent the calculated magnetic-field-induced shift of the luminescence arising from inelastic exciton-exciton collision (exc-exc curve) and the electron Landau-level shift with a 20% mass renormalization ($1/2 \hbar\Omega$ curve). The horizontal arrows indicate the $n = 2$ heavy-hole-exciton transition measured in absorption.

tion from the electronic linear behavior, which is similar to the line shape of the exciton-exciton scattering curve (dashed line in Fig. 6).

IV. DISCUSSION

The data presented in this work clearly show evidence of the role played by lifetime effects in the magnetoluminescence properties of the dense electron-hole gas in quantum wells. We recall that the samples of this study have identical structural parameters and the only difference lies in the presence of the thick $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layer underneath the MQW heterostructure of sample 2, which favors optical amplification of the luminescence. Therefore the difference in the observed spectral behavior only lies in the carrier lifetimes. The first important result of our study is that band-gap renormalization does occur in quantum wells in a high magnetic field. The expected electron-hole-plasma behavior of the band-gap renormalization is indeed observed under stimulated-emission conditions (sample 2), i.e., in QW samples with short carrier lifetimes, in contrast to the original assumption of the magnetic-field-induced saturation of the band-gap shrinkage in MQW's.^{12,13} On the other hand, QW samples having long carrier lifetimes (sample 1) exhibit the density independence of the BGR first observed

by Potemski *et al.*,^{12,13} which is in fact not predicted by the most recent theories.^{16,17}

In what follows, we discuss a few possible mechanisms responsible for the saturation of the band-gap renormalization observed in sample 1. Although these speculative attempts only provide a qualitative picture, they are helpful in clarifying different aspects of the dense electron-hole-plasma physics in strong magnetic fields. A possible explanation for the observed decrease of the BGR is that carriers belonging to different Landau levels do not interact. Assuming that each Landau level renormalizes according to its own population might explain the strongly reduced BGR exhibited by sample 1 where the total carrier density is divided among many (> 10) Landau levels. A similar situation occurs for the quantum-well subbands.^{26,30} The renormalization of a given subband is much more sensitive to its own population than to the density of carriers in other subbands. However, this assumption is not correct for the case of electrons confined in Landau levels. As shown in the Appendix, the exchange energy contribution due to inter-Landau-level interactions is comparable to the contribution coming from intra-Landau-level interactions. This suggests that the energy of carriers belonging to a certain Landau level does not renormalize independently of the other levels. Furthermore, this assumption would not be consistent with the BGR exhibited by sample 2 which corresponds to the total density of photogenerated carriers, even though the radiative recombination is dominated by the stimulated emission from the first Landau level only.

The longer carrier lifetime suggests the possibility of a phase transition in sample 1, i.e., the formation of a condensate phase. In this case, an increase of the excitation intensity would increase the volume of the liquid droplet, but not the density. However, the observed BGR of 18 meV in sample 1 corresponds to a density of about $2 \times 10^{11} \text{ cm}^{-2}$, which appears too low for the formation of an electron-hole liquid, especially considering the high temperature of the photoexcited carriers. Furthermore, no threshold behavior with the excitation intensity or with the temperature is observed for the saturation of the BGR, which is in contrast to the well-known electron-hole-liquid formation process.¹

In a recent paper Maan *et al.* have proposed a slightly different explanation for the apparent density independence of the BGR.³¹ Their explanation is based on the observation that the luminescence line shape develops a broad low-energy tail which extends further into the gap as the carrier density is increased and behaves more or less like the normal BGR. The width of this tail may exceed easily the cyclotron energies involved, but nevertheless no Landau-level-like structures can be observed in its shape. At the same time the Landau-level peaks observed on the high-energy side of the luminescence are seen to broaden substantially with increasing density, although they remain at the same energy as we observe here in sample 1. The explanation they propose is that there is a two-phase system. One phase consists of particles which scatter so rapidly that they cannot complete one cyclotron orbit before being scattered ($\Omega\tau \ll 1$). These particles do not show Landau-level quantization,

