Dynamics of Excitonic Polaritons in Semiconductor Heterostructures with Quantum Wells

A. V. Trifonov, Yu. P. Efimov, S. A. Eliseev, V. A. Lovtcius, P. Yu. Shapochkin, and I. V. Ignatiev
St. Petersburg State University, Russia

Abstract— Exciton dynamics is experimentally studied for a heterostructure with a wide InGaAs/GaAs quantum well. The exciton-phonon, exciton-exciton, and excitons-carrier scattering is studied by means of reflectance spectroscopy in different experimental conditions. The non-radiative broadening of exciton resonances is extracted from the experimental data and used as a measure of the scattering efficiency. The exciton-phonon scattering is studied at different sample temperatures. The exciton-exciton and exciton-carrier scattering is studied at an additional illumination of the sample with radiation of a CW tunable laser with different photon energies. Direct information about the characteristic time of relaxation processes is obtained in the spectrally resolved pump-probe experiments.

1. INTRODUCTION

Modern development of molecular beam epitaxy technology allows one to grow heterostructures of ultimate quality where the dynamics of optical excitations is mainly governed by fundamental physical processes. Exciton properties of such systems, in particular, exciton dynamics are intriguing problems attracting researcher interest for a long time. Exciton-light coupling in heterostructures with quantum wells (QWs) is extensively discussed in textbook by Ivchenko [1]. Early studies of exciton dynamics are summarized in the well-known textbook by Shah [2]. Further progress in the structure growth led to creation of high-Q microcavities with QWs, where the strong exciton-light coupling allows one to observe large splitting of exciton polariton modes, Bose-Einstein condensation, multi-stability, vacuum Rabi oscillations, and many other interesting effects [3].

The direct excitons in III-V semiconductor heterostructures with QWs may be strongly coupled with electromagnetic radiation so that the basic states of the system are the exciton-polariton states [3]. These states may be split by the exciton-light interaction into the polaritonic states with the energy separation called as the vacuum Rabi splitting, which is larger than the broadening of respective polariton transitions. This is the case for the high-quality microcavities with QWs. In this paper, we will discuss another case when the Rabi splitting is small comparing to the broadening and we may speak about the exciton transitions. This is a typical situation for the heterostructures with no microcavities. The fundamental process causing the exciton line broadening is the radiative decay. Characteristic time of the process for radiative excitons in GaAs/AlGaAs and InGaAs/GaAs QWs is of order of 10 ps or shorter that corresponds to the broadening of several tens of μeV [4].

Besides this process there are several other mechanisms causing the exciton line broadening. In real structures there is some amount of defects in the QW or neighboring barrier layers, which may create a fluctuating potential for excitons localizing them. The localization results in a spread of the exciton transition energies and in a spread of the exciton lifetimes. These effects greatly hinder the study of fundamental exciton processes so that the high quality of heterostructures with QWs is of principal importance to obtain reliable data about fundamental dynamic processes in an excitonic ensemble.

In present work we discuss several fundamental processes occurring in the exciton ensemble in a high-quality heterostructure with the relatively wide $In_{0.02}Ga_{0.98}As/GaAs$ QW, which width $L = 95$ nm. Details of the heterostructure growth and optical characterization are described in Ref. [5]. The large width of the QW allows one to observe several quantum confined exciton states and respective transitions in the reflectance and photoluminescence (PL) spectra. The ultimate quality of the structure is evidenced in the absence of noticeable Stokes shift of PL lines relative to those observed in reflectance spectra. The fundamental processes discussed here are the exciton radiative decay, the exciton-phonon interaction, the exciton-exciton scattering, and the exciton-free-carrier scattering. These processes may be studied by both the CW and time-resolved spectroscopy. Details of experimental technique are described in Ref. [5].

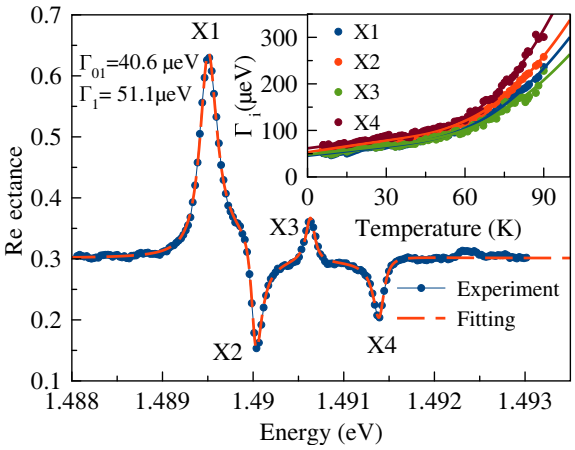

Figure 1: Reflectance spectrum of a heterostructure with the 95-nm InGaAs/GaAs QW (solid noisy curve) and the fit of the spectrum by the generalized Ivchenko model (red dashed curve). Sample temperature $T = 4\,\mathrm{K}$. Inset shows the temperature dependence of the non-radiative broadening, $\hbar\Gamma_1,\ldots,\hbar\Gamma_4$.

