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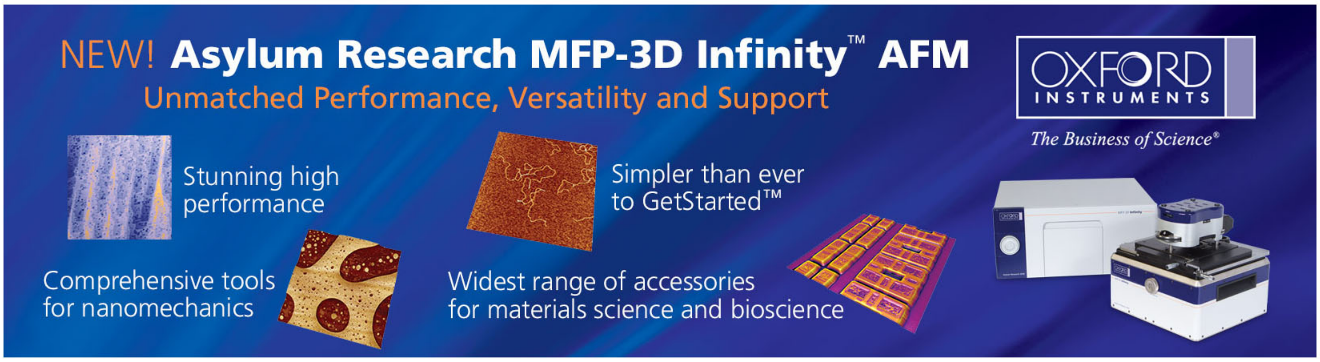

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Effects of GaAsSb capping layer thickness on the optical properties of InAs quantum dots

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The optical properties of GaAsSb-capped InAs quantum dots (QDs) with different capping layer thickness are investigated. Both the emission energy and the recombination lifetime are found to be correlated with the capping layer thicknesses. Theoretical calculations indicate that the quantum confinement and the wave function distribution of hole states are sensitive to the GaAsSb capping layer thickness. The Sb induced change in QD size also plays a role in the optical properties of GaAsSb-capped QDs. Controlling the GaAsSb capping layer thickness is a feasible way to tailor the InAs QDs for long-wavelength applications. © *2011 American Institute of Physics*. [doi:10.1063/1.3624464]

Recently, GaAsSb-capped InAs/GaAs quantum dots (QDs) have attracted much attention because of its capability of extending the emission wavelength to 1.55 μm or beyond.[1,2] It has been demonstrated that the impacts of the GaAsSb capping layer (CL) on the underlying InAs QDs are manifold. First, like the conventional InGaAs CL,[3] the GaAsSb CL acts as a strain reducing layer for the QDs, resulting in a redshift in the emission wavelength. Second, the reduced strain in the CL together with the surfactant effect of Sb atoms would suppress the decomposition of InAs QDs during the capping processes[4,5] and thereby preserving the island height as compared with GaAs-capped QDs. The third and the most prominent effect is the large valence band offset at the InAs-GaAs$_{1-x}$Sb$_x$ heterointerface, which could lead to the formation of type-II QDs as x exceeds $\sim$0.14.[6,7] Experimental evidences for the type-II QDs have been reported based on photoluminescence (PL)[7] and time-resolved PL (TRPL) measurements.[8,9] The long recombination lifetime of spatially indirect excitons and the confinement of only one carrier species make the type-II QDs very promising for memory devices[10] and solar cells.[11] Therefore, tailoring the transition energy, the band alignment, the wave function overlaps, and hence the carrier dynamics are desirable for specific applications. Variation of the Sb content in the GaAsSb CL (Refs. 5 and 6) and post-growth thermal treatments[12] have been employed to achieve this goal. Another approach is to change the GaAsSb CL thickness, which is expected to affect the quantum confinement of hole states and the strain distribution surrounding the type-II QDs. However, not much attention has been paid to the evolution of optical properties of the GaAsSb-capped InAs/GaAs QDs with the CL thickness. In this letter, we investigate the evolutions of emission energy and recombination lifetime of the GaAsSb-capped InAs/GaAs QDs with the CL thickness. The effects of the CL thickness on the hole states and their wave function distributions are discussed and compared with eight-band $\mathbf{k}\cdot\mathbf{p}$ model calculations.

