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Gain measurements and band-gap renormalization in GaAs/$Al_xGa_{1-x}As$ multiple-quantum-well structures

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Using a pump and probe technique with nanosecond excitation pulses we measure the nonlinear optical properties of GaAs/$Al_{0.3}Ga_{0.7}As$ multiple-quantum-well (MQW) structures under quasistationary excitation conditions. The main aspects of our work are the phenomena of optical gain, the renormalization of the fundamental band gap and of the higher subbands ($n_z=2$, $n_z=3$), due to the electron-hole population of the lowest ($n_z=1$) subbands. We present the first systematic analysis of the gain spectra to determine the renormalization of the $n_z=1$ subband and we interpret the data in terms of effective exciton parameters in combination with a pure two-dimensional theory.

In the present work we investigate the nonlinear optical properties of a quasi-two-dimensional electron-hole system at high densities in GaAs/$Al_{1-x}Ga_xAs$ multiple quantum wells (MQW) under quasistationary excitation conditions by applying laser pulses of typically 15 ns full width at half maximum (FWHM). In recent years, most contributions on high-excitation phenomena in MQW deal with picosecond and subpicosecond excitation times,[1,2] and therefore mainly study various aspects of relaxation processes of the electron-hole system under nonequilibrium conditions. Using a pump-and-probe-beam technique we study the change of the optical properties induced by high-excitation effects such as phase-space filling, exchange interaction, optical gain, band-gap renormalization, or screening of the Coulomb interaction under quasistationary conditions.

From our experimental findings we derive the density-dependent renormalization of the fundamental band gap by analyzing the gain spectra at various excitation levels. Strong influences of the excitonic enhancement on the line shape of optical gain in different temperature and density regimes become obvious[3,4] thus complementing first reports of the appearance of gain in MQW structures under fs excitation as shown in Ref. 5. Another aspect of our research is the study of the higher subband renormalization due to intersubband interaction, i.e., the shift of the $n_z=2$ and $n_z=3$ subband transitions.

The MQW samples consist of either 100 or 50 periods of 100-Å GaAs wells and 100-Å $Al_{0.3}Ga_{0.7}As$ barriers. The GaAs substrate has been carefully removed by etching in order to perform the transmission experiments. The pump-light source consists of an excimer-laser-pumped dye laser emitting pulses of typically 15 ns (FWHM); the weaker broad-band probe pulse has a duration of 3 ns (FWHM) and was temporally and spatially centered with respect to the pump pulse.

The pump photon energy $\hbar\omega_{exc}=1.675$ eV is chosen below the absorption edge of the $Al_{1-x}Ga_xAs$ barriers or cladding layers so that the photoexcited carriers are directly created in the wells slightly above the $n_z=2$ exciton. Since a considerable fraction of the pump beam is transmitted, we can anticipate almost homogeneous excitation of all 100 quantum wells (QW). The detection system consists of a multichannel analyzer. The absorption spectra are obtained by careful substraction of the luminescence, especially the stimulated emission, which appears mainly in the plane of the layers.

In Fig. 1 an overview of the density-dependent changes of the absorption spectra is plotted for a lattice tempera-

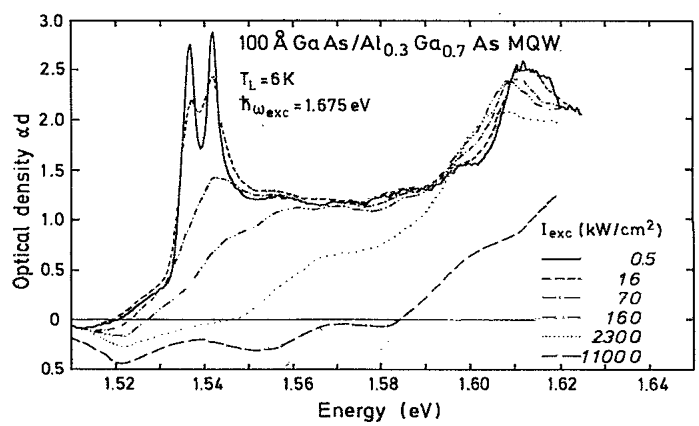

FIG 1. Absorption spectra of a (100-Å GaAs)/(100-Å $Al_{1-x}Ga_xAs$) MQW structure under quasistationary excitation conditions (pump intensity I_{exc}) at $T_L=6$ K.

