Time-resolved luminescence of InP quantum dots in a $Ga_{0.5}In_{0.5}P$ matrix: Carrier injection from the matrix

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We conducted time-resolved luminescence spectroscopy on self-assembled InP dots in a $Ga_{0.5}In_{0.5}P$ matrix at different photon excitation energies. In comparison with results on $Ga_{0.5}In_{0.5}P$ without dots, the influence of the $Ga_{0.5}In_{0.5}P$ matrix on luminescence-decay profiles of InP dots was clarified. By excitation at the $Ga_{0.5}In_{0.5}P$ matrix, the long-decay component ($\sim$50 ns), as well as the 400 ps radiative-decay component, was observed in luminescence of InP dots. This long-decay component reflects carrier lifetime in the $Ga_{0.5}In_{0.5}P$ matrix. Carrier injection time from the $Ga_{0.5}In_{0.5}P$ matrix to InP dots is estimated to be $\sim$100 ps, and is determined by carrier diffusion in the matrix. [S0163-1829(98)00903-5]

Nanostructures showing three-dimensional size confinement have attracted increasing attention due to their unique physical properties and potential device applications. Direct growth of self-assembled dot systems using molecular-beam epitaxy or metal organic vapor-phase epitaxy is promising, since it enables small, good-quality quantum dots to be fabricated.[1–6] Room-temperature lasing operation in a self-assembled dot system was reported recently.[7,8] In such applications, carriers are injected into a matrix and transported to dots. In many basic studies of optically excited self-assembled dot systems, the matrix rather than dots was photoexcited. This is because dots are distributed in a thin layer, making the number of directly excited carriers in dots very small. Research on carrier dynamics from the matrix to dots and on the effect of the matrix on dot luminecence properties, which remains relatively limited, is quite important.

Here we report time-resolved luminescence spectroscopy of InP dots in a $Ga_{0.5}In_{0.5}P$ ($Ga_{0.5}In_{0.5}P$ lattice matched to GaAs) matrix under excitation at different wavelengths. By comparing results on $Ga_{0.5}In_{0.5}P$ without dots, carrier dynamics from the matrix to dots is discussed.

Of the two samples used, one was $Ga_{0.5}In_{0.5}P$ without dots and the other was self-assembled InP dots embedded in a $Ga_{0.5}In_{0.5}P$ matrix (InP/$Ga_{0.5}In_{0.5}P$). Note that the term ''matrix'' refers to the $Ga_{0.5}In_{0.5}P$ matrix in InP/$Ga_{0.5}In_{0.5}P$, not to the $Ga_{0.5}In_{0.5}P$ sample without dots. The $Ga_{0.5}In_{0.5}P$ sample without dots was 500 nm thick and deposited on a 300 nm GaAs buffer layer grown on a GaAs(100) substrate using gas-source molecular-beam epitaxy. The InP/$Ga_{0.5}In_{0.5}P$ sample nominally has 4 ML InP sandwiched by the 300 nm $Ga_{0.5}In_{0.5}P$ layers grown on GaAs. The typical diameter of InP self-assembled islands is 70 nm, their typical height 7 nm, and their density $\sim 10^{10}$ cm^{-2}, as are measured by atomic force microscopy.

The excitation source was $\sim$300 fs pulses of 1.85–2.03 eV photon energy from an optical parametric amplifier whose source was a titanium-sapphire laser regenerative amplifier. The laser spectral width was $\sim$30 meV and the repetition rate 200 kHz. Laser light was focused onto a spot $\sim$200 μm in diameter. Samples were directly immersed in superfluid helium at 2 K. Sample photoluminescence was spectrally resolved using a 0.6 m monochromator and detected using a charge-coupled device camera. Luminescence spectra were corrected by the wavelength-dependent sensitivity of the apparatus. Temporal changes of luminescence were obtained by a single-photon counting technique using a microchannel plate photomultiplier (time range 100 ns), or by using a syncroscan streak camera (1000 ps).

