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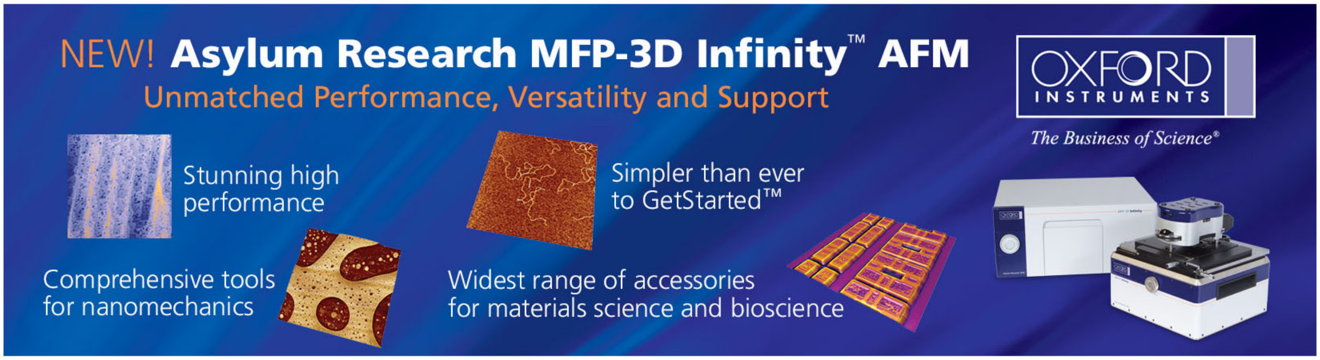
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Carrier dynamics of type-II InAs/GaAs quantum dots covered by a thin $\mathrm{GaAs}_{1-x}\mathrm{Sb}_x$ layer

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Carrier dynamics of InAs/GaAs quantum dots (QDs) covered by a thin $\mathrm{GaAs}_{1-x}\mathrm{Sb}_x$ layer were investigated by time-resolved photoluminescence (PL). Both the power dependence of PL peak shift and the long decay time constants confirm the type-II band alignment at the GaAsSb–InAs interface. Different recombination paths have been clarified by temperature dependent measurements. At lower temperatures, the long-range recombination between the QD electrons and the holes trapped by localized states in the GaAsSb layer is important, resulting in a non-single-exponential decay. At higher temperatures, optical transitions are dominated by the short-range recombination with the holes confined to the band-bending region surrounding the QDs. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2964191]

Self-assembled InAs/GaAs quantum dots (QDs) are important active materials for many optoelectronic device applications,[1] particularly for long wavelength GaAs based QD lasers.[2] In order to extend the emission wavelength into the 1.3–1.6 μm range for telecommunication applications, InAs/GaAs QDs were usually covered by an In(Al,Ga)As strain-reducing layer.[3] Recently, InAs/GaAs QDs covered by a thin $\mathrm{GaAs}_{1-x}\mathrm{Sb}_x$ layer have attracted much attention.[4–6] Emission wavelength beyond 1.5 μm and room-temperature continuous-wave operation of a 1.3 μm QD laser have been demonstrated.[7]

The $\mathrm{GaAs}_{1-x}\mathrm{Sb}_x$ covered InAs QDs are believed to exhibit a type-II band alignment when the Sb composition reaches $x \gtrsim 14\%$.[5,6] Experimental evidences for the type-II transition have been reported based on power dependent photoluminescence (PL) measurements.[5,6] However, no time-resolved PL (TRPL) has been reported, which would provide a more direct evidence on the nature of radiative recombination in such type-II QDs. Here, we report a TRPL study on the GaAsSb covered InAs QDs. The much longer decay time confirms a type-II band lineup at the GaAsSb–InAs interface. Different recombination paths were discussed and identified by temperature dependent measurements.