ture $T_L=6$ K. The figure shows the absorption of the sample at the $n_z=1$ and $n_z=2$ subbands at different pump intensities I_{exc}. The spectrum at $I_{exc}=0.5$ kW/cm^2 differs from the unexcited sample only in a slightly reduced $n_z=1$ heavy-hole–exciton (1hh-ex) absorption peak which is in the unexcited case higher than the $n_z=1$ light hole (1lh-ex) one. An analysis of the nonlinearities related to the $n_z=1$ exciton resonances shows clearly two domains.

(i) Starting with lowest excitation levels one observes a reduction of the oscillator strength of the 1hh and 1lh excitons. This initial process is mainly due to phase-space filling (PSF) and to the short-range exchange interaction (EI) as the screening of the long-range Coulomb interaction is strongly reduced in a two-dimensional (2D) system.[6] The 1hh-ex is affected by PSF and EI of both its electron and its hole (1hh) whereas the 1lh-ex, which shares only the electron with the 1hh-ex, is not affected by the PSF and EI associated with the 1hh. Hence, the 1hh-ex bleaches faster (see Fig. 1) with increasing population density until the 1lh subband becomes also occupied. The energetic positions of the two excitons remain unchanged under excitation which is proof for the almost complete balance between band-gap renormalization (BGR) and the reduction of the exciton binding energy due to intrasubband many-body processes in 100-Å QW.[7]

(ii) Above approximately 60 kW/cm^2 the excitonic structures at the $n_z=1$ subbands have vanished; the residual shape of the absorption edge reflects the steplike density of states of a two-dimensional system and is influenced by the Sommerfeld enhancement.[8] With further increased pump level (I_{exc} above 100 kW/cm^2) band filling plays the dominant role in the absorptive changes. This process leads to a net blue shift of the absorption edge whereas the fundamental energy gap E_g' renormalizes further to lower energies (see Fig. 2).

If we now look at the $n_z=2$ subbands the features are significantly different. With increasing pump level (up to 700 KW/cm^2) one observes a red shift (accompanied by a reduction) of the 2hh exciton peak ΔE_{2hh} of up to 5.6 meV thus indicating a renormalization of the second subband.[1] A closer analysis shows that the 2hh exciton peak shifts down by approximately 3 meV ($I_{exc}=160$ kW/cm^2) without any significant occupation of the $n_z=2$ subband. In this regime, indicated by the complete recovery of absorption energetically below the 2hh absorption edge, only a few states of the $n_z=2$ subbands are populated due to the fast intersubband-relaxation times of the photoexcited carriers [<10 ps in a 116-Å QW (Ref. 9)]. We would like to stress that the shift ΔE_{2hh} itself already indicates intersubband interaction as the main reason for the observed exciton shift, as many-body effects arising from occupation of the $n_z=2$ subband would not alter the $n_z=2$ exciton transition energy (see remark to the $n_z=1$ resonance above) until the exciton peak vanishes into the continuum.

At further increased excitation levels the occupation of the second subbands, described by the distribution functions of the carriers, successively leads to a complete bleaching of the 2hh exciton by PSF and EI and, moreover, to a reduction of the band-to-band absorption, and the $n_z=2$ subband shifts to lower energies. In addition, we

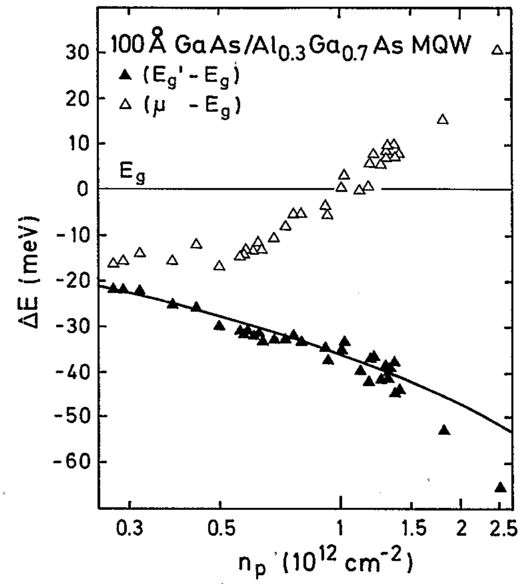

FIG 2. Renormalization of the fundamental band gap and chemical potential as function of the two-dimensional e-h density n_p for lattice temperatures $T_L=6$ K and 13 K. The solid line refers to the calculation in a dynamical SPP with parameters given in the text.

observe a red shift of the 3hh exciton peak which is slightly less than ΔE_{2hh} under comparable excitation conditions.

