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Time-resolved studies of exciton recombination in direct-gap GaAs/AlAs superlattices

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Abstract. The temperature dependence of the recombination dynamics of excitons was investigated by time-resolved photoluminescence spectroscopy in direct-gap GaAs/AlAs superlattices. Peculiarities of excitonic photoluminescence decay were established in the dependence of the well width. In particular, it was shown that a reduction of the well width results in faster exciton recombination due to an enhancement of interface influence.

Introduction

GaAs/AlAs superlattices (SL's) are considered to be a very perspective system for device applications, including light diodes and low-threshold lasers. For these purposes, detailed knowledge of the recombination processes which take place in SL's is very important. Previously, it was shown that the measured radiative lifetime of free excitons in such structures lies in the nanosecond time regime, which significantly exceeds the expected value [1, 2]. Exciton scattering on alloy disorder, interface corrugations, etc. results in a decrease of the exciton coherence length so that the radiative lifetime is enhanced. A reduction of the well width leads to a strong increase of the exciton binding energy. At the same time, the influence of the interfaces on exciton recombination is significantly enhanced.

In this paper, we report a detailed study of the photoluminescence (PL) dynamics in several direct-gap GaAs/AlAs SL's with different well thickness. In the SL with a layer thickness of the order of the exciton Bohr radius, the carrier dynamics was found to be dominated by the transfer from light-hole to heavy-hole excitons. When the layer thickness becomes smaller than the exciton radius, the dynamics of free excitons at low temperatures is controlled by localization.

1. Experimental

The $(GaAs)_n/(AlAs)_m$ SL's, where n and m denote the respective layer thickness in monolayers, were grown by molecular beam epitaxy on semi-insulating (100)-GaAs substrates. The well and barrier thicknesses were, respectively, $n = 9$, 14, 21, 36 ML's and $m = 4$, 7, 21, 36 ML's. We will label the samples as n/m SL.

The time-resolved PL experiments were performed in a He-flow cryostat allowing a temperature variation between 4.2 K and room temperature. The samples were excited by an Ar^+-ion laser pumped, frequency-doubled Ti:sapphire laser operating at 3.37 eV with a pulse width of 150 fs. The PL signal was dispersed by a 0.22 m monochromator

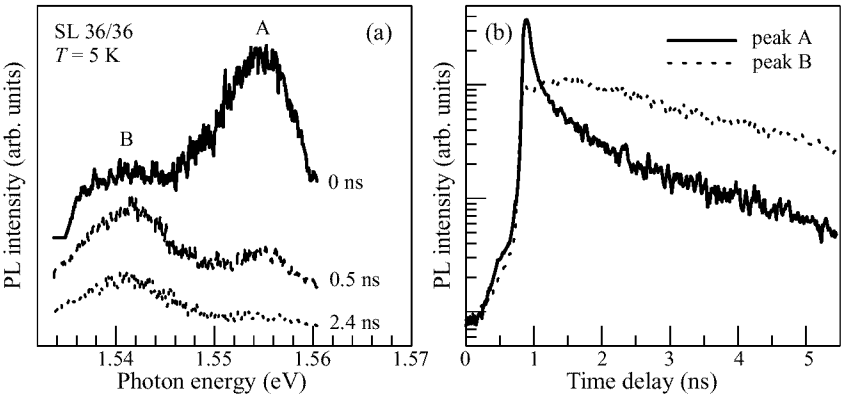

Fig. 1. (a) Time evolution of the PL spectrum in the 36/36 SL at 5 K. The two peaks at 1.555 and 1.541 eV are labelled A and B, respectively. (b) PL intensity vs time for two detection energies, which correspond to the two maxima of the PL spectrum.

using gratings with 1200 and 600 lines/mm. The temporal resolution of the PL signal was achieved by a streak camera system (Hamamatsu C5680) operating in synchro-scan and single-shot modes. The minimum time resolution is 2 ps for synchro-scan and 50 ps for single-shot operation. The spectral resolution is 0.2 nm for a 1200 lines/mm grating. The excitation power of the laser was adjusted to 70 μW.

2. Results and discussion

For direct-gap SL's, the recombination takes places between electrons and holes at the Γ-point of the GaAs wells. A typical PL spectrum of a direct-gap SL consists of one narrow line due to the recombination of direct excitons, i.e., electrons and heavy holes are confined within the same well. However, if the splitting between the heavy- and light-hole subbands is less than LO-phonon energy, thermalization of unequilibrium holes into the heavy-hole subband is slowered and an additional PL line due to light-hole exciton recombination can be observed [3]. For the 36/36 SL, the PL spectra shown in Fig. 1(a) consist of two peaks at 1.555 and 1.541 eV labelled A and B, respectively. The energy separation of 14 meV is close to the calculated splitting of 18 meV between the heavy- and light-hole states. Since a typical variation of the confinement energy due to monolayer fluctuations is about 3 meV for one monolayer, we cannot explain the observed splitting by monolayer fluctuations.