The samples were grown on GaAs substrates by molecular beam epitaxy. After the growth of a 200 nm thick GaAs buffer layer, an InAs QD layer [(2.7 ML) (monolayer)] was grown at 500 °C and subsequently capped by a 4.5 nm $\mathrm{GaAs}_{1-x}\mathrm{Sb}_x$ layer. Two samples with nominal x of 16% and 21% were prepared. A sample with GaAs covered InAs QDs ($x=0\%$) was also grown as a reference of type-I QDs. All samples were finally capped by a 50 nm GaAs layer. Atomic force microscopy of uncapped samples reveals that the QDs are lens shaped, with an average height of $\approx 8 \pm 0.5$ nm, a diameter of ≈ 20 nm, and a density of $3 \times 10^{10}\ \mathrm{cm}^{-2}$. PL was excited by an argon ion laser (488 nm), analyzed by a 0.5 m monochromator and detected by an InGaAs photomultiplier tube. TRPL was performed using either a 200 fs Ti:sapphire laser (780 nm/80 MHz) or a 50 ps pulsed laser diode (405 nm/5 MHz). The decay traces were recorded using the time-correlated single photon counting technique with an overall time resolution of ~150 ps.

Figure 1(a) shows the PL spectra for the QD samples with different Sb compositions. A redshift in peak energy was observed with the increasing x. The power dependent PL spectra for the 16% sample are displayed in Fig. 1(b). Increasing the excitation power (P_{ex}) results in a blueshift in QD emission peaks. The energy shift shows a linear dependence on $P_{\mathrm{ex}}^{1/3}$ [see Fig. 1(c)], consistent with the behavior expected for a type-II band alignment.[6,8] The PL peak can thus be identified as the recombination of ground-state (GS) electrons in the QDs with holes in the GaAsSb layer confined by the field induced band bending surrounding the QDs. The QD excited state (ES) appeared under higher P_{ex}'s. A similar power dependence of the blueshift was also observed for the QD ES.

Figure 2 shows the TRPL of the investigated samples measured at $T=12$ K. The decay time for the reference

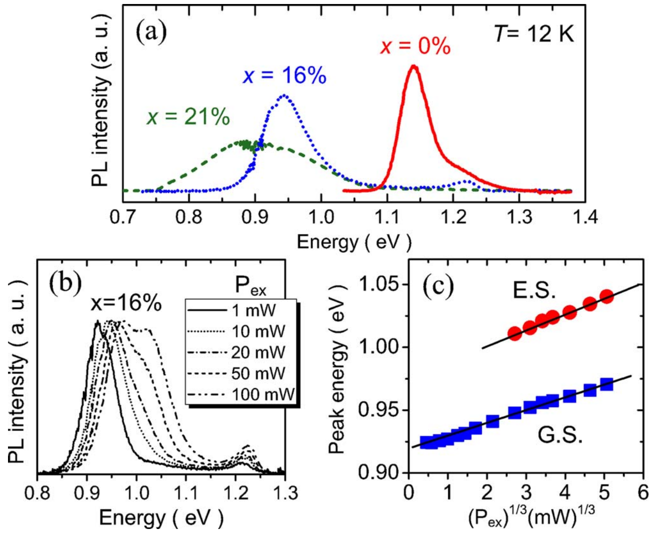

FIG. 1. (Color online) (a) PL spectra of the $\mathrm{GaAs}_{1-x}\mathrm{Sb}_x$ covered InAs QDs with different x. (b) Power dependent PL spectra of the 16% sample. (c) The GS and the ES peak energies as a function of $P_{\mathrm{ex}}^{1/3}$ for the 16% sample.

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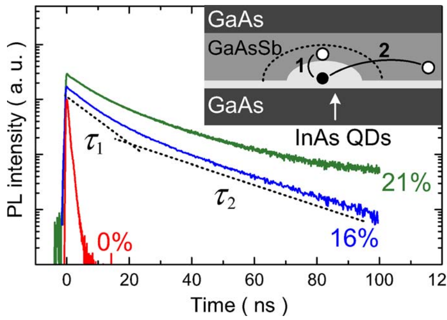

FIG. 2. (Color online) TRPL decay traces for the investigated QD samples. The inset depicts the sample structure and the underlying recombination processes responsible for the faster (1) and slower (2) decay components.