These findings underline that the $n_z>1$ edges renormalize due to the population of carriers in the $n_z=1$ subbands. There are two possibilities to explain the observed intersubband BGR,[10] namely intersubband screening and exchange interactions (EI). Intersubband screening should be small in quasi-2D systems because of the great wave-vector mismatch of states in two different subbands arising from the confinement of the wave functions perpendicular to the potential wells.[10] On the other hand, intersubband-EI (see also Ref. 11) should be negligible due to the orthogonality of the wave functions in different subbands. Moreover, the EI is in general of minor importance for the fundamental BGR at moderate plasma densities as is shown, e.g., in Ref. 3. In any case, the intersubband BGR in QW is small compared to the BGR of the fundamental gap: In Fig. 3 we compare the red shift of the $n_z=1$ gap ΔE_g and the shift ΔE_{2hh} in the density regime, before a strong occupation of the $n_z=2$ subband results in complete bleaching by PSF and EI. We deduce ΔE_g from the low-energy edge of the gain spectra as discussed below (the 1hh-exciton binding energy is $E_{1S}=8.7$ meV[12,13]). The values for the plasma density of the $n_z=1$ subband given on the upper x axis in Fig. 3 are deduced from the measured BGR (see Fig. 2). We find in this regime $\Delta E_{2hh}\simeq 0.16\Delta E_g$. Even if one assumes that the peak is no longer the 2hh exciton at elevated plasma densities but the enhancement of the subband edges as calculated by Ref. 14, the shift of the $n_z=2$ subband is still much smaller than ΔE_g, thus excluding the assumption of a rigid shift of the whole subband structure.[15]

In Fig. 1, at excitation intensities above $I_{exc}=160$ kW/cm^2 we observe optical amplification of the probe light. The chemical potential μ is given by the crossover

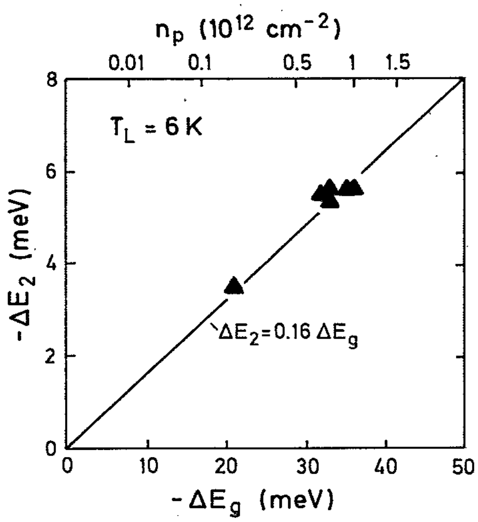

FIG 3. Red-shift of the 2hh-exciton resonance vs renormalization of the fundamental band gap. n_p indicates the e-h density in the $n_z=1$ subband.

from gain to absorption and shows a blue shift with increasing plasma density. In Fig. 1 the line shape of the optical gain is considerably disturbed by Fabry-Perot modes. At elevated temperatures and high e-h pair densities the spectra offer the single particle result according to a constant density of states of a 2D system[16] (with some final-state damping) while at lower temperatures and densities the influence of the e-h correlation is more pronounced.[16,4]