2. CW SPECTROSCOPY

A careful analysis of reflectance spectra allows one to obtain quantitative information about the exciton line broadenings caused by the exciton radiative decay, $\hbar\Gamma_{0N}$, and by various non-radiative processes, $\hbar\Gamma_N$. Here N numerates the quantum confined exciton states. An example of the reflectance spectrum of the structure under study is shown in Fig. 1. Several peculiarities of the spectrum marked by X1, ..., X4 are due to the exciton transitions from the ground (vacuum) state of the structure to the different quantum confined exciton states in the QW.

The spectrum is well described by a phenomenological model developed by Ivchenko and Andreani [1] and generalized in Ref. [5]. The reflectance spectrum is described by expressions:

$$R(\omega) = \left| \frac{r_{01} + r_{QW} e^{2i\psi}}{1 + r_{01} r_{QW} e^{2i\psi}} \right|, \tag{1}$$

$$r_{QW} = \sum_{N=1}^{N_{max}} \frac{i(-1)^{(N-1)} \Gamma_{0N} e^{i\phi_N}}{(\omega_{0N} - \omega) - i(\Gamma_{0N} + \Gamma_N)}. \tag{2}$$

Here ω_{0N} is the frequency of the N-th exciton transition, Γ_{0N} is the exciton decay rate and Γ_N is the rate of various scattering processes broadening the exciton line. Other notations are described in Ref. [5]. In Fig. 1, parameter Γ_{0N} determines the amplitude of peaks and dips while sum $(\Gamma_{0N} + \Gamma_N)$ determines their width. So, a study of reflectance spectra at different experimental conditions varying the dynamic processes in the exciton ensemble and thus changing the radiative and non-radiative constants allows us to study these processes.

3. EXCITON-PHONON SCATTERING

Exciton-phonon scattering in the GaAs based heterostructures is relatively weak at low sample temperatures. Respective relaxation time of hot excitons with emission of acoustic phonons is of order of several tens of picoseconds [2]. This time corresponds to a weak exciton line broadening of order of several μeV, which is small comparing to the radiative broadening of the lowest exciton transition in the QW heterostructures.

The exciton-phonon scattering may be strongly enhanced when the sample temperature is increased. Inset in Fig. 1 shows the non-radiative broadening of four exciton transitions as a function of sample temperature obtained by the experimental study of reflectance spectra at different temperatures followed by the data processing using Eqs. (1), (2). The temperature dependence of the phonon-mediated line broadening can be described by function [6, 7]:

$$\hbar\Gamma_N = \hbar\Gamma_N(0) + \hbar\gamma_{\mathrm{ac}} T + \frac{\hbar\gamma_{LO}}{\exp\left(kT/E_{LO}\right) - 1}. \tag{3}$$

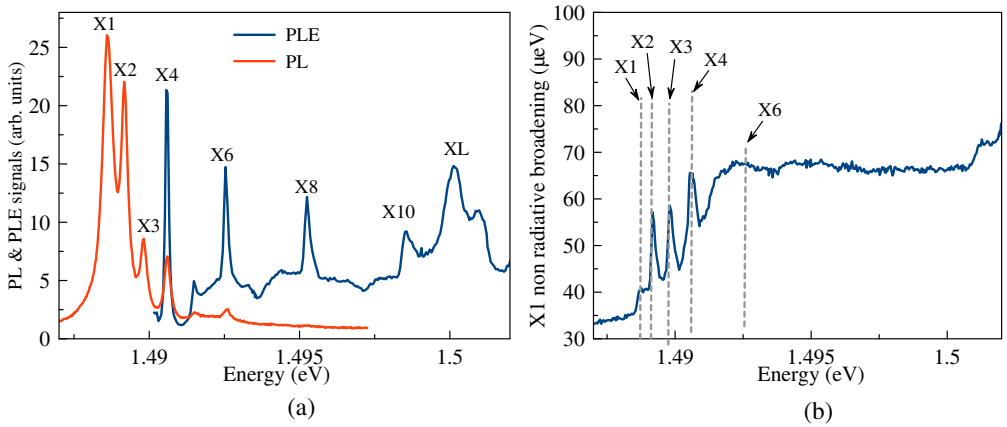

Figure 2: (a) PL and PLE spectra of the sample in the range of several exciton transitions marked by X1, ..., X10. The transition to the light-hole exciton is marked as XL. (b) The non-radiative broadening of the lowest exciton transition, $\hbar\Gamma_1$, obtained from reflectance spectra as a function of photon energies of an additional excitation by a CW tunable laser. The power density of the excitation is of about 0.1 W/cm^2. Sample temperature, $T = 5$ K.