The samples were grown by molecular beam epitaxy. A layer of self-assembled InAs QDs (2.7 MLs) were grown at 500 °C on the GaAs buffer layer and subsequently capped by a GaAs$_{0.8}$Sb$_{0.2}$ layer with a thickness t. Four samples with $t = 0$, 2.5, 5, and 10 nm have been grown. The samples were finally capped by a 50 nm GaAs layer. Atomic force microscopy revealed that uncapped surface QDs are lens shaped, with an average height of 8.0 ± 0.5 nm, an average diameter of 20 nm, and an areal density of about 3×10^{10} cm^{-2}. PL was excited by an Ar$^+$ laser (488 nm) and detected by an InGaAs photomultiplier tube. TRPL measurements were performed using a 50 ps pulsed laser diode (405 nm/2.5 MHz) and recorded using the time-correlated single photon counting technique with a temporal resolution of $\sim$150 ps.

Figure 1(a) shows the PL spectra measured at T = 12 K for the QD samples under a low excitation power ($P_{ex} = 10\,\mu$W). A clear redshift of the PL peak with the increasing GaAsSb CL thickness is observed. For the nominal Sb content of $x = 0.2$ in the CL, the InAs-GaAsSb interface is expected to exhibit a type-II band alignment.[5–7] Therefore, the PL redshift with the increasing CL thickness can be attributed to the combined effects of the formation of type-II QDs,[5–7] the reduced quantum confinement of the hole states, as well as the modifications in the strain distribution in the CL layer. Besides, the GaAs$_{1-x}$Sb$_x$ capping (with $x > 0.2$) could increase in the dot height due to the suppressed QD decomposition.[4,5] However, the evolution of QD size with the GaAsSb CL thickness remains unknown. To gain information about the structural changes by the GaAsSb capping, cross-sectional transmission electron microscopy (TEM) have been performed, which are shown in Figs. 1(c)–1(f). For the GaAs-capped QDs, the islands are flat in shape, with dimensions of about $h = 2.5$ nm in height and $d = 18$ nm in diameter. After the GaAsSb capping, a gradual increase in the QD size with the CL thickness is observed. The estimated heights (diameters) are 3.1 nm (21 nm), 4.1 nm (21 nm), and 5.2 nm (24 nm) for CL thickness $t = 2.5$, 5, and 10 nm, respectively. Although accurate determinations of the QD size and shape are hindered by the strong strain field contrast in the TEM images, a clear

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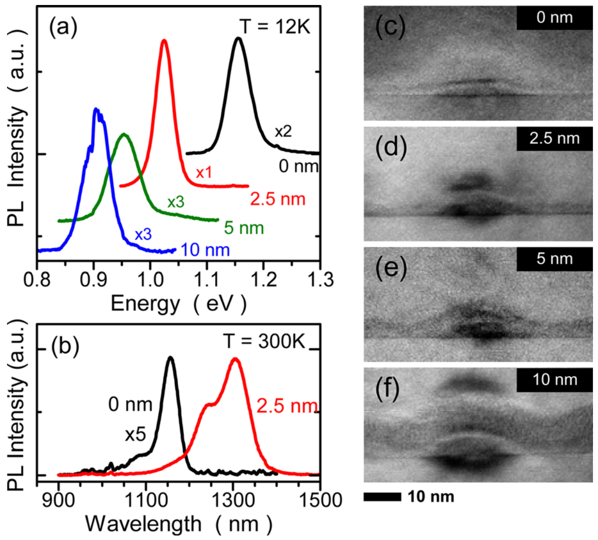

FIG. 1. (Color online) (a) The PL spectra measured at T = 12 K for the investigated QD samples. (b) The room-temperature PL spectra for the samples with $t = 0$ and 2.5 nm. (c)–(f) The cross-sectional TEM images for the samples with different CL thicknesses.

increasing trend of the QD size with the CL thickness can still be inferred. This means that the enlarged QD size should also be considered in the PL redshift with the CL thickness.

To clarify the major effect of the CL thickness, we have performed power dependent PL measurements, which are shown in Fig. 2. For the GaAs-capped QDs, the ground-state peak energy remains nearly constant in the investigated power range. By contrast, the GaAsSb-capped samples with $t = 5$ and 10 nm show large blueshifts with the increasing excitation power, which are clear signatures of the formation of type-II QDs after GaAsSb capping.[1,7,9] However, as the CL thickness was reduced to $t = 2.5$ nm, only a moderate blueshift of 15 meV is observed. This indicates that a thinner CL thickness tends to reduce the type-II character of the GaAsSb-capped InAs QDs.