From the optical spectra we determined the energetic position of the chemical potential $\mu = \mu_e + \mu_h$ and the fundamental band edge E_g' (from the low-energy edge of the gain spectra). We calculate the two-dimensional e-h pair density n_p from the spectral width of the gain spectra according to a simple three-band model, which takes into account the $n_z=1$ electron subband and the two lowest subbands for holes (we approximated the actual dispersion by $m_e=0.068$, $m_{hh}=0.35$ and $m_{lh}=0.06$; for the problem of in-plane masses (see, e.g., Ref. 17):

$$n_h = \frac{k_b T}{\pi \hbar^2} \sum_j m_j \ln\left[1+\exp\left(\frac{\mu_h - E_j}{k_B T_p}\right)\right], \tag{1}$$

where j indicates the light- and heavy-hole contributions, E_j are the subband energies, and μ_h is the quasichemical potential of the holes (a similar formula is valid for the electrons). For the offset of the light-hole subband we use 7 meV.[17] Although the influence of the carrier temperature T_p on the evaluation of n_p is small in 2D systems, we assumed a $T_p > T_L$ which increases with the e-h density (see, e.g., Ref. 18) with regard to the reduced cooling rates of GaAs MQW.[1,19] Hence, we use $T_p=150$ K (see, e.g., Ref. 20) in the highest density regime ($n_p \geq 1.5 \times 10^{12}$ cm^{-2}).

In Fig. 2 we present our results concerning the renormalization of the fundamental band gap $E_g'(n_p)$. The data points refer to measurements at $T_L=6$ K and $T_L=13$ K. The difference between the two temperatures is negligible compared to the elevated plasma temperature; actually there is no systematic difference in the experimental data for $T_L=6$ K and $T_L=13$ K, respectively. Data for higher temperatures will be given in Ref. 16. The solid line refers to the calculation in a dynamical-random-phase approximation (RPA) with a single-plasmon-pole approximation (SPP) (Ref. 4) for $m_e/m_h=0.2$ and $T_p=150$ K. Although the carrier temperature is a function of n_p the theory reveals a dependence of the renormalization on T_p which is of minor importance[20] in the density regime under consideration.

The theory is derived for the pure 2D case for which the binding energy of the exciton is $E_{1S}^{2D}=4R_y$ (R_y is the 3D Rydberg constant) and the wave function is

$$\Phi(r) = \frac{1}{\sqrt{2\pi}} \frac{2}{a_0} \exp\left(-\frac{2r}{a_0}\right) \tag{2}$$

(a_0 is the 3D Bohr radius: the convention of Schmitt-Rink and co-workers[3,4] for the 2D Bohr radius a_{2D} is the amplitude radius [see Eq. (3)] $a_{2D}=a_0/2$). On the other hand, the experimental situation of a quasi-2D system is known to be characterized by modified exciton parameters, i.e., the binding energy E_{1S} and the exciton radius a'_{2D}.[12,13] As shown in Ref. 6 it is possible to treat QW as a quasi-2D system using 2D formula but with effective parameters: In the study of the quantum-confined-Stark effect,[13] the authors derived the exciton radius using a variational wave function by assuming a 1S-like orbital in the plane of the layers

$$\Phi(r) = \left(\frac{2}{\pi}\right)^{1/2} \frac{1}{\lambda} \exp\left(-\frac{r}{\lambda}\right), \tag{3}$$

where λ is the amplitude radius of the exciton orbit used as an adjustable parameter. The analysis for 100-Å QW and zero field yields $\lambda=126$ Å and $E_{1S}=8.7$ meV. Using these effective parameters (we define $a_{2D}=\lambda$ according to the convention of Schmitt-Rink and co-workers[3,4]) together with the SPP we get an excellent agreement with the experimental data of up to $n_p=1.4\times10^{12}$ cm^{-2}. Consistently, the measured points for the chemical potential will coincide with a calculated curve $\mu=E_g'(n_p)+\mu_h+\mu_e$.

The deviation at higher densities [see also (Refs. 16 and 20)] could be explained in terms of an enhanced intersubband screening due to the occupation of the $n_z=2$ subband; see, e.g., the absorption spectrum at $I_{exc}=11$ MW/cm^2 or $n_p=2.5\times10^{12}$ cm^{-2}, respectively. The validity of the $n_p^{1/3}$ law[4,10,18] which is an approximation to the full calculation at $T_p=0$ K will be discussed in Ref. 20.

In conclusion, it is shown once more that it is possible to use strict 2D theory to describe QW under the condition that effective parameters are used for the natural units.

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