Photoluminescence studies of self-assembled InSb, GaSb, and AlSb quantum dot heterostructures

E. R. Glaser, B. R. Bennett, B. V. Shanabrook, and R. Magno
Naval Research Laboratory, Washington, D.C. 20375-5347

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Photoluminescence (PL) spectroscopy has been performed on a set of self-assembled InSb, GaSb, and AlSb quantum dot (QD) heterostructures grown on GaAs. Strong emission bands with peak energies near 1.15 eV and linewidths of ~80 meV are observed at 1.6 K from 3 monolayer (ML) InSb and GaSb QDs capped with GaAs. The PL from a capped 4 ML AlSb QD sample is weaker with peak energy at 1.26 eV. The PL bands from these Sb-based QD samples shift to lower energy by 20–50 meV with decreasing excitation power density. This behavior suggests a type II band lineup. Support for this assignment, with electrons in the GaAs and holes in the (In,Ga,Al)Sb QDs, is found from the observed shift of GaSb QD emission to higher energies when the GaAs barrier layers are replaced by $Al_{0.1}Ga_{0.9}As$. [S0003-6951(96)03625-X]

Compared to the arsenic and phosphorous material systems, relatively little work has been reported on antimony-based quantum dots (QDs).[1–9] Watanabe *et al.*[10] fabricated InSb QDs on Se-terminated GaAs. We recently demonstrated the self-assembled growth of InSb, GaSb, and AlSb QDs on GaAs by molecular beam epitaxy (MBE).[11] Raman scattering measurements[12] revealed phonons that provide chemical information on the composition of these (In-,Ga,Al)Sb QDs. Emission properties of GaSb/GaAs QDs have been reported recently by Hatami *et al.*[13] In this letter, the optical properties and electronic structure of InSb, GaSb, and AlSb QDs grown with thin GaAs cap layers on GaAs (001) are studied by photoluminescence spectroscopy. The importance of including strain effects in predicting the band structure of these QDs is illustrated in this work.

The photoluminescence (PL) studies were performed on 3 ML InSb, 3 ML GaSb, and 4 ML AlSb self-assembled quantum dots employing the Stranski–Krastanov growth mode. The samples were grown on semi-insulating GaAs (001) substrates by MBE at ~500 °C for the GaSb and AlSb QD structures and ~410 °C for the InSb QD sample. The lattice constants of (In,Ga,Al)Sb are 14.6%, 7.8%, and 8.5% larger, respectively, than that of GaAs. After initial growth of a planar 1–3 ML wetting layer, the QDs spontaneously form as more material is deposited.[14] A 3 ML InAs/GaAs QD sample was also grown[15] in order to compare the optical properties of the Sb-based QDs with the better known emission properties of the InAs QD system.[3,4,7] In addition, a 3 ML GaSb QD sample with $Al_{0.1}Ga_{0.9}As$ barrier layers was fabricated to gain more insight into the electronic band structure of these QD systems. Additional details of the growth conditions and mechanisms are described elsewhere.[11,16] The deposition of a thin GaAs cap layer (~300 Å) was necessary to observe PL from these QD structures. The cap layers were grown at temperatures ~100 °C below the growth temperature of the (In,Ga,Al)Sb and InAs QDs in order to minimize the possibility of altering or destroying the QDs.

Atomic force microscopy (AFM) was employed to obtain the size and density of QD samples grown under the same conditions as the structures studied in this work except without the thin GaAs cap layers.[11,16] The AFM measurements revealed samples with dot densities of $\sim 10^9 - 10^{10}$ cm^{-2}, heights of 50–100 Å, and diameters of 400–600 Å.

The PL from the QD heterostructures was excited by the 488 nm line (2.54 eV) of an Ar^+ laser. Spectra were obtained for excitation power densities of $2.8 \times 10^{-3} - 28$ W/cm^2. The samples were studied at ~1.6 K. The emission was detected by a Ge photodiode.

The PL spectra obtained at 1.6 K under similar excitation power conditions (~2.8 W/cm^2) from the 3 ML InSb, 3 ML GaSb, and 4 ML AlSb QD samples are shown in Fig. 1. Strong emission bands are observed from the InSb and GaSb QD samples with peak energies at 1.13 and 1.16 eV, respectively. The emission from the 3 ML GaSb QD sample is very similar to that reported recently by Hatami *et al.* for GaSb/GaAs QD structures.[13] The PL from the AlSb QDs is about three orders of magnitude weaker with peak energy at 1.26 eV. The emission observed in the low- and high-energy wings of this band is due to recombination from the under-

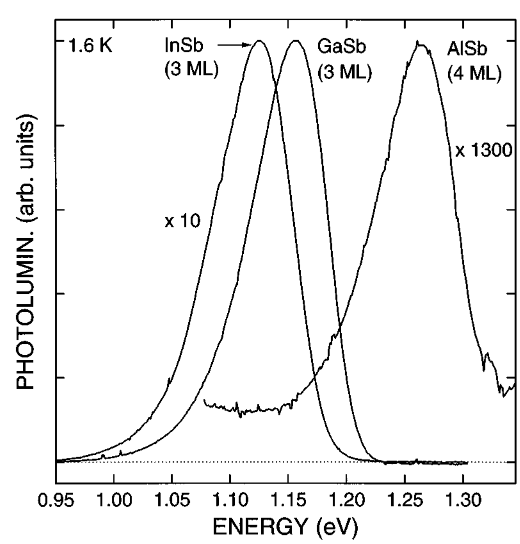

FIG. 1. The photoluminescence spectra obtained at 1.6 K from the 3 ML InSb, 3 ML GaSb, and 4 ML AlSb QD samples ($P = 2.8$ W/cm^2).