Here the first term describes the non-radiative broadening at zero temperature, the second term describes the increase of the broadening related to the exciton-acoustic-phonon scattering, and the last term described the effect of exciton-LO-phonon scattering. At temperatures $T \leq 60$ K, the broadening linearly rises with temperature that indicated that the exciton-acoustic-phonon scattering plays the main role in this temperature range. Quantity $\hbar\gamma_{\rm ac}$ characterizing this process is: $\hbar\gamma_{\rm ac} = 0.65$ μeV/K for the X1 exciton resonance and $\hbar\gamma_{\rm ac} = 0.75$ μeV/K for the X4 resonance. These values are a bit smaller than that obtained in Ref. [7]: $\hbar\gamma_{\rm ac} = 1.1$ μeV/K.

At higher temperatures, $T > 60$ K, a stronger increase of the non-radiative broadening is observed. It is explained by the exciton-LO-phonon scattering well described by the last term of Eq. (3) with parameters: $\hbar\gamma_{LO} = 5.3$ meV (13 meV) and $E_{LO} = 29$ meV (33 meV) for resonances X1 (X4). These date are similar to those reported in Ref. [7].

Careful analysis of reflectance spectra measured at different temperatures shows that there is no valuable variation of the exciton-light coupling constant, $\hbar\Gamma_{0N}$, within experimental error of about ±3 μeV. The energy position of exciton resonances nicely follows the temperature variations of the band gap energy for GaAs.

4. EXCITON-EXCITON AND EXCITON-CARRIER SCATTERING

Exciton-exciton scattering is another important process strongly modifying the exciton dynamics. Under the non-resonant optical excitation above the lowest exciton transition, the excitons with large in-plane wave vector may be created due to the phonon-mediated relaxation. When the wave vector is larger than that of photons that is the excitons are beyond the light cone, they do not interact with light and therefore cannot radiatively recombine. In the high-quality structure, these excitons may live for a long time (nanoseconds) and be accumulated up to a large density at relatively weak optical excitation, as experiments show [5]. The scattering of radiative excitons with the excitons from this non-radiative reservoir gives rise, in particular, to the exciton line broadening.

We have studied the effect of line broadening in reflectance spectra when the exciton reservoir was populated by an additional optical excitation. For the excitation, we used a CW tunable laser with photon energies varied in the large range above the lowest exciton transition. An example of experimental results is shown in Fig. 2.

In Fig. 2(a), the PL and PL excitation (PLE) spectra clearly show the exciton resonances corresponding to the optical transitions from (or to) the quantum confined exciton states. The peak amplitude of the resonances gradually decreases with the photon energy rise that points out the decrease of oscillator strength of the transitions theoretically predicted, e.g., in Ref. [8]. Besides the resonances, a background signal (pedestal) in the PLE spectrum is observed for the photon energies $\hbar\omega > 1.4915$ eV that is approximately 3 meV above the lowest exciton transition seen in the PL spectrum. This non-resonant signal is mainly due to optical transitions with creation of free

electrons and holes. Although the exciton binding energy is of about 4 meV, the excitons created with the excess energy $\Delta E > 3$ meV, e.g., in the 2s-state, may dissociate into free electrons and free holes acquiring a deficient energy from the crystal lattice due to its finite temperature.

Figure 2(b) shows the behavior of non-radiative broadening of the X1 exciton resonance in reflectance spectra when the sample is additionally illuminated by a CW laser radiation with different photon energies. The spectrum, which can be called as the excitation spectrum of non-radiative broadening, is obtained by means of theoretical analysis with the use of Eqs. (1), (2) of the reflectance spectra measured for each photon energy of the illumination. As seen in the small spectral range, $\Delta E < 3$ meV, above the X1 exciton transition, the excitation spectrum reveals exciton resonances similar to those observed in the PL and PLE spectra. At the same time, the lowest exciton resonance, X1, is strongly suppressed. This means that the CW excitation to this resonance does not affect its width. The physics is clear: such excitation creates cold excitons with small in-plane wave vector, which rapidly emit photons and, therefore, cannot be accumulated. Correspondingly, the exciton-exciton scattering is a rare event for the small power density of the illumination (< 100 mW/cm^2) not noticeably affecting the exciton line width.

The CW excitation into resonances X2, ..., X4 creates hot excitons. Their relaxation to the lowest energy states with emission of acoustic phonons may create the non-radiative excitons with large wave vectors acquired from the phonons. As it is already mentioned above, such excitons may be accumulated and may efficiently scatter the radiative excitons. This is the reason why the CW excitation into the X2, ..., X4 resonances strongly increases the width of the X1 resonance. Between the resonances, the light absorption is low and the non-radiative reservoir is inefficiently populated.

