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Localization of Excitons in AlGaAs/GaAs Quantum Wells Observed in the Time Resolved Photoluminescence Spectroscopy

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We present the detailed analyses of temporal profiles of photoluminescence from heavy-hole excitons in AlGaAs/GaAs quantum wells. We have observed the two-component exponential decay when the excitation density is low. We have confirmed by measuring the lateral spatial motion of excitons that the fast component is attributed to radiative recombinations of free excitons while the slowly decaying component is due to localized excitons. To the best of our knowledge, this is the first direct observation of localization of 2-dimensional excitons in quantum wells.

Introduction

The optical properties of quantum wells (QW's) are dominated by 2-dimensional (2D) excitons up to room temperatures due to the large binding energies. Free 2D excitons are expected to have long coherence length, leading to a rapid radiative decay rate and a large non-linear optical coefficients[1]. These properties are essential to the proposed operations of optical devices. The localized excitons caused by the potential fluctuations, as interface roughness and alloy disorder, can hamper achieving these devices. In AlGaAs/GaAs system, even with the advanced growth technique, monolayer fluctuations at interfaces seem to persist, resulting in the localization of excitons at low temperatures[2].

The quality of QW's are evaluated mainly through cw photoluminescence (PL) and photoluminescence excitation (PLE) spectra. Though these methods are indispensable for the evaluations, they are not sufficient for identifying the localized excitons and the degree of interface roughness, as demonstrated in the followings. In this report we present the detailed analyses of the PL temporal profiles of the AlGaAs/GaAs QW's and, for the first time to our knowledge, demonstrate the direct observation of localized excitons by measuring the lateral spatial motion of excitons. From these observations we clarify the difference of radiative decay rates between localized and free excitons in QW's.

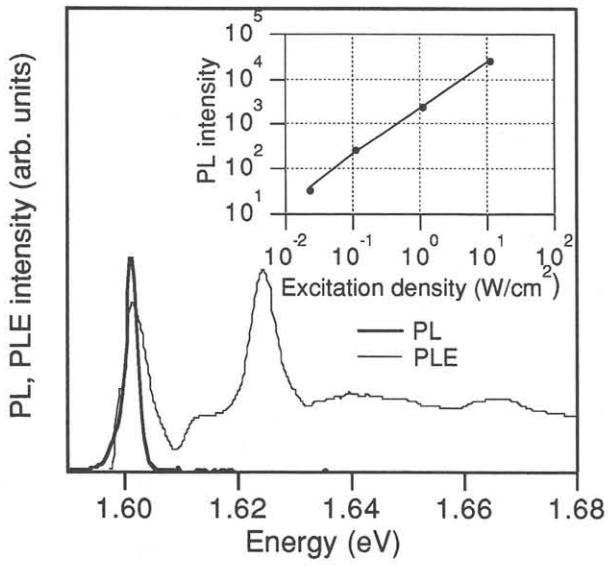

Fig. 1 PL (thick line) and PLE (thin line) of the sample at 4 K. The inset shows the PL intensity as function of excitation power density.

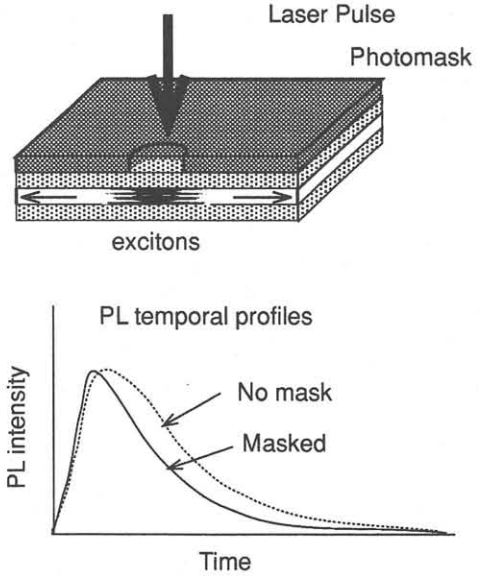

Fig. 2 Exciton time-of-flight technique to measure transport.

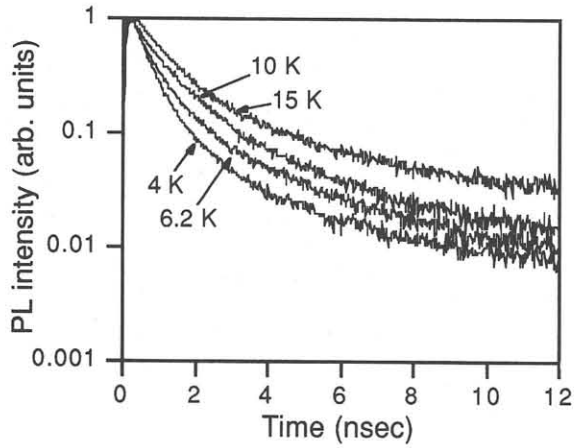

Fig. 3 Temporal profiles of PL at 4 K, 6.2 K, 10 K, and 15 K excited at 1.71 eV. Initial carrier density is estimated to be 10^8 cm^{-2}.

