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# Photoluminescence dynamics of weakly confined excitons in GaAs thin films

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**Abstract**

We investigate the dynamics of weakly confined excitons in GaAs thin films measured by time-resolved photoluminescence (PL) technique. When excitation energy was above the resonant energy of the exciton, a long PL rise time of about 200 ps was observed. It is considered that an exciton formation process from excited continuum energy states to discrete energy states of the exciton in the thin film causes the slow PL rise. The observed PL decay time constant was about 14 ns due to high quality fabricated samples. The observed population dynamics can be surely ascribed to the specific features of weakly confined excitons.


*Keywords:* Weakly confined exciton; Time-resolved photoluminescence; Exciton formation

Nowadays, the optical property of an exciton in quantum structures has been one of intense researches for ultrafast optical devices. When the center-of-mass motion of exciton is confined in a thin film with a particular thickness, namely the exciton Bohr radius is smaller than the thickness of the film, an ultrafast and large nonlinear optical response was theoretically reported under the nonlocality-induced double resonance in energy and size (NIDORES) condition [1]. In case of picosecond (ps)-pulse laser irradiation, large and ultrafast optical responses of an exciton were measured by a transient grating technique [2,3]. On the other hand, in case of femtosecond-pulse laser irradiation, the optical response was reported to have both the ultrafast response and the long decay component measured by the transient grating [4]. Since the origin of the two components is unclear, detailed study of population dynamics is needed. In this study, we investigate exciton population dynamics measured by a time-resolved photoluminescence (PL) technique.

The used samples were grown by molecular beam epitaxy on (0 0 1) GaAs wafer. The samples consist of three double-hetero-structure layers, which were made of 110-nm GaAs active layer sandwiched by 5.7-nm $Al_{0.3}Ga_{0.7}As$ layers. Excitons in the active layer were weakly confined because its Bohr radius of about 12 nm was smaller than the thickness of films. Time-resolved PL measurements were performed using a streak camera with a single monochromator. An excitation laser was a mode-locked Ti:sapphire laser. The duration and the repetition rate were 100 fs and 80 MHz, respectively. The laser linewidth was varied by a spectral filter unit using a slit and grating mirror. The variation range was from 1 to 25 meV, which corresponded to several ps to 100 fs pulse. The sample was set in cryostat and was kept at 4 K. All measurements were performed under a normal incident condition and a cross-Nicol configuration. An average power of the excitation laser was about 1.0 $W/cm^2$, in order to avoid a many-body effect of photo-generated excitons.

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Fig. 1 shows the observed photo-reflection and PL spectra. The reflection spectrum clearly shows the peak energy due to the energy states of the weakly confined exciton. These energies agree with the results estimated with the weakly confined exciton model. The PL spectrum seems to be the corresponding peaks to those of the reflection spectrum. The PL peak with large intensity at 1.514 eV is originated from the relaxation of the second excited energy level of the exciton, which indicates that the weakly confined excitons exist certainly in the thin films.

Fig. 2 shows the trace of time-resolved PL result with a ps-pulse laser under the resonant excitation condition. The PL trace showed a long decay constant of several nanoseconds. Note that the fast decay component consists of not only the PL component but also the laser signal. The PL decay measured with slow repetition rate laser is shown in the inset in Fig. 2. The observed decay time was estimated to be 14 ns. The faster decay constant about 2 ns near the origin can be affected to the system resolution, and long decay component is due to the lifetime of the exciton. The long lifetime is considered to originate from the high quality of fabricated GaAs thin films, because impurity states were hardly observed in the PL spectrum.

As the excitation energy was increased, slow PL rises were observed under ps-laser irradiation (Fig. 3). The PL spectra and traces of bulk structure are also shown. The slow PL rises were observed in the PL trace for the excitation energy above 1.525 eV. The rise is rarely observed in the bulk structure. Therefore, the slow PL rise is specific property of the weakly confined excitons.