Figure 1(a) shows luminescence spectra of $Ga_{0.5}In_{0.5}P$ (above) and InP/$Ga_{0.5}In_{0.5}P$ (below). The excitation photon energy was 2.03 eV and the density 0.1 $\mu J/cm^2$. In the $Ga_{0.5}In_{0.5}P$ sample, luminescence was located at 1.952 eV. Luminescence at about 1.75 eV also appeared in the InP/$Ga_{0.5}In_{0.5}P$ sample, and is attributed to InP dots. No lu-

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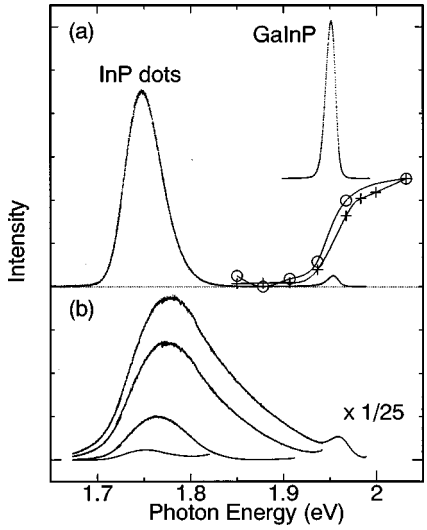

FIG. 1. (a) Luminescence spectra of $Ga_{0.5}In_{0.5}P$ (above) and $InP/Ga_{0.5}In_{0.5}P$ (below) at 2 K. The excitation photon energy is 2.03 eV and the density 0.1 $\mu J/cm^2$. Crosses show the excitation spectrum of InP dot luminescence. Circles show contribution of the long decay component to decay profiles of InP dot luminescence. (b) Luminescence spectra of $InP/Ga_{0.5}In_{0.5}P$ at 2 K under excitation photon energies of 2.03, 1.97, 1.94, and 1.85 eV (top to bottom). Excitation densities are fixed at 10 $\mu J/cm^2$.

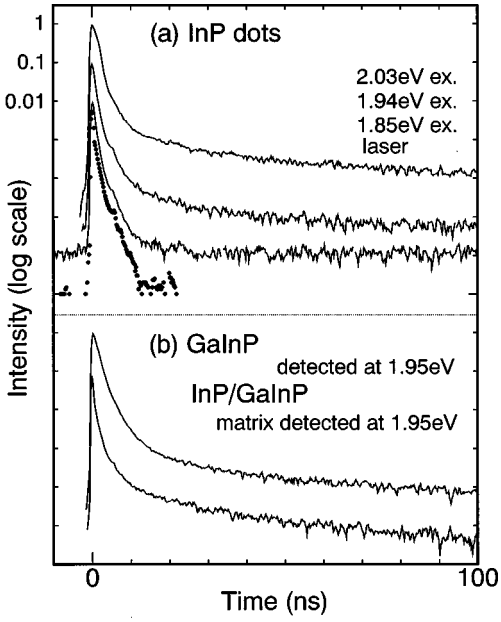

FIG. 2. (a) Temporal changes of the luminescence of InP dots detected at 1.75 eV (detection width 10 meV) under excitation photon energies of 2.03, 1.94, and 1.85 eV (top to bottom). The dotted line is the measured laser profile, indicating system response. (b) Temporal changes of $Ga_{0.5}In_{0.5}P$ luminescence at 1.95 eV (detection width 2 meV) from the $Ga_{0.5}In_{0.5}P$ sample without dots (above) and the $Ga_{0.5}In_{0.5}P$ matrix including InP dots (below). The excitation photon energy is 2.03 eV. In (a) and (b), excitation densities are fixed at 10 $\mu J/cm^2$, and the temperature is 2 K. Results are plotted on a log scale, and displaced vertically for clarity.

minescence was observed from a two-dimensional wetting layer, and this $InP/Ga_{0.5}In_{0.5}P$ sample is therefore assumed to not have such a layer.