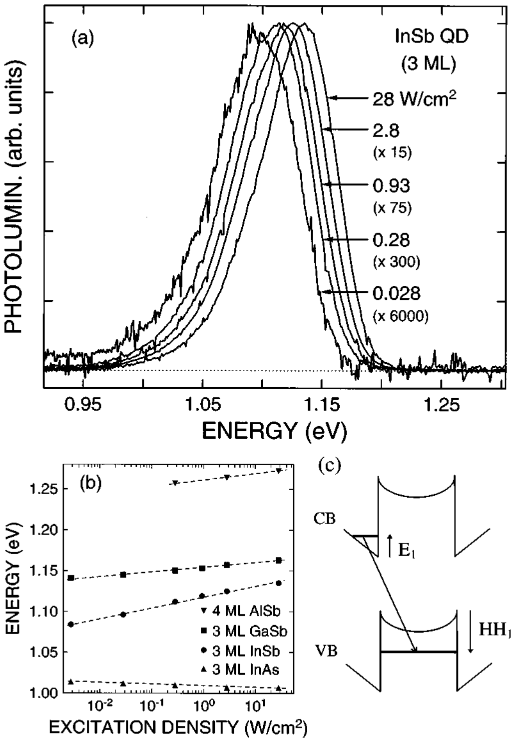

FIG. 2. (a) The PL spectra obtained from the 3 ML InSb/GaAs QD structure for several excitation power densities. (b) The peak energies of the emission bands from the InSb, GaSb, AlSb, and InAs QD samples vs excitation power density. The dashed lines are guides to the eye. (c) Illustration of band bending that occurs at interfaces of a heterostructure with type II band alignment under high excitation power densities.

lying GaAs substrate as confirmed by the PL obtained with the excitation light incident on the substrate side of the sample. In addition, it should be noted that recent Raman scattering experiments[12] on 4 ML AlSb QD samples grown with and without GaAs cap layers revealed phonon modes that provide evidence for segregation of Ga from the GaAs buffer layer into the AlSb QDs. Thus, it is likely that the present nominal 4 ML AlSb/GaAs QD sample capped with GaAs is actually better described as $Al_xGa_{1-x}Sb$ QDs with $0.7<x<0.9$.

We note that the peak energies of the emission from the 3 ML InSb and GaSb QD samples occur at energies much larger than the low-temperature bulk InSb (0.24 eV) and GaSb (0.81 eV) band gap. However, the peak energy of the PL band observed from the 4 ML AlSb/GaAs QD heterostructure is smaller than the low-temperature AlSb indirect band gap (~1.7 eV). As seen in Fig. 1, the PL linewidths for each sample are nearly identical (~80 meV). A contribution to the breadth of these PL lines arises from the convolution of emission from QDs with different sizes since a large number of dots are excited with the 1.5-mm-diam light source employed in this study.

The PL spectra obtained from the 3 ML InSb QD structure for several excitation power densities are shown in Fig. 2(a). The peak energy of this emission band is found to shift to lower energy by 50 meV as the excitation power density is reduced from 28 to 0.002 8 W/cm^2. The peak energies of the emission bands from the 3 ML InSb, 3 ML GaSb, 4 ML AlSb, and 3 ML InAs QD samples are shown in Fig. 2(b). The emission from both the 3 ML GaSb and 4 ML AlSb QD samples is also found to shift to lower energies with decreasing excitation power density. The smaller intensity of the emission from the 4 ML AlSb QD structure compared to the other samples made it difficult to obtain PL spectra of the AlSb QD sample at very low power densities. The shift of the emission band from the 3 ML GaSb QD sample by 22 meV is comparable to that reported recently by Hatami *et al.* from similar GaSb/GaAs QD heterostructures.[13] In sharp contrast to the behavior observed for the Sb-based QD samples, the peak energy of the emission band from the 3 ML InAs reference QD sample is found to have a weak dependence on the excitation power density. The band is observed to shift to slightly higher energies with decreasing power density.

The shift of the PL band to higher energy for the InSb, GaSb, and AlSb QDs is consistent with a type II band alignment with electrons and holes separated in real space. Specifically, the peak shifts with power density for type II structures due to the band bending that occurs at the interfaces [see Fig. 2(c)]. Thus, at high excitation power densities, the resultant Hartree potential shifts the relative energies of the electron and hole states (E_1 and HH_1) and causes a shift of the PL band to higher energy. This behavior is well known, for example, from PL studies of type II GaAs/AlAs short-period superlattices.[17] Hatami *et al.*[13] proposed a similar interpretation of their PL spectra from GaSb QDs. The weak excitation power dependence found for the emission bands from the 3 ML InAs QD sample in this work and from InGaAs QD structures reported by another group[5] is consistent with the assignment[3] of a type I band lineup for these QD systems.

