Luminescence of HH-excitons in GaAs/GaAlAs superlattices under resonant excitation

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Abstract. The spectra and kinetics of photoluminescence decay of GaAs/GaAlAs superlattice were measured under resonant excitation in the temperature range of 5–40 K. A narrow peak resonant with the frequency of excitation radiation was found in the luminescence spectra. This peak can be attributed to the resonant luminescence of excitons. The exciton radiative recombination rate and its dependence on temperature were determined experimentally.

Introduction

The probability of radiative recombination of free excitons in quasi-2D structures (quantum wells and superlattices) is proportional to the number of sites in a crystal lattice participating in collective excitation, i.e. it is determined by the exciton coherence length. According to theoretical estimations [1, 2], in an ideal heterojunction, where this length is limited only by the wavelength of radiation, the time of exciton radiative recombination in GaAs quantum wells is of the order of several picoseconds. These estimates were confirmed by recent pump-probe experiments on high quality GaAs/AlAs superlattices [3]. In the structures of lower quality, the scattering on interface inhomogeneities and internal defects leads to a reduction of exciton coherence length and finally results in its localization. The localization of excitons in GaAs quantum wells was observed in experiments on four-wave mixing [4]. According to the above statements, the radiative recombination rate is defined by the exciton coherence length. This means that a degree of excitons localization in the structure could be directly estimated by the measurement of the excitonic luminescence kinetics. Modern high-speed techniques allow us to perform such measurements in real time, which simplifies the experiments and makes their interpretation more reliable. Such experiments were performed on GaAs/AlAs structures under nonresonant excitation [5].

1 Experimental

In this paper we present a study of luminescence spectra and its kinetics in the GaAs/GaAlAs superlattice under resonant excitation. We investigated a structure of standard quality, grown by solid-source molecular-beam epitaxy (MBE) without interruption on heterointerfaces. The structure consisted of 50 periods of the superlattice 3.0 nm GaAs/3.8 nm Ga_{0.56}Al_{0.44}As. Photoluminescence (PL) was excited with a tunable Ti-sapphire laser with a pulse duration of about 7 ps. The resultant PL was dispersed by a double-grating spectrometer (U1000) with a photon counting system. We used a streak-camera for registration
2 Results and analysis

At low (5–10 K) temperatures the sample exhibits very strong PL with a single excitonic line at \( \approx 1638 \text{ meV} \) with a half-width of 4.5 meV. The photoluminescence excitation spectrum (PLE) also has an excitonic peak with almost the same half-width (5 meV) and is shifted to a high-frequency range by 3 meV. The Stokes shift comparable to the linewidth indicates a significant inhomogeneous broadening of an excitonic band. The luminescence spectrum under selective resonant excitation is presented in Fig. 1. In addition to the wide band of excitonic luminescence, a narrow peak was observed, the spectral position of which coincides with the frequency of the pumping light. The peak width is equal to the spectral resolution of the registration system. When the temperature increases to 40 K (Fig. 1) the luminescence completely disappears, and the intensity of the narrow peak significantly decreases. It is clear that, at this temperature, the PL is completely quenched and only scattered laser radiation is registered. Since the efficiency of scattering should not depend on temperature, the essential enhancement of the narrow peak intensity at low temperature can be interpreted as a result of a resonant luminescence, spectrally coincident with the laser radiation.

This conclusion was confirmed by the results of time-resolved measurements in which the kinetics of PL as a function of detection wavelength was measured. As is shown in Fig. 2, the PL intensity has a distinctly asymmetric shape—a sharp forward front and a longer tail. Under the resonance conditions, when the excitation and detected wavelengths coincide, the PL response has the shortest duration; however, its decay time still considerably exceeds the time resolution of the recording system.
Excitons in Nanostructures

From the above discussion it follows that the main contribution to the signal at the resonant frequency is from the luminescence of the sample rather than from a scattered laser radiation. Thus, the narrow luminescence peak is caused by the radiation of excitons resonantly selected from an inhomogeneously broadened band by the excitation radiation. The luminescence kinetics in a wide excitonic band is characterized by the delayed forward front and greatly extended tail. This behaviour of the PL decay is a characteristic attribute of spectral diffusion. The rather high integral intensity of the narrow peak means that the radiative recombination rate under resonant conditions \((1/\tau_r)\) does not concede to the rate of the spectral diffusion \((1/\tau_{sd})\). The relaxation time of a resonant PL \((\tau)\) which can be obtained experimentally is determined by the equation:

\[
1/\tau = 1/\tau_r + 1/\tau_{sd}.
\]

The ratio of the integral intensity of the narrow peak to the total luminescence intensity is represented by:

\[
I_r/I = (1/\tau_r)/(1/\tau_r + 1/\tau_{sd}).
\]

The analysis of the spectral and kinetic measurements provides the value of the exciton radiative recombination rate in the sample to be \(1/\tau_r = 1/200 \text{ ps}^{-1}\). As was mentioned above, the radiative recombination rate strictly corresponds to the exciton coherence length \((r_{coher})\):

\[
\tau_r/\tau_{r0} = (\lambda/r_{coher})^2,
\]

where \(\lambda = 200 \text{ nm}\) is the wavelength of the resonant radiation in GaAs. Experimentally determined \(\tau_r\) corresponds to the \(r_{coher} = 30 \text{ nm}\), which is larger than an excitonic Bohr radius in the structure \(a_B = 10 \text{ nm}\).

Based on the above results, we can conclude that the excitons in the studied sample are not totally localized, however, at low temperature, the efficiency of the energy exchange between the excitons, specifying the rate of the spectral diffusion, is not too high.

As the temperature increases, both the relaxation time and PL intensity decrease, which indicates the presence of an additional channel of nonradiative degradation of excitation. At the same time, the analysis shows (see Fig. 3), that with increasing temperature, the exciton radiative recombination rate decreases. The decrease of the 2D-exciton's radiative
Recombination rate with temperature was observed by several authors [6–8]. However, up to now these results have no unambiguous interpretation. According to [6, 7], the main reason for this effect is thermal population of nonradiative excitonic states characterized by large magnitudes of the wavevector. Theoretical evaluation predicts, for this case, a linear temperature dependence of $\tau_r$ [6], which does not agree with our experimental data (see Fig. 3). Another reason for the increase of $\tau_r$ is a decrease of the exciton coherence radius as a result of scattering of excitons by thermal phonons [8]. In this case, the temperature dependence of $\tau_r$ can be described by the relation:

$$\tau_r(T) = \tau_r(0) / [1 - A \exp(\frac{E_{ph}}{kT})].$$

Here, $E_{ph}$ is the energy of the scattered phonon, and the coefficient $A$ is specified by the exciton-phonon coupling. The dependence calculated using this relationship (solid line in Fig. 3) is in excellent agreement with the experimental results (The value $E_{ph}$, used as a fitting parameter, is about $13 \text{ cm}^{-1}$). Thus, we can conclude that the growth of $\tau_r$ with temperature, in our sample, is determined by the exciton-phonon scattering, which reduces the radius of the exciton coherence.

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**References**