For study of origin of slow PL rise, the measurements at various temperatures under nonresonant excitation

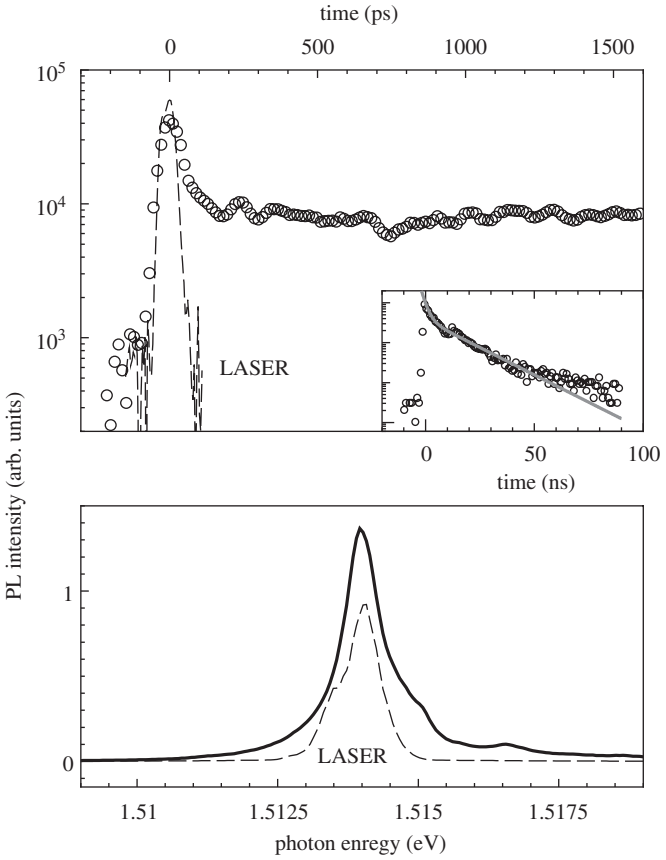


Fig. 2. (Top) Time-resolved PL trace under ps-pulse laser irradiation and (bottom) corresponding PL spectrum (thick solid line) with ps-laser spectrum (dashed line). (Top inset) Time-resolved PL trace measured with 4 MHz repetition rate laser at 10 K.

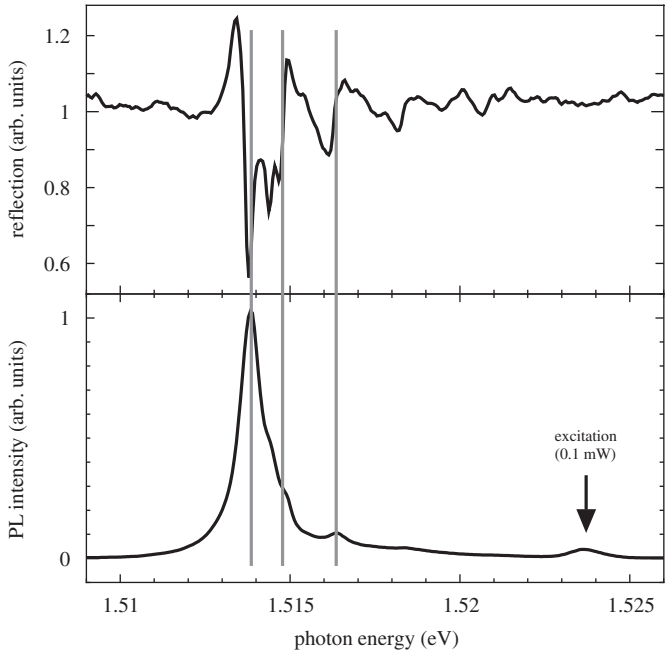


Fig. 1. (Top) Reflection spectrum measured under the Brewster angle incident condition and (bottom) PL spectrum at 4 K. The solid vertical lines indicate the confined exciton energy levels.

conditions with the ps-laser irradiation were performed. Temperature dependence of the rises is shown in Fig. 4. Rise time constant is estimated to be about 200 ps at 5 K. The slow PL rise can be caused by an exciton formation process, because the behavior of the observed profiles is fairy similar to the reported one [5]. When the excitation energy is larger than 1.524 eV, the continuum state is excited. Then, the photo-generated electron–hole pairs relax from continuum states to discrete states of the weakly confined exciton with phonon emission. The slow PL rise was reported in quantum well structure, and the reported rise time constant was comparable with the observed one in our sample. Therefore, the exciton formation due to the electron–hole relaxation, from the excited continuum states of the thin film to the discrete energy levels of exciton, causes the observed slow PL rise.