Experimental

The time resolved PL measurements have been performed on several samples with varying well widths. We will focus on the sample with a 5-nm QW in the following discussions. The sample of a $Al_{0.3}Ga_{0.7}As/GaAs$ QW was grown by molecular-beam epitaxy on a semi-insulating GaAs (100) substrate. It contains a single QW of 5 nm in width. The sample was cooled in a variable-temperature liquid-He cryostat. For the PL and PLE measurements we used a cw Ti:Sapphire laser pumped by an Ar laser. We measured cw PL and PLE spectra to examine the quality of the sample. Figure 1 shows the cw PL and PLE spectra at 4 K. The PL emission line is sharp and its overlap with the PLE peak is excellent free from a Stokes shift. We further tested the PL intensity as a function of excitation laser intensity. As can be seen in the inset of Fig. 1, the PL intensity is proportional to the excitation density down to 0.02 W/cm^2. Furthermore, no discernible shift of the peaks was observed when the excitation density was changed. From these observations we have concluded that the influence of bound excitons and non-radiative recombinations are negligible in our sample.

In the time resolved measurements, we used a cavity-dumped dye laser pumped by a mode-locked Ar laser. A pulse duration was 5 psec using Pyridine 2 as a laser dye. The excitation photon energy was fixed at 1.71 eV (723 nm). The laser beam was loosely focused onto the sample with a beam spot diameter of 150 μm. The laser intensity was adjusted so that the carrier density for one laser pulse was of the order of 10^8 cm^{-2}. (It is important to keep the excitation density low because the relative PL intensity of localized excitons to free excitons becomes stronger as the excitation density becomes lower.) The luminescence was dispersed through a 35-cm monochromator, detected using a cooled microchannel plate photomultiplier in a photon-counting mode. The overall time resolution was better than 70 psec.

In order to observe the lateral spatial motion of excitons, we have adopted an exciton time-of-flight technique which was discussed in detail by Hillmer et al.[3]. In this method an opaque photomask with an array of small transparent holes (10 μm in diameter, and separated from the nearest neighbor by 50 μm) was prepared. The PL temporal profiles are recorded successively with and without a photomask on the sample. With the mask placed on the sample (Fig. 2), the laser pulse generates photo-carriers only inside the hole area. After the excitation, the photo-carriers drift out of the hole area, and we detect only the PL from the excitons which remain inside the hole area. The temporal profile of the PL is determined both by the radiative decay and the escape of excitons from the hole area. By comparing the PL temporal profile with and without the photo-mask, one can obtain the information on the lateral motion of excitons in a QW. There is a possibility that an exciton travels to the neighboring hole and emits a photon, but we have confirmed this effect is negligibly small by a Monte Carlo simulation in our case.

Results and discussions

Figgure 3 shows the PL temporal profiles at 4.0, 6.2, 10, and 15 K in logarithmic scale. The detection wavelength has been fixed at the PL emission peak, and the excitation density has been of the order of 10^8 cm^{-2}. We have accumulated the data long enough to ensure high statistical precision in the whole time range of our interest. The decay curve of PL is not described by a single exponential curve as expected for the radiative recombination of free excitons, but requires two-component exponentials. The two-component exponential decay has been observed in other samples of different well width especially in low excitation conditions. And the relative strength of fast and slow components is sample-dependent. As shown in Fig. 3, the decay rates of the fast component are temperature dependent, while those of the slow component are the same at four temperatures within statistical uncertainties. The least squares fitting to the double exponential function shows that the decay time of the fast component increases from 0.47 nsec at 4.0 K to 0.94 nsec at 15 K almost linearly.

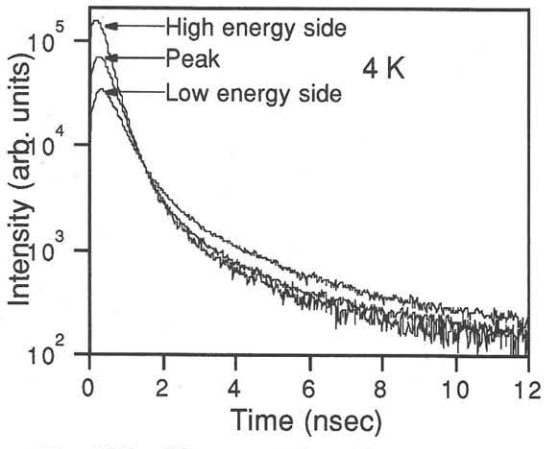

Fig. 4 The PL temporal profiles at various detection energies. The detection energies are 1.604 eV (PL peak) ± 0.002 eV.