Figure 1(b) shows luminescence spectra of the $InP/Ga_{0.5}In_{0.5}P$ sample excited at 2.03, 1.97, 1.94, and 1.85 eV for an excitation density of 10 $\mu J/cm^2$. This density is 100 times larger than that used in Fig. 1(a). The top spectrum in Fig. 1(b) is broader than the dot luminescence spectrum in Fig. 1(a), and the peak energy shows a blueshift. This indicates state filling in dots.[9] In Fig. 1(b), the luminescence intensity decreases with excitation photon energy. The excitation spectrum of the dots is thus obtained as plotted by crosses in Fig. 1(a). Intensity decreases rapidly at about 1.952 eV, indicating that carriers luminescing from dots are created mainly in the $Ga_{0.5}In_{0.5}P$ matrix. When the matrix was excited by photon energy exceeding 1.952 eV, the dot luminescence intensity was $\sim$30 times larger than that obtained by exciting only dots with a photon energy less than 1.952 eV.

Figure 2(a) shows temporal changes of the luminescence of the InP dots measured at 1.75 eV, that is, luminescence peak of InP dots in Fig. 1(a). The excitation photon energies were 2.03, 1.94, and 1.85 eV, and the excitation density was 10 $\mu J/cm^2$. A dotted line in the figure is a measured laser profile showing the system response. All three profiles show a fast decay component resembling the laser profile.[10] When higher dot levels were excited directly by photon energies from 1.85 to 1.91 eV, no slow luminescence rise was observed. This means the absence of a ''phonon bottleneck.''[11]

In addition to the fast decay component, a longer decay component with a $\sim$50 ns decay time was observed in the top profile in Fig. 2(a). This longer component was observed even at a lower excitation density of 0.1 $\mu J/cm^2$. In Fig. 2(a), the long decay component decreases with excitation energy. The bottom profile has no longer component, and can be fitted well to the convolution of the 400 ps single exponential decay and the system response over three orders of luminescence magnitude. This decay time (400 ± 50) ps agrees with reported data,[12,13] and is attributed to the radiative lifetime of InP dots. By subtracting the bottom profile taken for excitation at 1.85 eV from other decay profiles, we estimated the contribution of the longer decay component in all profiles. Circles in Fig. 1(a) show the time-integrated intensity of the long decay component plotted against the excitation photon energy. This shows that the long component decreases rapidly with excitation energy crossing $Ga_{0.5}In_{0.5}P$ energy of 1.952 eV. This result suggests that the long component is attributable to the $Ga_{0.5}In_{0.5}P$ matrix.

Figure 2(b) shows temporal changes of $Ga_{0.5}In_{0.5}P$ luminescence at 1.952 eV from the $Ga_{0.5}In_{0.5}P$ sample without InP dots (above) and the $Ga_{0.5}In_{0.5}P$ matrix in the $InP/Ga_{0.5}In_{0.5}P$ sample (below). The excitation photon energy was 2.03 eV and the density 10 $\mu J/cm^2$. Both profiles are similar, having a longer decay component, and resemble that of dot luminescence obtained using 2.03 eV excitation shown as the top profile in Fig. 2(a). Therefore, it is concluded that the long decay component in dot luminescence stems from the $Ga_{0.5}In_{0.5}P$ matrix. When the $InP/Ga_{0.5}In_{0.5}P$ sample is excited by photon energy exceeding 1.952 eV, carriers are produced in the $Ga_{0.5}In_{0.5}P$ matrix and transported to InP dots. Thus the decay time of the InP dot luminescence is governed by the decay time of the matrix luminescence, that is, by carrier lifetime in the $Ga_{0.5}In_{0.5}P$ matrix.

