Temporal Evolution of the Dense Electron-Hole Plasma Phase in GaAs Quantum Wires

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Abstract

We report a spectroscopic investigation of the stationary and transient recombination processes in rectangular GaAs quantum well-wires. The ground level exciton states are studied by linear and non linear photoluminescence excitation spectroscopy, providing information on the one-dimensional exciton binding energy and on the actual band gap edge of the wires. The carrier density dependence and the temporal evolution of the band gap renormalization in GaAs quantum wires have been determined by a line-shape analysis of the time resolved electron-hole plasma luminescence. The obtained data are compared with recent theoretical calculations.

1. INTRODUCTION

When a dense electron hole plasma (EHP) forms in a semiconductor, the electron-hole pair self-energy changes due to many-body interactions, leading to the exciton ionization and to the renormalization of the band gap. The quantitative study of these effects requires a precise determination of the unperturbed ground level parameters of the crystal, namely, the exciton binding energy and the band gap. In this work we have used linear and non-linear (two-photon absorption induced) photoluminescence excitation spectroscopy to precisely characterize the quantum wire array. Based on these studies, we have performed high-excitation intensity time-resolved photoluminescence (PL) measurements to determine the carrier density dependence and the temporal evolution of the band gap renormalization (BGR) and the exciton screening threshold. The absolute values of the BGR were obtained by a line-shape fitting of the time resolved PL spectra at different delays after the exciting pulse and at different excitation intensities.

2. EXPERIMENTAL

The investigated quantum wire array was fabricated by holographic lithography and plasma etching from a GaAs/ $Al_{0.36}Ga_{0.64}As$ multiple quantum well heterostructure (the well and barrier widths were 10.6 and 15.3 nm , respectively). The resulting quantum wires had a lateral width of 60 ± 5 nm. Details on the investigated structure are reported in Refs.[1,2]. The optical characterization of the samples was performed by usual c.w. spectroscopy and by two-photon absorption measurements [3]. The latter experiments were performed by exciting the wires in the transparency region ($\hbar\omega_{exc} \simeq E_g/2$) with a tunable Nd:Yag pumped dye laser down-converted by a low-pressure H_2 Raman cell. The excitation geometry was selected in order to have the laser polarization parallel or perpendicular to the carrier confinement direction ($\epsilon \parallel y$ or $\epsilon \perp y$, respectively). For the time resolved PL experiments the sample was optically excited by the second harmonic of a mode-locked Nd:Yag laser with pulse duration of about 25 ps and repetition frequency of 2 Hz. The temporal evolution of the luminescence was analyzed by a single-shot streak-camera equipped with cooled CCD detector.

3. RESULTS and DISCUSSION

In Fig.1 we compare the one-photon absorption and the two-photon absorption photoluminescence excitation spectra (OPA-PLE and TPA-PLE, respectively) measured at 10 K. In the relatively wide quantum wire array investigated here, the Bohr radius of the 1s exciton is about four times smaller than the wire width. As a consequence the envelope function of the exciton is laterally confined and the center of mass motion of the exciton is quantized [4]. This results in the sharp peaks observed in the linear PLE spectrum of Fig.1a, originating from the radiative recombination of excited center of mass states with odd quantum numbers [5]. The TPA-PLE spectrum, on the other hand, exhibits different features depending on the polarization direction of the exciting field with respect to the carrier confinement direction [6,7].

In particular, when the polarization vector of the exciting field is perpendicular to the carrier confinement direction ($\epsilon \perp y$ geometry), the matrix elements of the TPA process are not vanishing for transitions involving quantum wire states with the same quantum numbers (selection rule $\Delta n_y = 0$). Conversely, in the $\epsilon \parallel y$ geometry only intersubband transitions with $\Delta n_y = \pm 1, \pm 3 \dots$ are allowed. In addition, if excitons are included in the 1D theory of two-photon absorption [6,7], only 2p exciton states related to $\Delta n_y = 0$ transitions are allowed as final states of the TPA process in the $\epsilon \perp y$ configuration. On the contrary, 1s exciton states are allowed in the $\epsilon \parallel y$ geometry for $\Delta n_y \neq 0$ transitions. These additional constraints result from the requirements of parity conservation in the two-photon absorption process. In the light of these theoretical arguments we can interpret the experimental spectra of Fig. 1b. The TPA-PLE spectrum (measured in the $\epsilon \perp y$ geometry) exhibits five structures whose relative energy splittings are the same as those observed for the $\Delta n_y = 0$ transitions in the time resolved [3] and in the stationary luminescence spectra [8] under high photoexcitation condition, i.e. they directly reflect the 1D subband spacing. Furthermore, the first resonance of the TPA-PLE spectrum in Fig.1b is about 3 meV below the onset of the continuum in Fig.1a. This confirms that indeed 2p excitonic states are observed. The estimated binding energy of 3 meV for the 2p state is in good agreement with that predicted for a quantized 1D exciton (4 meV in ideal GaAs quantum wires). We should mention that, in agreement

with the theory, the $\epsilon \parallel y$ TPA spectrum exhibits distinct features corresponding to $\Delta n=0$ transitions with 1s final exciton state (see ref.[3]).