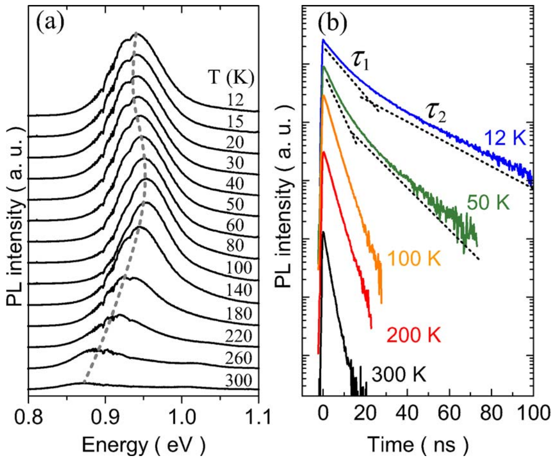

FIG. 3. (Color online) Temperature evolution of the PL spectra (a) and decay traces (b) for the $x=16\%$ sample.

InAs/GaAs QDs was $\tau \approx 0.8 \pm 0.2$ ns, comparable with the reported typical value of ~1 ns.[9] In contrast, the GaAsSb covered QDs exhibit much longer decay times than the reference InAs QDs. This can be attributed to the reduced spatial overlap between the electron and hole wavefunctions due to the type-II band alignment.[10] We notice that the decay traces for the GaAsSb covered QDs are non-single-exponential, which can be decomposed into a faster initial component and a slower tail component. In order to obtain decay time constants, they are fitted by a double-exponential function: $I(t)=A_1 \exp(-t/\tau_1)+A_2 \exp(-t/\tau_2)$. For the 16% sample, the deduced time constants are $\tau_1 = 7.5$ ns and $\tau_2=24$ ns, with a relative ratio of $A_2/A_1=0.4$. The time constants for the 21% sample are somewhat longer ($\tau_1 \approx 8.2$ ns, and $\tau_2 \approx 29$ ns), with a more pronounced slower decay component ($A_2/A_1 \approx 0.5$).

Similar non-single-exponential decays have been observed in GaSb/GaAs type-II QDs.[11,12] A quite general picture considering a time-dependent recombination rate of nonequilibrium carriers has been established. For the GaAsSb covered InAs/GaAs QDs investigated here, the underlying processes can be understood as follows. After the carrier excitations, electrons are captured rapidly into the QDs, exhibiting a band bending in the surrounding. On the other hand, holes are captured into the GaAsSb quantum well (QW) and then attracted by the nonequilibrium electrons. The induced band bending tends to confine the hole closer to the QDs and hence increases the electron-hole wavefunction overlap. The faster initial decay time τ_1 can thus be related to the *short-range* radiative recombination of the QD electrons with the holes confined in the surrounding band bending.[11,12] As the nonequilibrium electrons recombine continuously, the band bending reduces, leading to a reduced wavefunction overlap and hence a lower recombination rate. The band-bending effect would eventually become negligible when most of the nonequilibrium carriers have recombined. The longer decay time τ_2 thus reflects the *long-range* radiative recombination of the QD electrons with the holes in the GaAsSb QW states.[11,12]

As elucidated above, the effect of nonequilibrium carriers is expected to be negligible under lower excitation conditions.[11] However, we found that the PL decay traces are still non-single-exponential even though the initial carrier density N_0 was reduced to less than 1 electron/dot. To further clarify this point, temperature dependent TRPL has been performed. The results for the $x=16\%$ sample are displayed in Fig. 3. In this study, the P_{ex} was kept low so that only the GS peak was observed (i.e., $N_0 \lesssim 2$ electrons/dot). We found that the PL peak exhibits a so-called *S*-shaped energy shift with the temperature. This is a typical feature of carrier localization effect and has already been observed in many alloy systems, such as InGaN/GaN and GaInNAs/GaAs QWs.[13,14] Because no *S*-shaped feature was observed in the reference InAs/GaAs QDs, the carrier localization must be in the GaAsSb capping layer, due possibly to alloy fluctuations and/or Sb clustering.