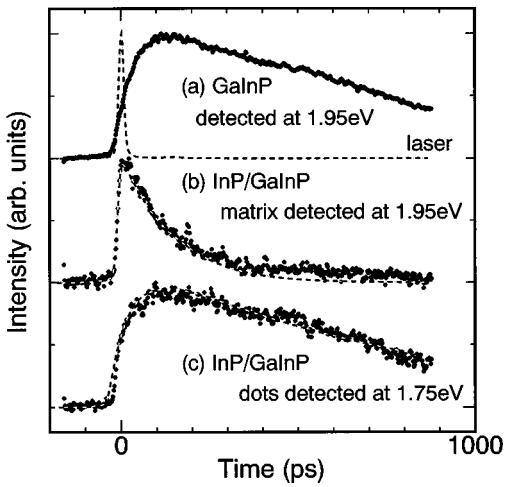

FIG. 3. Expanded temporal changes (up to 1000 ps time region) of $Ga_{0.5}In_{0.5}P$ luminescence at 1.95 eV (detection width 6 meV) from (a) the $Ga_{0.5}In_{0.5}P$ sample without dots and (b) the $Ga_{0.5}In_{0.5}P$ matrix including InP dots, and of (c) the InP dot luminescence detected at 1.75 eV (detection width 6 meV). The excitation photon energy is 2.03 eV, the density is 3 μJ/cm^2, and the temperature is 2 K. Results are plotted on a linear scale. The dashed line in (a) shows the measured laser profile, indicating the system response. The line in (b) [or (c)] is a fit of the convolution between function $\exp(-t/\tau_d)$ [or $\exp(-t/\tau_d)\{1-\exp(-t/\tau_r)\}$] and the laser profile.

The long decay component can be explained by localized electrons and/or holes in tail states below conduction and/or valence-band edges in $Ga_{0.5}In_{0.5}P$. One possible origin of these tail states is defects or alloy fluctuations in $Ga_{0.5}In_{0.5}P$. Another is $Ga_{0.5}In_{0.5}P$ ordering. Certain growth conditions are known to lead to a spontaneous long-range ordering of the usually disordered Ga-In sublattice of the III-V zinc-blende structure.[14] In such ordered $Ga_{0.5}In_{0.5}P$, long photoluminescence decay components are observed.[15] In addition, ordered $Ga_{0.5}In_{0.5}P$ is reported to show lower luminescence energy than disordered $Ga_{0.5}In_{0.5}P$,[14] and polarization-dependent photoluminescence.[16] These two phenomena were observed also in our samples. Thus, the 50 ns long decay component shown in Fig. 2 may come from ordered $Ga_{0.5}In_{0.5}P$, although further study may be needed to confirm this. At the very least, it is clarified that InP dot luminescence shows the long decay component reflecting the carrier lifetime in the $Ga_{0.5}In_{0.5}P$ matrix.

Although the 50 ns decay component exists similarly, decay profiles less than 10 ns region differ in Fig. 2(b). Dots in Fig. 3 show expanded temporal changes of the $Ga_{0.5}In_{0.5}P$ luminescence at 1.95 eV in (a) the $Ga_{0.5}In_{0.5}P$ sample without InP dots and (b) the InP/$Ga_{0.5}In_{0.5}P$ sample. In Fig. 3(c), the temporal change of InP dot luminescence at 1.75 eV is shown. The excitation photon energy was 2.03 eV and the density 3 μJ/cm^2.

The dashed line in Fig. 3(a) is a measured temporal change of the laser. The time resolution is $\sim$20 ps. The luminescence decay profile of Fig. 3(a) shows slow luminescence rise. This may be attributable to state filling in low-energy tail states and carrier relaxation from higher-energy states of $Ga_{0.5}In_{0.5}P$. It is because fast luminescence rise less than the time resolution and fast decay less than 100 ps were observed at a higher luminescence energy of 1.96 eV.

Although the decay profile of Fig. 3(a) shows a slow luminescence rise, the decay profile of Fig. 3(b) shows a fast rise less than the time resolution and a faster decay than that of Fig. 3(a). The line in Fig. 3(b) is a fit of the convolution between the laser profile and a single exponential decay with 130 ps decay time. The difference in decay profiles of Figs. 3(a) and (b) can be attributable to InP dot introduction to $Ga_{0.5}In_{0.5}P$. In quantum-well systems, carrier capture time was studied.[17,18] While quantum capture or carrier thermalization dominates the capture time for samples with thin barriers, carrier transport to wells dominates the time for samples with barriers thicker than 50 nm.[17] Because the width of the $Ga_{0.5}In_{0.5}P$ matrix in the InP/$Ga_{0.5}In_{0.5}P$ sample used in Fig. 3(b) is 300 nm, the difference between decay profiles of Figs. 3(a) and (b) is attributed to the carrier transport from the $Ga_{0.5}In_{0.5}P$ matrix to InP dots. Carriers are injected into InP dots at the time constant of the decay time in Fig. 3(b). The rise times differ in Figs. 3(a) and (b), because state filling does not occur in the $Ga_{0.5}In_{0.5}P$ matrix in the InP/$Ga_{0.5}In_{0.5}P$ sample. This in turn is due to the low-energy tail states of $Ga_{0.5}In_{0.5}P$ not occupied long enough by the carrier for the escape to dots.