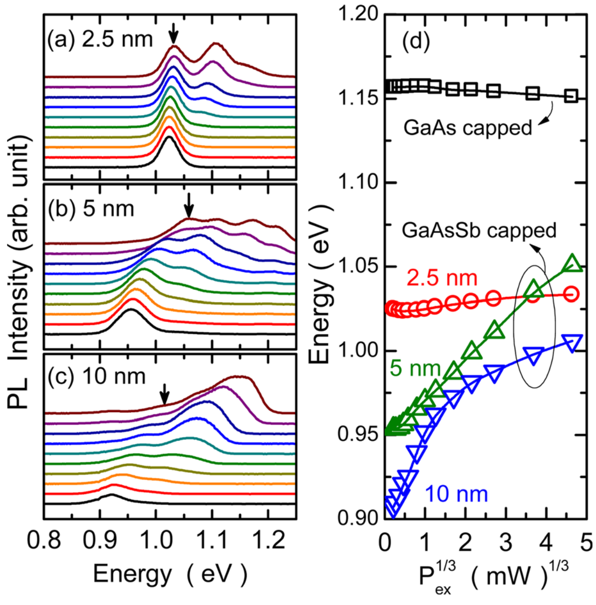

FIG. 2. (Color online) Power-dependent PL spectra for the GaAsSb-capped samples with a CL thickness of (a) 2.5 nm, (b) 5 nm, and (c) 10 nm. All the PL spectra have been offset and the intensities have been normalized to their ground-state peak. (d) The ground-state peak energy of the QDs as a function of $P_{ex}^{1/3}$.

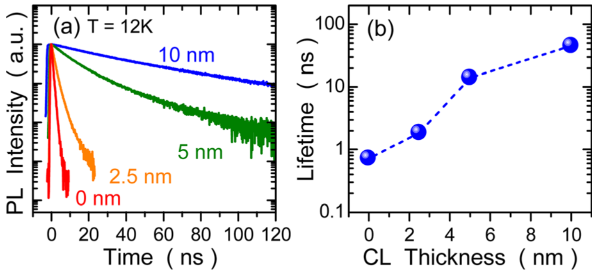

FIG. 3. (Color online) (a) Time-resolved PL spectra and (b) the deduced decay time for the investigated QD samples.

The effect of CL thickness on the radiative recombination lifetime in the GaAsSb-capped QDs has also been investigated by TRPL measurements. For a type-II system, the spatially separated electrons and holes would increase the radiative recombination lifetime τ_R, which is inversely proportional to the square of the overlap integral of the electron and hole wave functions and proportional to the emission energy E_{PL}, i.e., $1/\tau_R \propto |\langle \varphi_e(\mathbf{r})|\varphi_h(\mathbf{r})\rangle|^2/E_{PL}$, where $\varphi_{e(h)}(\mathbf{r})$ is the electron (hole) wave function. Since $\varphi_e(\mathbf{r})$ is still well-confined in the QDs even after the GaAsSb capping, the measured τ_R can thus be a measure of the proportion of $\varphi_h(\mathbf{r})$ that remains in the QDs. Figure 3(a) shows the PL decay recorded at the PL peak under low excitation conditions. The determined τ_R as function of CL thickness are shown in Fig. 3(b). For the GaAs-capped InAs QDs, we obtain $\tau_R = 0.77$ ns, which is comparable to the value reported in literature.[13] By contrast, a gradual lengthening of the PL decay time with the increasing CL thickness is observed for the GaAsSb-capped samples. The deduced τ_R are 1.9, 14, and 45 ns for the samples with $t = 2.5$, 5, and 10 nm, respectively. If we assume that $\langle \varphi_e(\mathbf{r})|\varphi_h(\mathbf{r})\rangle = 1$ in the type-I InAs QDs, the overlap in the GaAsSb-capped samples still has 58% for $t = 2.5$ nm, but decreases to 21% and 11% for $t = 5$ and 10 nm, respectively. This means that the hole wave function distribution in the GaAsSb layer is sensitive to the CL thickness, especially for $t < 5$ nm.

Theoretical calculations based on eight-band $\mathbf{k}\cdot\mathbf{p}$ model[14] have been performed in order to understand the effects of CL thickness quantitatively. For a comparison purpose, we model the InAs QD as a truncated pyramid with $\{101\}$ facets and having a conformal $GaAs_{0.8}Sb_{0.2}$ CL covering thereon with a thickness t. All the material parameters are adapted from Ref. 15, except that the unstrained valence band offsets and the deformation potentials are obtained from Refs. 16 and 17. The strain-induced piezoelectric polarization has also been included. In order to separate the effects of CL thickness on the hole states and the enlarged QD size on the electron states, we have performed two sets of calculations. In the first set, we considered a constant QD size ($h = 3.5$ and $b = 14$ nm) and varying the CL thickness from $t = 0$ to 10 nm. The calculated wave function distributions of the hole ground state on the (110) plane are displayed in Figs. 4(a)–4(d). For the GaAs-capped QD, the hole is well-confined in the QD with a high wave function overlap up to 98%. With the increasing t, the hole wave function penetrates gradually into the GaAsSb layer due to the reduced quantum confinement of