These results permit the precise determination of the quantization energies of quantum wire subbands with quantum numbers as high as $n_y=5$. Furthermore, from the 1s-2p splitting and taking into account the small 2p exciton binding energy, a ground level energy gap of 1.566 eV and the exciton binding of 12 ± 1 meV have been determined. These quantities will thus be used in the following to characterize the unperturbed ground level of the investigated quantum wires .

We can now turn to the time-resolved high-excitation-intensity photoluminescence (HEI-PL) spectra. In Fig.1c, we show a typical HEI-PL spectrum recorded 210 ps after the exciting pulse and with excitation intensity of the order of $1 \text{ MW}/\text{cm}^{-2}$. The whole spectrum has been blue-shifted by 14 meV in order to fix the lowest energy peak at the energy of the continuum E_g . This allows a direct comparison of the PL peaks with the first five $\Delta n=0$ transitions observed in the $\epsilon \perp y$ TPA- PLE spectrum. Clearly, the PL peaks originate from true interband transitions among quantum wire states whose energy splitting roughly coincide with that measured from the 2p exciton peaks in the TPA-PLE spectrum. Actually, a little difference in the energy position is observed because of the small 2p exciton binding energy in the TPA-PLE spectrum. This clearly indicates that under strong photogeneration rate free carriers dominate the recombination processes and the band gap moves towards low energy (the spectrum has been blue-shifted by 14 meV).

In Fig.2 we show several HEI-PL spectra recorded at different power densities, and for delay times of the order of 1 ns , until the PL signal extinguishes completely. As shown by the vertical lines, the relative positions of the PL structures reflect the energy splitting of the first five interband transitions in the wires. In general, at short delays after the exciting pulse the PL spectra exhibit broad tails, indicating the presence of a hot-carrier distribution (high energy tail) and band gap renormalization (low energy tail). With increasing delays, energy relaxation of hot carriers occurs. The spectra narrow and distinct interband transitions

become distinguishable. This time dynamics reflects the temporal evolution of the carrier density in the quantum wire states, which is expected to be maximum after the laser pulse and then to decrease following the e-h pair lifetime in the plasma. Following this plasma dynamics the BGR and the hot carrier temperature are expected to decrease either with decreasing the excitation intensity or with increasing the delay time after the exciting pulse. Qualitatively, a time (i.e. density) dependence of the BGR and of the carrier temperature can be inferred by the progressive narrowing of the low and high energy tail, respectively, observed at long delays in the PL spectra of Fig.2.

To get quantitative information on the temporal evolution of the EHP we have performed a statistical analysis of the luminescence line-shape. Electrons and holes in the plasma are described by Fermi-Dirac distribution functions with common carrier temperature (higher than the crystal temperature). Assuming that the many-body interactions modify the single particle energy states causing a rigid band shift and a quasiparticle broadening, the resulting spectral density of emission for a constant transition matrix element reads

$$I(\hbar\omega) \simeq \sum_{j=1}^N \int dE' \int dE_e \int dE_h \delta(\hbar\omega - E_e - E_h - E_g^*) \cdot f_e(\epsilon_{je}) f_h(\epsilon_{jh}) D(E' - E_g^* - E_{je} - E_{jh}) L_e(E_e, \epsilon_{je}(E')) L_h(E_h, \epsilon_{jh}(E')) \quad (1)$$

where the summation is over N occupied subbands, E_g^* is the renormalized band gap,

$$\epsilon_{je,h} = \frac{m_{h,e}}{m_e + m_h} \cdot (E' - E_g^* - E_{je} - E_{jh}) + E_{je,h} \quad (2)$$

consistent with the assumption of momentum conservation in the recombination, and $D(E)$ is the joint one-dimensional density of states proportional to $E^{-1/2}$. In this model the quasiparticle spectral functions are given by functions $L_{e,h}$ with constant damping parameters $\Gamma_{e,h}$.

Within this model , given the values of the electron and hole 1D subband edges E_{je} and E_{jh} previously determined , and varying the total e-h pair density (n), E_g^* , the temperature T and Γ as fitting parameters, a satisfactory lineshape analysis of the spectra is obtained. The renormalized band gap falls in the low energy tail of the PL spectra and is red shifted

with respect to the $n_y=1$ gap of the unperturbed quantum wire by an amount which depends on the excitation intensity and/or the delay time. The fits get worst for the spectra recorded at short delays and very high carrier densities.

