Unusual intensity dependence of rise and decay times of free exciton photoluminescence in high purity GaAs and AlGaAs alloys

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Abstract. Rise up and decay dynamics in high purity GaAs and direct-gap AlGaAs alloys were studied. In contrast to the previous results both the rise and decay times of the free exciton PL line show strongly non-monotonic behaviors with increasing in excitation power. Sharp increase of these times with the pulse power increasing from 0.02 pJ to 0.1 pJ is a result of the change exciton formation mechanisms from geminate to bimolecular. The decrease in rise and decay times at high excitation powers can be due to a decrease of the exciton lifetime due to Auger-like processes.

1. Introduction

At present the dynamics of exciton formation and relaxation in III-V semiconductor structures has been the subject of intense research for fundamental and applied reasons. The experimental method commonly utilized for the investigation of exciton formation processes is time-resolved photoluminescence (PL). In the last decade it has been shown for GaAs quantum well structures [1,2] that the rise and decay times of exciton PL monotonically increase with increasing excitation power. Since only excitons with low center-of-mass kinetic energies can recombine radiatively, this phenomenon was explained in terms of a cooling of the excitonic gas in the time after excitation [2].

We report here on an unusual rise up and decay dynamics in high purity GaAs and direct-gap AlGaAs alloys. It was found that in contrast to the previous results both the rise and decay times of the free exciton PL line show strongly non-monotonic behaviors with excitation power. These non-monotonic behaviors cannot be explained by the effect of cooling of the excitonic gas.

2. Experimental

The studied layers of high quality GaAs and $Al_xGa_{1-x}As$ over the $0 \le x \le 0.21$ composition range were obtained by molecular beam epitaxy. The details of the layer growth were

described elsewhere [3]. The time-resolved PL was excited using a dye laser synchronously pumped by a mode-locked Ar-ion laser. This resulted in about 70 ps duration pulses at a wavelength of 580 nm. The intrinsic repetition rate of 80 MHz set by the Ar ion laser cavity length was reduced to 4 MHz using a cavity damper. The PL was analyzed using a CROMEX 250IS spectrometer and detected by a Hamamatsu C4334 Streakscope camera. The time resolution of the system was better than 50 ps. Samples were placed in a closed-cycle CTI-Cryogenics cryostat. The laser spot focused on the sample had a Gaussian shape with a half-width of 250 μm. The image of the excited spot was projected onto the entrance slit of the monochromator without magnification. Since the monochromator slit widths of 10–20 μm were used, the data were recorded with a quasi-uniform excitation.

3. Results

Figure 1 shows the rise and decay dynamics of free exciton PL intensity in high purity GaAs layer under non-resonant excitation at different excitation pulse powers.

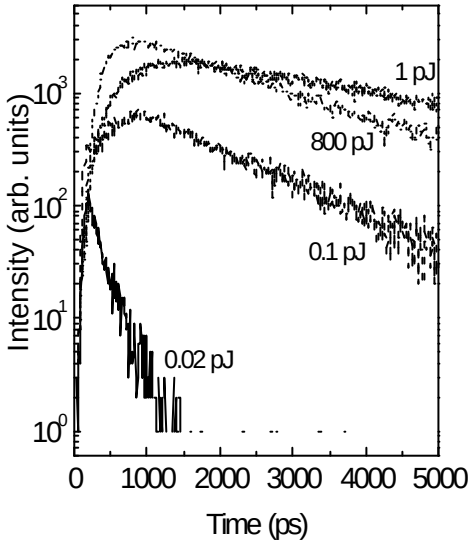

Figure 1. Rise and decay dynamics of free exciton PL intensity in high purity GaAs under non-resonant excitation at excitation pulse powers of 0.02, 0.1, 1, and 800 pJ.

One can see that the character of exciton dynamics drastically changes with increasing excitation pulse power. At the low excitation pulse power of 0.02 pJ the exciton intensity sharply rises in 200 ps after the excitation pulse, and then decays with a time constant of 210 ps. An increase of the pulse power (P) from 0.02 pJ to 0.1 pJ results in slowing down the rise of excitonic PL for as long as 900 ps, while the decay time constant increases up to 1500 ps. Since we have shown recently that under these excitation conditions the contribution of non-radiative recombination is negligible in our layers [3], the changes in rise and decay times are not a result of saturation of non-radiative traps.