Carrier injection from the matrix to dots is confirmed by the slow luminescence rise of InP dots shown in Fig. 3(c). The line in Fig. 3(c) is convolution fitted between the laser profile and function $\exp(-t/\tau_d)\{1-\exp(-t/\tau_r)\}$, where the decay time $\tau_d = 800$ ps and the rise time $\tau_r = 50$ ps.

The 130 ps decay time in Fig. 3(b) is expected to coincide with the 50 ps rise time in Fig. 3(c). This difference is probably induced by intense laser excitation of 3 μJ/cm^2. When excitation density was 10 μJ/cm^2, the luminescence decay time of the $Ga_{0.5}In_{0.5}P$ matrix was 150 ps and the rise time of InP dots was 30 ps. The difference became larger, probably due to state filling of low-energy tail states of $Ga_{0.5}In_{0.5}P$. Intense excitation causes filling of higher $Ga_{0.5}In_{0.5}P$ states and, in turn, carrier relaxation to lower $Ga_{0.5}In_{0.5}P$ states. This effect observed in the $Ga_{0.5}In_{0.5}P$ sample without dots in Fig. 3(a) prolongs the decay time of $Ga_{0.5}In_{0.5}P$ luminescence in the InP/$Ga_{0.5}In_{0.5}P$ sample. Carrier injection directly from higher $Ga_{0.5}In_{0.5}P$ states to InP dots shortens the rise time of the dot luminescence. In the limit of weak excitation, the luminescence decay time of the $Ga_{0.5}In_{0.5}P$ matrix and the rise time of InP dots would agree. Given these considerations, the carrier injection time from the $Ga_{0.5}In_{0.5}P$ matrix to InP dots is estimated to be on the order of 100 ps.[19]

We analyzed decay curves in Fig. 3 using phenomenological functions to obtain the carrier injection time.[18] Although carrier dynamics in the 300-nm-thick $Ga_{0.5}In_{0.5}P$ matrix sandwiching InP dots could be described using a one-dimensional diffusion equation,[20] a simple estimation should be sufficient.[17] When we assume that carrier diffusion length at 100 ps is 300 nm in the InP/$Ga_{0.5}In_{0.5}P$ sample, the diffusion coefficient at 2 K is 9 cm^2/s. This may be a reasonable value, since the coefficient of $Ga_{0.5}In_{0.5}P$ at room temperature is reported to be 6.9 cm^2/s.[21] The diffusion length at 25 K in the lateral direction was estimated to be 1 μm by cathodoluminescence in the sparse InP dot sample in $Ga_{0.5}In_{0.5}P$.[9] Using the above diffusion coefficient of 9 cm^2/s, we calculate the carrier lifetime in $Ga_{0.5}In_{0.5}P$ to be

900 ps. This is about the same as the dominant luminescence decay time of the $Ga_{0.5}In_{0.5}P$ sample shown in Fig. 2(b) or Fig. 3(a).

In summary, we have conducted time-resolved luminescence spectroscopy on self-assembled InP dots in a $Ga_{0.5}In_{0.5}P$ matrix at different photon energy excitations. These results were compared to those for $Ga_{0.5}In_{0.5}P$ without dots grown under the same condition. Long decay component ($\sim$50 ns) was observed for InP dot luminescence decay profiles, reflecting carrier lifetime in the $Ga_{0.5}In_{0.5}P$ matrix. The carrier injection time from the $Ga_{0.5}In_{0.5}P$ matrix to InP dots was estimated to be $\sim$100 ps. The carrier diffusion in the matrix was found to determine this injection time.

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