but are responsible for the low-energy tail and show BGR. The other phase consists of a class of particles within this rapid scattering continuum, which shows a relatively long-lived and locally correlated motion of electrons and holes. This class of particles is thought to give rise to sharp emission peaks at the Landau-level transition energies. This correlated motion is long lived because $\Omega\tau \gg 1$, since they can be observed as sharp peaks, and local, because they must be of the size of cyclotron orbit (8.5 nm at 10 T). The broadening of the peaks with increasing density is then explained by a gradual shortening of the correlated motion, which broadens the peaks, and eventually merging of the correlated motion with a continuum. In this sense the magnetospectroscopy can be seen as a form of time-resolved spectroscopy on the time scale of the cyclotron frequency. This model has the appealing feature that it reconciles the contradiction between the apparent saturation of the BGR deduced from the density independence of the Landau-level peaks, with the conventional experimental and theoretical wisdom that BGR remains strongly density dependent. Furthermore, this hypothesis is fully compatible with our present experimental results where we do see the normal BGR in samples with short lifetimes, whereas the same density-independent Landau levels are observed in samples with long lifetimes. An unsatisfactory aspect of the model is, however, that it gives no explanation of why the levels are all shifted by an amount of twice the exciton binding energy.

Another possibility, somewhat analogous, is that a high-density liquid forms, but the luminescence is dominated by a low-density gas being in thermodynamic equilibrium with the liquid. An appealing candidate for such a high-density phase seems to be the condensate of weakly attractive magnetoexcitons described by Paquet *et al.*,¹⁷ as its luminescence is predicted to be strongly suppressed. Moreover, in this case, the low-density phase can be a gas of excitons decaying through inelastic-scattering events in which two excitons give rise to an electron-hole pair and a photon.²⁹ According to the discussion of Sec. III C, it is thus expected that the luminescence at high magnetic fields is dominated by lines corresponding to the free-carrier Landau levels diminished by twice the exciton binding energy, as we observe in our experiments (see the fan chart of Fig. 4). The excitonic condensate has so far been studied in the ultraquantum limit, i.e., with carriers in the first Landau level only, and at zero temperature.¹⁷ At present it is not clear whether such a simple picture is valid for the case in which many Landau levels are populated and the thermal energy becomes comparable to the cyclotron energy. Further studies are needed to clarify the validity of this assumption.

V. CONCLUSIONS

We have presented a detailed spectroscopic investigation of the magnetoluminescence and the temporal evolution of the recombination of a dense electron-hole plasma confined in GaAs quantum wells. We demonstrate that lifetime effects strongly change the optical properties of the free-carrier plasma in the presence of high magnetic

fields. Heterostructures with long carrier lifetimes exhibit richly structured magnetoluminescence spectra independent of the photogeneration rate and clear saturation of the band-gap renormalization. Conversely, multiple quantum wells in which the free-carrier lifetime is very short exhibit the usual density-dependent gap shrinkage with magnetoluminescence spectra strongly dependent on the photogeneration rate. These results suggest that two different phases can form in the crystal in the presence of high magnetic fields: a long-living highly correlated particle system, analogous to a condensed exciton or electron gas at the equilibrium density, which does not show any spectral shift of the luminescence with increasing excitation intensity; and a short-living free-carrier population, which exhibits the usual electron-hole plasma behavior with density-dependent band-gap reduction. We believe that our results can stimulate further studies to fully understand the electronic and optical properties of the dense electron-hole plasma in high magnetic fields.