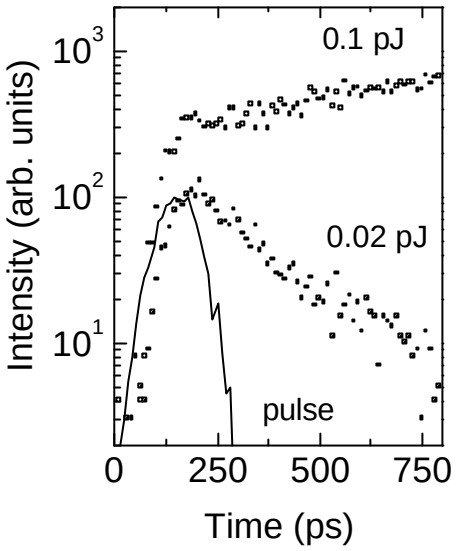

Figure 2. Rise and decay dynamics of free exciton PL intensity in high purity GaAs at excitation pulse powers of 0.02 and 0.1 pJ.

We would like to note that the exciton dynamics at P = 0.1 pJ as it is clearly seen in fig.2 is a sum of two curves. There one of these curves results in a rise on the exciton dynamics at the time of 200 ps and is similar to the low excitation pulse power curve. An increase of the excitation pulse power up to 1 pJ leads to an eliminating of the rapid component in exciton dynamics and both the rise and decay times increases with increasing P to reach the maximum values of 1700 ps and 3800 ps, respectively. A further increase of P leads to a monotonic decrease of these values down to 700 ps and 2000 ps, respectively, at P=800 pJ. The AlGaAs layers show the same character of intensity dependence of excitonic rise and decay times. However, the maximum values of the rise and decay times slowly decrease with the increase of the AlAs fraction in the alloy down to values of 700 ps and 2100 ps, respectively, at x=0.21. The rise and decay times of exciton PL intensity in GaAs layer as shown in fig.3 as a function of excitation power.

4. Discussion and Conclusions

The experimental data show that the rise and decay times of exciton PL intensity strongly change with excitation power. We found that (i) the rise and decay exciton dynamics actually contains two components a fast and a slow part. The fast component dominates at a low excitation power, while the slow component increase rapidly with increasing excitation power and finally overwhelms the fast component. (ii) The duration of the slow component non-monotonically depends on excitation power.

Considering first the origin of the two components in exciton dynamics at low excitation power, these are connected with two different mechanisms of the free exciton formation geminate and bimolecular, which are realized at non-resonant photoexcitation [4]. The geminate formation process can be described as follows: excitation of an

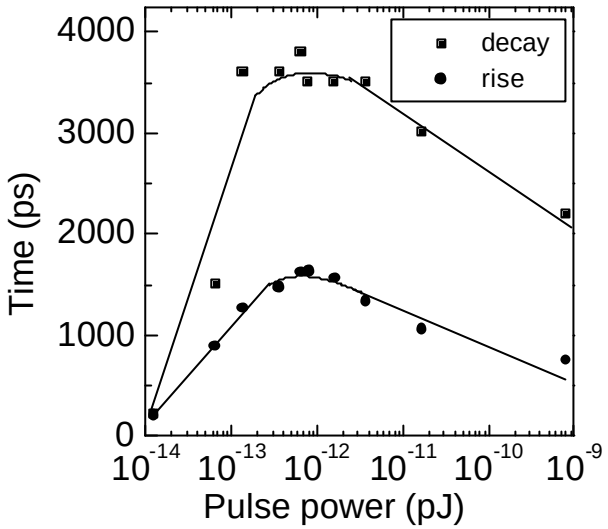

Figure 3. Rise and decay times of exciton PL intensity as a function of excitation power. Solid lines are used as …

electron-hole pair by a photon, propagation of the created pair through the crystal with total momentum close to zero, and finally conversion of the correlated pair into the exciton via interaction with a phonon. In the bimolecular process exciton formation consists in random binding of non-correlated electrons and holes via Coulomb interaction. We are not aware of any theoretical works devoted to the probability of exciton formation in bulk GaAs via the geminate process. However, the computation for GaAs/AlGaAs quantum well structures shows that excitons have to form mainly via geminate processes at low excitation and via bimolecular processes at high excitation [5]. Indeed, the geminate process is linear with excitation and realized during the pump pulse only, while the bimolecular process is quadratic with excitation and it can last during long time after the pump pulse.

Thus increase in pulse power to a moderate level results in a domination of the bimolecular exciton formation process. As it has been shown recently [2] excitonic rise and decay times in this case should increase with initial temperature of excitonic gas, which increases with excitation power. Following Yoon *et al.* [2] we estimated the initial temperature of the excitonic gas from the high-energy slope of band-to-band recombination and observed that this temperature monotonically increases with pulse power in the entire power range used. However, both rise and decay times monotonically decrease with increasing excitation for pulse power higher than 1 pJ. We suppose that this phenomenon is a result of decrease in the exciton lifetime due to the appearance of an additional rapid channel for exciton decay. Since at high excitation pulse power the exciton concentration becomes very high, we associate this rapid channel with Auger-like processes.

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