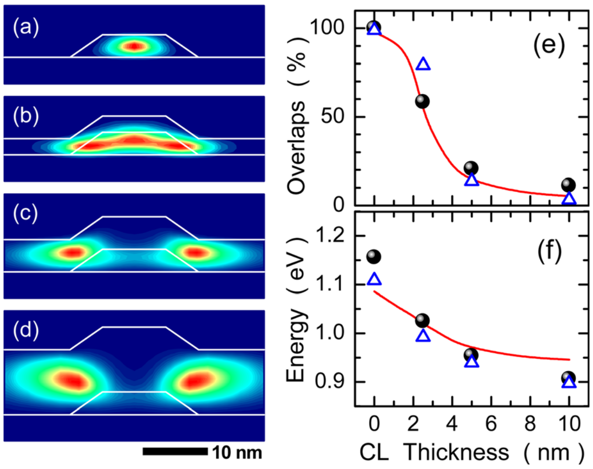

FIG. 4. (Color online) The calculated wave functions of the hole ground state of the InAs QD with a GaAsSb CL thickness of (a) 0 nm, (b) 2.5 nm, (c) 5 nm, and (d) 10 nm. (e) The electron-hole wave function overlaps and (f) the ground state transition energy as a function of the CL thickness, where the solid symbols are experimental data, while the solid curves (open symbols) are calculated results obtained from the first (second) set of calculations.

hole states in the CL. The hole wave function is localized close to the QD base, in consistent with recent calculations.[18] As t is further increased from 5 to 10 nm, the hole wave function becomes more extended in the GaAsSb layer. On the other hand, the electron states, as well as their wave function distributions, are nearly unchanged by the GaAsSb capping. As shown in Fig. 4(e), the calculated wave function overlap (solid curve) decreases gradually from $t = 2$ to 5 nm and become less dependent on the CL thickness for $t > 5$ nm, in agreement with the experimental data (solid symbols). In Fig. 4(f), the calculated transition energy also shows a redshift with the increasing CL thickness. The overall redshift from $t = 0$ to 10 nm is 140 meV, which is however smaller than the experimental redshift ($\sim$250 meV). In fact, we have also calculated different QD sizes (but keeping a constant size for all t) and found that only minor changes in the overall redshift in the transition energy. This indicates that the different CL thicknesses, which affect predominantly the hole states, cannot fully account for the observed PL redshift. Therefore, in the second set of calculations, we further consider the enlarged QD size induced by the GaAsSb capping[5] according to our TEM analysis. All other parameters are kept the same. As shown in Figs. 4(e) and 4(f), the experimental energy shift is well reproduced by the second set of calculations (open symbols). This result indicates that the modification in QD size by the GaAsSb capping still plays a nonnegligible role in the evolution of the optical property of the InAs QDs with CL thickness.

We would like to mention that the GaAsSb-capped sample with $t = 2.5$ nm exhibits a stronger PL intensity and a narrower PL linewidth at $T = 12$ K. This sample also shows a room-temperature PL emission at 1.3 μm with a large enhancement in the integrated intensity ($\sim 7\times$) as compared with the GaAs-capped QDs [see Fig. 1(b)]. Such an improvement in the optical properties is very appealing for long-wavelength emitters. Although the increased dot height of the GaAsSb-capped QDs[4,5] is beneficial for extending the emission wavelength, the formation of type-II QDs for higher Sb contents[6,7] on the other hand hinders them from being efficient light emitters. A trade-off might be researched by optimizing the Sb content in the GaAsSb CL.[5] Our present study suggests that a careful control of the GaAsSb CL thickness ($t < 2.5$ nm) is an alternative approach for extending the emission wavelength while retaining the type-I characters of the QDs.

We have used PL and TRPL measurements to study the emission energy and the recombination lifetime of GaAsSb-capped InAs QDs with different CL thicknesses. Theoretical calculations indicated that the PL redshift and the lengthening of PL lifetime arise not only from the modifications in the quantum confinement of hole states in the GaAsSb layer, but also from the Sb induced structural changes in the QDs. Controlling the GaAsSb CL thickness can be an alternative approach for tailoring the optical properties of GaAsSb-capped InAs QDs.

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