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APPENDIX

In this appendix it is shown that in the limit of large filling factors ($\hbar\Omega \ll \mu$) the exchange interaction energy of the two-dimensional electron gas scales according to the total density of carriers, irrespective of their principal Landau quantum number. Therefore, even though eigenfunctions belonging to different levels are orthogonal, inter- and intra-Landau-level many-body interactions are, in general, comparable. The Landau eigenfunctions can be written as

$$\Psi_{k,n}(x,y) = \frac{Ae^{iky}}{\sqrt{2^n n! l_0}} \exp\left[\frac{(x + kl_0^2)^2}{2l_0^2}\right] H_n\left[\frac{x + kl_0^2}{l_0}\right], \quad (\text{A1})$$

where l_0 is the magnetic length

$$l_0 = \left[\frac{\hbar}{eB_0}\right]^{1/2},$$

H_n are Hermite polynomials

$$H_n(x) = (-1)^n e^{x^2} \frac{d^n}{dx^n} e^{-x^2},$$

and A is a normalization constant (independent of n , k , and l_0). Using the Fourier representation of the Coulomb potential and the following result:³²

$$\begin{aligned} & \left| \int dx e^{iq_x x} \Psi_{k,n}^* \Psi_{k-q_y, n'} \right| \\ &= B \left[\frac{2^{n'} n'!}{2^n n!} \right]^{1/2} (ql_0)^{n-n'} e^{-(ql_0/2)^2} \\ & \times \left| L_{n'}^{n-n'} \left[\frac{q^2 l_0^2}{2} \right] \right| = J_{n,n'}(ql_0) B, \end{aligned} \quad (\text{A2})$$

with ($n \geq n'$), where B is a constant (independent of n , n' , k , l_0 , and q), L_n^m are Laguerre polynomials

$$L_n^m(x) = \frac{1}{n!} e^{x^2} x^{-m} \frac{d}{dx^n} (e^{-x^2} x^{m+n}),$$

and $J_{n,n'}$ depends only on the magnitude of q ; the contribution to the energy of an electron in the Landau level n' due to the exchange interaction with the full Landau level n can be expressed as

$$\begin{aligned} \Delta E_{\text{ex}}^{n,n'} &= - \int d^2q \frac{2\pi e^2}{q} [J_{n,n'}(ql_0)]^2 \\ &= -C \frac{e^2}{l_0} \int_0^\infty dx \frac{m!}{(m+p)!} x^{p-1/2} e^{-x} [L_m^p(x)]^2, \end{aligned} \quad (\text{A3})$$

where $m = \min(n, n')$, $p = |n - n'|$, and C is a positive constant (independent of n , n' , and l_0). Writing the Laguerre polynomials in terms of confluent hypergeometric functions

$$L_m^p(x) = \frac{(m+p)!}{m! p!} F(-m, p+1, x),$$

Eq. (A3) reduced to³³

$$\begin{aligned} \Delta E_{\text{ex}}^{n,n'} &= -C \frac{e^2}{l_0} \sqrt{\pi} \frac{(2p-1)!!}{(2p)!!} \left[1 + \frac{m(-\frac{1}{2})(\frac{1}{2})}{1^2(p+1)} + \frac{m(m-1)(-\frac{1}{2}-1)(-\frac{1}{2})(\frac{1}{2})(\frac{1}{2}+1)}{1^2 2^2(p+1)(p+2)} + \dots \right. \\ & \left. + \frac{m(m-1) \cdots 1[-\frac{1}{2}-(m-1)] \cdots [\frac{1}{2}+(m-1)]}{1^2 2^2 \cdots m^2(p+1)(p+2) \cdots (p+m)} \right]. \end{aligned} \quad (\text{A4})$$

The contribution to the energy of an electron in the first Landau level ($n'=0$) due to the exchange interaction with the electrons in ν full Landau levels ($n=0, 1, 2, \dots, \nu$) is, therefore,

$$\Delta E_{\text{ex}}(0; \nu) = \sum_{j=0}^{\nu} \Delta E_{\text{ex}}^{j,0} \simeq \left[-\frac{e^2}{l_0} \right] \left[\sum_{j=0}^{\nu} \frac{(2j-1)!!}{(2j)!!} \right]. \quad (\text{A5})$$