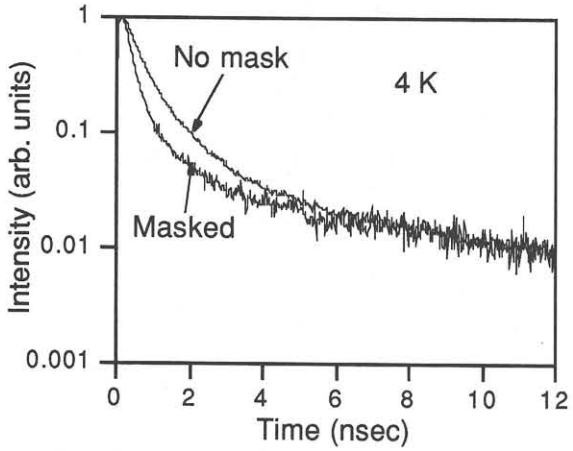

Fig. 5 The PL temporal profiles from the same sample with and without a photomask at 4 K. The excitation conditions are the same for both data.

On the other hand the slow component is independent of temperatures with a decay constant of 2.2 nsec. The linear dependence of decay time on the temperature is characteristic of free excitons, while localized excitons are expected to show temperature independent decay time, as reported both for AlGaAs/GaAs QW's[4] and for InGaAs/GaAs QW's[5]. The relative intensity of the slow component reduces considerably when the excitation density has been increased to 10^9 cm^{-2}. From these observations on the time resolved PL measurements it is likely that the fast component should be attributed to the radiative recombination of free excitons while the slow component is due to the localized excitons in the monolayer fluctuations.

We have further measured the PL temporal profiles at different detection energies. Plotted in Fig. 4 are the PL temporal profiles for several detection energies, at PL peak (1.604 eV) and 2 meV above and below the peak at 4 K. As the detection energy changes, relative strength of the fast and slow components varies; the slow component becomes stronger as the detection energy goes lower. This observation is consistent with the assertion of the ref. 2 that the e-hh exciton state is inhomogeneously broadened due to the monolayer fluctuations, and the localized states appear on the lower energy side.

We have proceeded to measure spatial motion of excitons to confirm the localization effect. We present in Fig. 5 the observed time evolution of emission from the bare (no photomask) and the masked sample at 4 K under the same excitation conditions. The data show that the decay time of the fast component becomes shorter when the sample is covered with a mask. The reduction of decay time is explained by the spatial motion of free excitons, that is, free excitons escape into the opaque region before they decay radiatively. Actually, assuming diffusive motion of excitons, we can deduce a diffusion constant by comparing temporal profiles of the masked and unmasked data[6]. We have obtained 50 cm^2/sec in this case. On the other hand, as for the slow component, the decay curves in both cases are the same within the statistical uncertainties, showing that the excitons are immobile in this case. Thus we have concluded that the fast component is attributed to the radiative recombination of free excitons, and the slow component is from localized excitons.

The difference of decay rates between free excitons and localized excitons can be attributed to the coherent nature of 2D excitons in QW's. In the ideal 2D crystal at low temperatures, a free exciton is a coherent excitation of the whole crystal, leading to a large dipole transition probability and fast radiative decay rate since the transition probability is proportional to the area of the coherent excitation as discussed in the ref. 1 and ref. 4. In the real crystal, however, an exciton undergoes the scattering with interface roughness and impurities, resulting in the localization of excitonic states at low temperatures. The coherent area of a localized exciton is determined by the localization length which is smaller than that of a free exciton (the spatial extent of the localized state is always smaller than that of the extended state). Thus the radiative decay time of localized excitons is much longer than that of free excitons.

The foregoing discussion indicates that the time resolved PL measurement alone (without measuring spatial motion) can be a very sensitive way to detect localization effect. Even when the PL and PLE measurements do not show the evidence of localized excitons, the time resolved data can clearly show their existence thanks to the difference of decay rates for free and localized excitons.

Conclusions

We have observed the two-component exponential decay of PL from the AlGaAs/GaAs QW in our high precision time resolved data. We have concluded by measuring the spatial motion of excitons that the fast component is attributed to the radiative recombination of free excitons while the slow component is due to the localized excitons. The difference of the decay rates can be attributed to the coherence length of wave functions for free excitons (extended states) and localized excitons.

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