We observed a slow PL rise time constant of about 200 ps, when the excitation energy was much higher than the resonant energy of the exciton states. The threshold energy seemed to be a little higher energy of the continuum state of a thin film. The slow PL rise is caused by the exciton formation process from the excited continuum states of the thin film to the discrete energy states of the

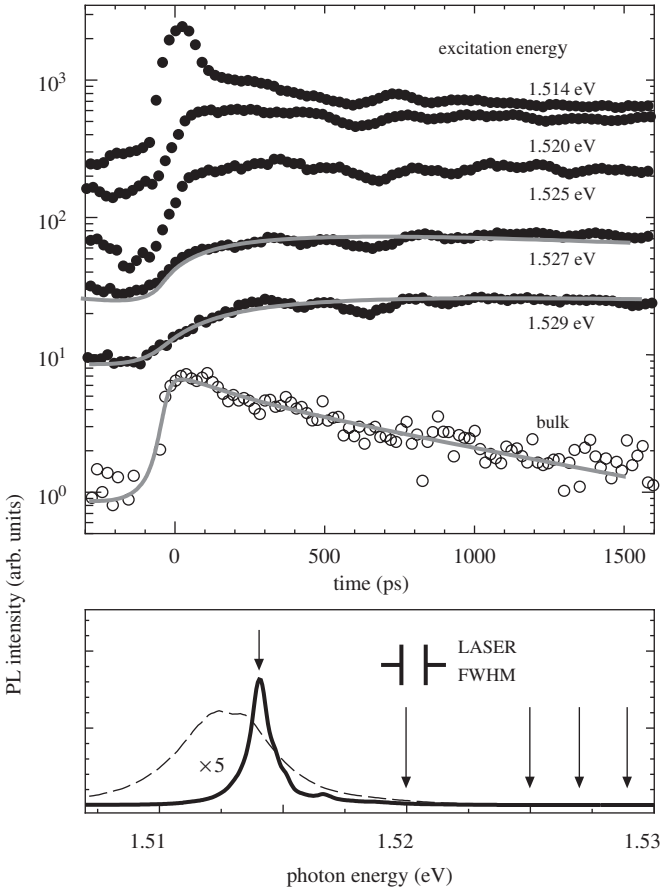


Fig. 3. (Top) Time-resolved PL traces at various excitation energies and (bottom) corresponding PL spectrum. The arrows indicate the excitation energies. The solid circles and solid line show PL signals of weakly confined excitons. The circles and dashed line show those of bulk structure. Solid gray lines are guidelines for eyes. The excitation energy for bulk structure was 1.532 eV.

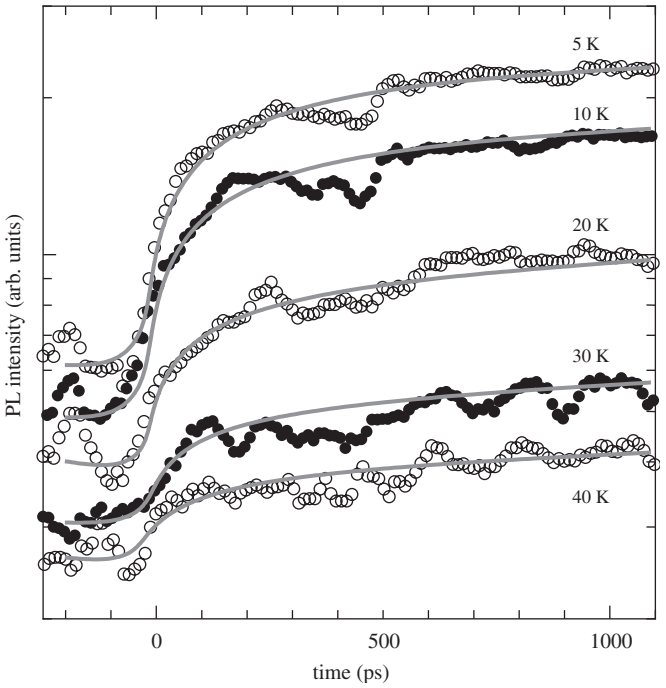


Fig. 4. Temperature dependence of time-resolved PL. Solid gray lines are guidelines for eyes.

weakly confined exciton. In addition, long PL decay time constant of 14 ns under resonant excitation conditions was measured. One of the origins of a long PL decay time of weakly confined excitons is ascribed to the high quality fabricated samples.

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