The CW illumination above the X4 resonance results in strong increase of the X1-resonance broadening, which becomes almost independent of photon energy of the illumination up to $E = 1.502$ eV. No resonant increase of the broadening at the exciton resonances X6, ..., X10 is observed in this spectral range. This means that the mechanism of the broadening is changed in this spectral range. We may assume that the broadening is due to exciton-free-carrier scattering. This scattering is more efficient, at least by one order of magnitude, than the exciton-exciton scattering [9, 10]. Further increase of photon energy of the illumination gives rise to the further increase of the X1-resonance broadening, which is probably caused with the increase of light absorption via transitions into the light-exciton (XL) states.

5. PUMP-PROBE EXPERIMENTS

Pump-probe experiments give more direct information about characteristic times of dynamic processes [2, 5]. We have performed the spectrally-resolved pump-probe experiments when the picosecond pump pulse excites the X1 exciton state while the spectrally broad femtosecond probe pulse is used to detect the reflectance spectrum at some delay after the pump pulse. An analysis of the spectra measured at different delays using Eqs. (1), (2) allows one to obtain the dynamics of all the characteristics of the exciton state under study.

Figure 3 shows as an example of experimental results obtained by such way, namely the energy position of exciton resonance X1 as a function of the delay. As seen, the resonance is shifted from its initial position to the higher energies (blue shift) at zero delay and then relaxes toward its initial position when the delay increases. The magnitude of the shift, $\delta E_{X1} \approx 60\,\mu$eV, is comparable to the half width at the half maximum (HWHM) of the resonance, HWHM $\approx 90\,\mu$eV, in the absence of pumping. This effect clearly shows that the standard pump-probe method when the dynamics is studied at the fixed photon energy may give a wrong decay time of the process under study.

The physical origin of the blue shift is probably similar to that observed for polaritons in structures with microcavities [3]. Namely, the pump pulse creates some amount of radiative excitons with repulsive interaction between them, which shifts the exciton state. The fast component of the shift is related to the decrease of exciton density caused by their radiative recombination. So, the decay time, $\tau_{X1} = 6.7$ ps obtained in this experiment corresponds to the recombination time of the excitons.

There is also a slow component of the decay, which is extensively discussed in Ref. [5]. It is related to the non-radiative excitons with large in-plane wave vectors, which can be created even at the resonant excitation because of the finite spectral width of the pump pulses (of about 1.4 meV) and of the exciton-acoustic-phonon scattering. The effective time of the slow decaying component varies from the units to tens of nanoseconds, depending on the experimental conditions [5].

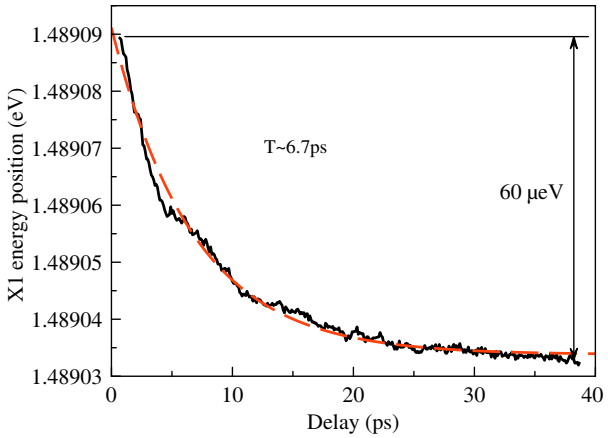

Figure 3: Dynamics of the blue shift of the exciton resonance X1 measured in pump-probe experiments.

6. CONCLUSION

The study performed for a high-quality heterostructure with a wide QW shows that the dynamics of radiative excitons, besides the radiative recombination, is governed, in a general case, by several fundamental processes, namely, the exciton-phonon, exciton-exciton, and exciton-carrier scattering. Contribution of each of the processes strongly depends on the experimental conditions used. At the strictly resonant excitation into the lowest exciton transition when only the radiative excitons are created, the dynamics is mainly determined by the recombination process. Other experimental conditions to make this process preferable are the low sample temperature and the weak excitation power, when the areal density of radiative excitons is low. With increase the temperature, the exciton-phonon scattering broadens exciton resonances and creates non-radiative excitons, which are accumulated due to their long lifetime. The non-radiative excitons are also created at the non-resonant excitation of excitons. The accumulation of the non-radiative excitons gives rise to an efficient exciton-exciton scattering broadening the exciton resonances. When the photon energy is large enough to create free electrons and holes, the exciton-carrier scattering efficiently broadens the exciton resonances even at the small excitation density. The increase of the excitation density strengthens the exciton-exciton and exciton-carrier scattering processes. Even when the resonant, but strong enough, excitation is used, the repulsive interaction of excitons causes noticeable dynamic blue shift of exciton resonances when the distance between the excitons becomes comparable with their diameter.

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