The temporal evolution of the PL intensities of lines A and B at 5 K, which are shown in Fig. 1(b), is very different. While line A exhibits a biexponential decay with time constants of 100 ps and about 1.6 ns, line B first grows with a time constant of 230 ps and then decays with a time constant of about 2.5 ns. The initial decay of line A and corresponding increase of line B can be understood by the transfer from light-hole to heavy-hole excitons. The second time constants are then indicative of the radiative recombination lifetimes of both excitons. The transfer from light- to heavy-hole excitons has been observed previously for a 30/27 SL [4]. The transfer time was determined to be 90 ps, which is in a good agreement with our result. In order to identify the type of exciton, we have investigated the temperature dependence of the PL intensities of peaks A and B. An increase of the lifetime with increasing temperature would indicate that the recombination is dominated by free excitons.

A different situation occurs for the 21/21 SL. The PL spectrum of this SL shown in

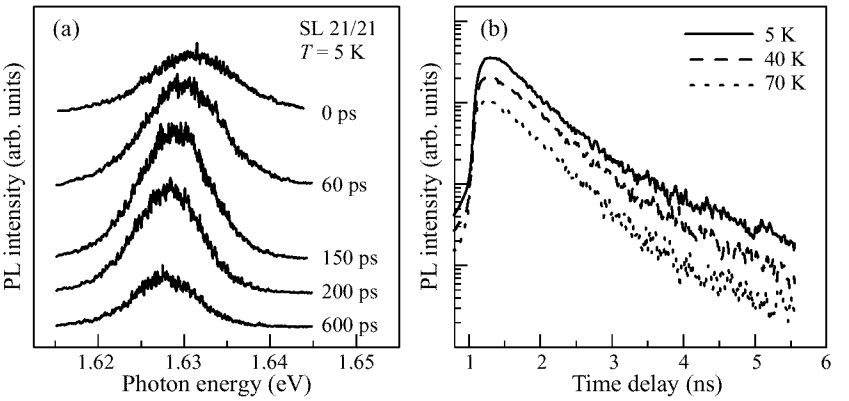

Fig. 2. (a) Time evolution of the PL spectrum for the 21/21 SL at 5 K. (b) PL intensity vs time measured at 5, 40, and 70 K.

Fig. 2(a) consists of a single peak at 1.631 eV. Within the first 150 ps, a spectral red-shift of 3 meV is observed, which is typical for exciton localization, indicating a transfer from free to localized excitons. This conclusion is in agreement with results reported by Gurioli *et al.* [5] who observed a Stokes shift of 3 meV at 5 K for a similar sample.

The PL decay of this line can be well described by a biexponential decay as shown in Fig. 2(b). The fast decay is assigned to the transfer of free to localized excitons, while the slow component is determined by the recombination of localized excitons. The decay time due to free excitons increases somewhat from 250 to 500 ps, when the temperature is increased from 5 to 80 K. At the same time, the decay time assigned to localized excitons remains almost constant (about 1.4 ns) in the low-temperature region and decreases to about 1 ns for $T > 30$ K. If the temperature is larger then 70 K, the PL decay is well described by a single exponential with a shorter time constant. This observation implies that localized excitons become delocalized above this temperature.

Let us consider now the recombination dynamics in the SL's with thinner wells (samples 14/7 and 9/4). Their PL spectra also consist of one narrow line due to heavy-hole exciton recombination. Note, that the direct bandgap in the 9/4 SL is caused by very thin barrier layers which leads to pushing up the AlAs-related X-level in the conduction band above the Γ-level of the well [6].

In contrast to the 21/21 SL, no temporal red-shift of the PL maxima is observed in the spectra of the 14/7 and 9/4 SL's at low temperatures. PL intensities decay exponentially with time constants of 300 and 200 ps, respectively. It implies that the excitons in these SL's are localized. However, starting from $T = 80$ K, a small temporal shift by 2–3 meV of the PL maximum towards lower energies takes place in the spectrum of the 14/7 SL. At the same time, the PL decay curves exhibit a biexponential behavior, indicating on progressive delocalization of excitons in this sample. On the other hand, in the 9/4 SL the PL intensity decays exponentially up to $T = 100$ K, which implies that excitons in this SL remain localized even at rather high temperatures.

3. Conclusions

Thus, if the well width is comparable with the exciton radius such as in the 36/36 SL, two separate PL lines are observed in the PL spectra, which are connected with heavy- and light-hole excitons. In SL's with thinner quantum wells such as the 21/21 SL, the excitons

are already confined by the fluctuating potential of the hetero-interfaces. However, in the time-resolved PL spectra, two lines are still observed indicating the rapid transition from free to localized excitons. In SL's with even thinner wells the influence of lateral potential fluctuations due to interface roughness completely governs exciton recombination.

Another peculiarity is a rapid decrease (from about 2.5 ns to 200 ps for the 36/36 and 9/4 SL's, respectively) of the PL decay time with the decrease of the well thickness, which could be explained by a progressive influence of the interface centers of nonradiative recombination.

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