In Fig.3 we plot the band gap renormalization versus the total carrier density. The BGR values were obtained by subtracting the best fit E_g^* values from the unperturbed $n_y=1$ quantum wire gap previously determined ($E_g=1.566$ eV). The analysis was performed on the whole set of spectra excited at different power densities or detected at different delays. The BGR values range between 12 and 25 meV in the carrier density range $10^5 < n < 5 \times 10^6 \text{ cm}^{-1}$. The smallest BGR value obtained from the fits amounts to about 12 meV at $3 \times 10^5 \text{ cm}^{-1}$. This value compares with the exciton binding energy of the investigated wires, indicating that exciton ionization occurs around this carrier density. In Fig.3 we also plot the carrier density dependence of the BGR calculated by Kuang-Hu and Das Sarma [9] for quantum wires of width 500 Å (continuous line). Although this theory neglects the hole population and considers an electron-plasma confined in the lowest conduction quantum wire subband only, it is satisfactory to see that experiment and theory get close. Furthermore, in Fig.3 we show the expected BGR for the 10 nm quantum well from which the wires were fabricated (dashed curve). The curve is given by the well known empirical formula $BGR \simeq .00368 n_{2D}^{-1/3}$ meV, valid for a 10 nm quantum well. In this case the two-dimensional carrier density has been obtained by the scaling rule $n_{2D} = n/L_y$, where L_y is the wire width and n is the one dimensional carrier density obtained from the best fits. This comparison clearly reveals the one-dimensional character of the observed BGR.

In Fig.4 we display the temporal evolution of the total carrier density (n in cm^{-1}), of the BGR and of the carrier temperature as obtained by the line shape fitting of the EHP luminescence excited under different power densities. As already observed in quantum wells [10], the BGR decreases at long delay times, following the reduction of the carrier density due to recombination. The initial photogenerated carrier density (short delays) depends on the intensity of the exciting pulse and shows an exponential decay. The slope of the carrier

density versus time slightly increases with increasing the injected density, indicating some shortening of the recombination time at high carrier density. The band gap renormalization follows this temporal evolution of the electron-hole plasma density. The transient BGR reduces for long delays and the crystal tends to recover the original value of the unperturbed energy gap. The carrier temperatures are deduced from the Fermi-Dirac thermal distribution of the carriers in the subbands, assuming the same average temperature for the electron and hole plasmas. At high excitation density the carrier temperature varies from about 30 meV to 6 meV following an exponential decay in the delay range between 300 and 820 ps. Within the experimental error the KT factor exhibits the same temporal evolution in the investigated carrier density range. Only at the lowest excitation intensity the line-shape fit was reliable enough at short delay and a KT value of about 7.5 meV has been obtained for nominally zero delay. At high carrier densities the KT values are plotted for delays longer than 200 ps, but still a hot plasma is present in the wires. From the data of Fig.4 we can estimate an energy loss rate for electrons and holes ranging between $7 \cdot 10^{-13}$ and 10^{-12} W/carrier. These data, though approximated, are smaller than those expected from the calculations of Campos and Das Sarma [11] valid for a 20nm*20nm wire with a single subband occupied at carrier density 10^6 cm^{-1} . More sophisticated experiments and detailed theories including the multisubband population are probably necessary to elucidate this aspect of the EHP physics in quantum wires.

In conclusion, we have determined the carrier density dependence and the temporal evolution of the band gap renormalization in GaAs quantum wires. The obtained data compare satisfactorily with recent theories.

Acknowledgements

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Fig.1 - (a) One-photon absorption photoluminescence excitation (OPA-PLE) spectrum of the wires at 10 K. The two peaks are quantized center of mass states of the free exciton ($n=1$ and $n=3$ states). The vertical line indicates the continuum (E_g).

(b) Two-photon absorption photoluminescence excitation spectrum (TPA-PLE) at 10 K. The peaks are the 2p excitons with selection rule $\Delta n_y=0$. The numbers indicate the electron and hole quantum wire subbands. The vertical lines correspond to the PL peaks (bottom spectrum).

(c) High excitation intensity time resolved photoluminescence spectrum (HEI-PL) recorded at 5 K and 210 ps after the exciting pulse.

Fig.2 - Time resolved electron-hole plasma luminescence spectra recorded under different excitation intensities ($I_0=0.5 \text{ MWcm}^{-2}$).

Fig.3 - Band gap renormalization (dots) versus total carrier density obtained from the fit of the luminescence spectra under different excitation intensities and after various delays. The continuous line is the calculated BGR curves for quantum wires of widths 500 Å (after ref. [9]). The dashed line is the expected BGR for the 10 nm quantum well from which the wires were fabricated.

fig. 2

fig. 3

Fig.4 - Temporal evolution of the total carrier density (a), of the band gap renormalization (b), and of the carrier temperature (c), obtained from the fit of the time resolved luminescence spectra under different excitation intensities ($I \simeq 0.5 MWcm^{-2}$).