Given a fixed total density of carriers per unit area ρ , the limit $\hbar\Omega \ll \mu$ corresponds to $\nu \rightarrow \infty$ and $l_0 \rightarrow \infty$ with

$$\frac{\nu}{2\pi l_0^2} = \rho,$$

where

$$\frac{1}{2\pi l_0^2}$$

is the degeneracy of each Landau level. Writing

$$\frac{(2j-1)!!}{(2j)!!} = \frac{(2j)!}{[2^j(j!)^2]}$$

and using Stirling's formula ($n! \rightarrow n^n e^{-n} \sqrt{2\pi n}$), for large j one obtains

$$\frac{(2j-1)!!}{(2j)!!} \simeq \frac{1}{\sqrt{j}}$$

and finally

$$\Delta E_{\text{ex}}(0; \nu \gg 1) \simeq \left[-\frac{e^2}{l_0} \right] \int^{\nu} \frac{dx}{\sqrt{x}} \simeq \frac{e^2}{l_0} \sqrt{\nu} \simeq -e^2 \sqrt{\rho}. \quad (\text{A6})$$

This behavior is the same as in the high-density limit in a zero magnetic field,³⁴

$$\Delta E_{\text{ex}} \rightarrow -\frac{2\pi e^2}{k_F} \rho \simeq -e^2 \sqrt{\rho}, \quad (\text{A7})$$

where k_F is the two-dimensional Fermi wave vector. Even though only the contribution of the unscreened exchange interaction is included, the results of Eq. (A6) are sufficient to conclude that when many Landau levels are populated it is by no means justified to estimate the many-body corrections by neglecting inter-Landau-level interactions.