By comparing the *S*-shaped feature with the temperature dependent decay traces, the carrier dynamics of the non-single-exponential decay become clear. As shown in Fig. 3(b), the slower decay component was observed only at $T<100$ K. Because photogenerated holes are trapped by the localization states in the GaAsSb QW at low temperatures, they are less mobile and unable to be attracted into the bend-bending region induced by the QD electrons. This explains why the slower decay component can be observed at $T<100$ K even under low excitation conditions. With the increasing T from 30 to 100 K, these trapped holes gain thermal energy and start to populate the two-dimensional (2D) state of the GaAsSb QW. As the temperature was further increased, most of the holes were delocalized to the 2D QW state and hence could be efficiently attracted into the bend-bending region. Accordingly, the decay traces exhibit a well-defined single decay time constant τ_1 at $T \gtrsim 100$ K, as expected for low excitation conditions.

Quantitatively, the hole localization energy can be deduced from the *S*-shaped energy shift. By using the relation combining the Varshni equation and localization effect.[15] $E(T)=E(0)-\sigma^2/kT-\alpha T^2/(\beta+T)$, with parameters $E_0=0.998$ eV, $\alpha=11\times 10^{-4}$ eV/K, $\beta=600$ K, and a localization energy $\sigma \approx 14$ meV, a reasonable fit can be obtained [solid line in Fig. 4(a)]. An Arrhenius plot of the time-integrated intensities of the faster ($I_1=A_1\tau_1$) and the slower ($I_2=A_2\tau_2$) decay components obtained from the TRPL traces is shown in Fig. 4(b). In the range of $T=30-100$ K (the shadow region), I_2 decreases while I_1 increases in such a way that the total intensity I_1+I_2 remains nearly constant. Such intensity changes clearly elucidate the carrier transfer pro-

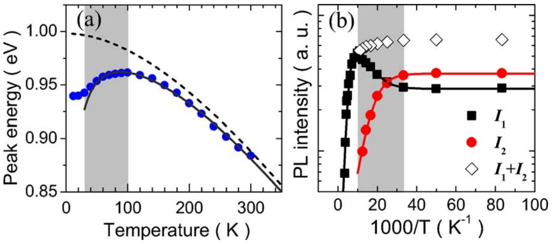

FIG. 4. (Color online) (a) The PL peak energy as a function T. Lines are fitting curves with (solid) and without (dashed) localization effects. (b) An Arrhenius plot of the time-integrated intensities for the faster (I_1) and the slower (I_1) decay components.

cess: the trapped holes are gradually delocalized by thermal energy and attracted into the bend-bending region surrounding the QDs. The activation energy for I_2 was found to be $E_A \approx 18$ meV, close to the deduced localization energy σ.

For $T \gtrsim 100$ K, the faster time constant is $\tau_1 = 3.3 \pm 0.4$ ns and is insensitive to T, as expected for well-confined zero-dimensional carriers. The time constant[16] is approximately four times longer than the reference InAs QDs, corresponding to an electron-hole wavefunction overlap of ~50% of the type-I QDs. This value is considerably larger than that expected for a type-II exciton.[10] The appreciable overlap may be enhanced by the quantum confinement of the GaAsSb/GaAs QW. In addition, the small band discontinuity between InAs and $GaAs_{0.84}Sb_{0.16}$ is also responsible for such an appreciable overlap.

In summary, carrier dynamics of type-II InAs/GaAs QDs covered by a thin GaAsSb layer have been investigated by TRPL measurements. Different recombination paths in such type-II QDs have been clarified by temperature dependent measurements. The long-range recombination with the holes trapped by localized states in the GaAsSb QW is significant at low temperatures. At higher temperatures, the recombination is dominated by the holes confined to the band-bending region surrounding the QDs.

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[16] The measured τ_1 at $T \simeq 100$ K can be approximated as the actual recombination lifetime because both the temporal PL shift caused by the nonequilibrium electrons and the hole localization effect are less significant. For $T > 200$ K, however, the rapid decrease in the integrated PL intensity indicates that the nonradiative processes are increasingly important.