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- ¹I. V. J. Kukushkin, S. V. Meshkov, and V. B. Timofeev, *Usp. Fiz. Nauk* **155**, 219 (1988) [*Sov. Phys.—Usp.* **31**, 511 (1988)].
- ²J. C. Maan, G. Belle, A. Fasolino, M. Altarelli, and K. Ploog, *Phys. Rev. B* **30**, 2253 (1984).
- ³D. C. Rogers, J. Singleton, R. J. Nicholas, C. T. Foxon, and K. Woodbridge, *Phys. Rev. B* **34**, 4002 (1986).
- ⁴X. L. Zheng, D. Heiman, and B. Lax, *Phys. Rev. B* **40**, 10523 (1989).
- ⁵T. T. J. M. Berendschot, H. A. J. M. Reinen, and H. J. A. Bluyssen, *Solid State Commun.* **63**, 873 (1987).
- ⁶S. Katayama and T. Ando, *Solid State Commun.* **70**, 97 (1989).
- ⁷T. Ando, in *High Magnetic Fields in Semiconductor Physics II*, Vol. 87 of *Springer Series in Solid State Sciences*, edited by G. Landwehr (Springer, Heidelberg, 1989), p. 164.
- ⁸M. S. Skolnick, K. J. Nash, S. J. Bass, P. E. Simmonds, and M. J. Kane, *Solid State Commun.* **67**, 637 (1988).
- ⁹J. M. Worlock, A. C. Maciel, A. Petrou, C. H. Perry, L. R. Aggarwal, M. C. Smith, A. C. Gossard, and W. Weimann, *Surf. Sci.* **142**, 486 (1984).
- ¹⁰T. Uenoyama and L. J. Sham, *Phys. Rev. B* **39**, 11044 (1989).
- ¹¹W. Chen, M. Fritze, A. V. Nurmikko, D. Ackley, C. Colvard, and H. Lee, *Phys. Rev. Lett.* **64**, 2434 (1990).
- ¹²M. Potemski, J. C. Maan, K. Ploog, and G. Weimann, *Proceedings of the 19th International Conference on PS*, edited by W. Zawadzki (IPPAS, Warsaw, 1988), p. 119; *Solid State Commun.* **75**, 185 (1990); *Surf. Sci.* **229**, 380 (1990).
- ¹³M. Potemski, J. C. Maan, K. Ploog, and G. Weimann, in *Scpectroscopy in Semiconductor Microstructures*, Vol. 206 of *NATO Advanced Study Institute, Series B: Physics*, edited by G. Fasol, A. Fasolino, and P. Lugli (Plenum, New York, 1989), p. 425.
- ¹⁴H. L. Störmer and R. W. Martin, *Phys. Rev. B* **20**, 4213 (1979).
- ¹⁵L. V. Keldysh and T. A. Onishchenko, *Pis'ma Zh. Eksp. Teor. Fiz.* **24**, 70 (1976) [*JETP Lett.* **24**, 59 (1976)].
- ¹⁶G. E. W. Bauer, *Phys. Rev. Lett.* **64**, 60 (1990).
- ¹⁷D. Paquet, T. M. Rice, and K. Ueda, *Phys. Rev. B* **32**, 5208 (1985).
- ¹⁸R. Cingolani, K. Ploog, A. Cingolani, C. Moro, and M. Ferrara, *Phys. Rev. B* **42**, 2893 (1990).
- ¹⁹G. Lasher and F. Stern, *Phys. Rev.* **133**, A533 (1963).
- ²⁰E. O. Göbel, J. Kuhl, H. J. Polland, and K. Ploog, *Appl. Phys. Lett.* **47**, 781 (1985).
- ²¹R. Cingolani, H. Kalt, and K. Ploog, *Phys. Rev. B* **42**, 7655 (1990).
- ²²R. Cingolani, M. Ferrara, M. Lugará, Y. Chen, F. Bassani, J. Massies, and F. Turco, *Europhys. Lett.* **7**, 751 (1988).
- ²³U. Ekenberg, *Phys. Rev. B* **40**, 7714 (1989).
- ²⁴S. Schmitt-Rink and C. Ell, *J. Lumin.* **30**, 585 (1985).
- ²⁵S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, *Adv. Phys.* **38**, 89 (1989).
- ²⁶C. Weber, C. Klingshirn, D. S. Chemla, D. A. B. Miller, J. E. Cunningham, and C. Ell, *Phys. Rev. B* **38**, 12748 (1989).
- ²⁷The exchange splitting of the heavy-hole-exciton ground state has been studied in detail by M. Potemski, J. C. Maan, A. Fasolino, K. Ploog, and G. Weimann, *Surf. Sci.* **229**, 151

- (1990).
- ²⁸R. Cingolani, K. Ploog, G. Peter, R. Hahn, E. O. Göbel, C. Moro, and A. Cingolani, *Phys. Rev. B* **41**, 3272 (1990).
- ²⁹E. Göbel, K. L. Shaklee, and R. Epworth, *Solid State Commun.* **17**, 1185 (1975).
- ³⁰J. A. Levenson, I. Abram, R. Raj, G. Dolique, J. L. Oudar, and F. Alexandre, *Phys. Rev. B* **38**, 13 443 (1989).
- ³¹J. C. Maan, M. Potemski, K. Ploog, and G. Weimann, *Proceedings of the International Winterschool on the Physics of Semiconductors*, Springer Series in Solid State Sciences, edited by G. Bauer, F. Kuchar, and H. Heinrich (Springer, Heidelberg, in press).
- ³²Bateman Manuscript Project, *Tables of Integral Transforms*, edited by A. Erdelyi (McGraw-Hill, New York, 1954), Vol. 2, Sec. 16.5, Eq. 30.
- ³³L. D. Landau and E. M. Lifshitz, *Quantum Mechanics, Course of Theoretical Physics* (Pergamon, New York, 1965), Vol. 3, Eq. f.6 of the mathematical appendixes.
- ³⁴C. Ell, R. Blank, S. Benner, and H. Haug, *J. Opt. Soc. Am. B* **6**, 2006 (1989), Eq. 2.15.