The electronic band structure of (In,Ga,Al)Sb/GaAs QDs capped with GaAs is unknown. However, calculations have been recently performed for the electron and hole levels of type I InAs/GaAs QD heterostructures, taking into account the strain distribution in and around the QDs located in a GaAs matrix.[18] A similar analysis is required to predict the optical transitions for the present Sb-based QD systems. The importance of including strain effects is perhaps best illustrated in this work for the InSb/GaAs QD structures. In particular, a type I band alignment, with electrons and holes located in the InSb QDs, is predicted using the natural valence band offsets from Harrison and Tersoff for the InSb/GaAs heterojunction.[19] However, as noted above, strain effects must be included for calculating the proper band alignment of this structure since the InSb QDs are embedded in a GaAs matrix. If we model the shape of the dots as approximately spherical, the major component of the strain tensor within the InSb dots is hydrostatic in nature.[18] For this situation, we find that the conduction-band (CB) minimum of the InSb QDs is raised above the CB edge of the adjacent GaAs layers. As also found when not including the effects due to strain, the ground-state hole subband is derived from the InSb valence-band (VB) edge. Confinement effects will remove the fourfold degeneracy of the uppermost ($J=3/2$) VB edge in the InSb QDs, with the lowest-energy state for holes derived from the twofold degenerate heavy hole (HH) VB. Thus, a type II band alignment with holes in the InSb QDs and electrons in the adjustment GaAs is predicted after

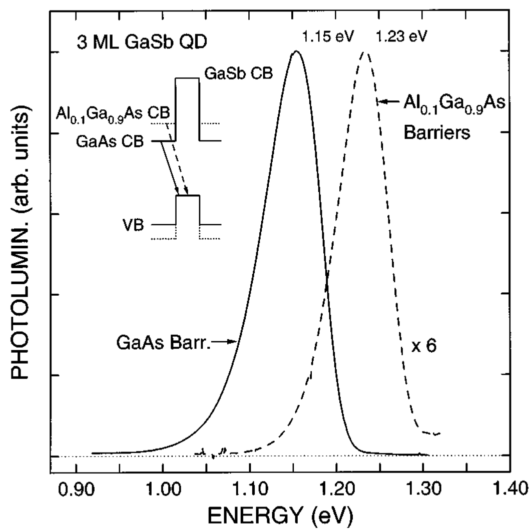

FIG. 3. The PL bands observed from the 3 ML GaSb QD heterostructures grown with either GaAs or $Al_{0.1}Ga_{0.9}As$ barrier layers. Inset: schematic diagram of conduction- and valence-band minima and lowest-energy optical transitions for GaSb/GaAs and GaSb/$Al_{0.1}Ga_{0.9}As$ QD structures.

taking into account both strain and quantum confinement effects. Similar analyses also reveal type II band alignments for the (Ga,Al)Sb/GaAs QD heterostructures and a type I alignment for InAs/GaAs QDs.[20]

In order to determine the location of the electrons and holes that participate in the recombination from these samples, a 3 ML GaSb QD sample was grown with $Al_{0.1}Ga_{0.9}As$ barrier layers instead of GaAs. A shift of the PL band by ~80 meV to higher energy is calculated for such a sample using a type II band lineup model with electrons in the (Al)GaAs barrier layers and holes in the GaSb QDs (see inset in Fig. 3). This shift was estimated applying the transitivity rule,[21] using a conduction-to-valence band offset ratio of 65:35, and assuming small changes in hole confinement energies due to the expected heavy mass of holes in the GaSb QDs.

A comparison of the PL bands observed from the 3 ML GaSb QD heterostructures grown with GaAs and $Al_{0.1}Ga_{0.9}As$ barrier layers is shown in Fig. 3. The peak of the emission band found from the 3 ML GaSb sample grown with $Al_{0.1}Ga_{0.9}As$ barrier layers is indeed found to be shifted by 80 meV above the peak energy of the PL band observed from the 3 ML GaSb QDs surrounded by GaAs. This result supports the assignment of a type II band lineup for these GaSb QD structures (and, by analogy, for the 3 ML InSb and 4 ML AlSb QD heterostructures), with electrons in the (Al)GaAs barrier layers and holes in the GaSb QDs.

In summary, photoluminescence at 1.6 K has been observed from self-assembled InSb, GaSb, and AlSb QDs on GaAs (001) fabricated by Stranski–Krastanov growth. Strong emission bands with peak energies near 1.15 eV are observed from 3 ML InSb and GaSb QD heterostructures. The PL from a 4 ML AlSb QD sample is weaker with peak energy at 1.26 eV. The results of excitation power density studies suggest a type II band lineup for these structures. Support for this assignment with electrons in the adjacent GaAs buffer and cap layers and holes in the (In,Ga,Al)Sb QDs, as predicted by theory that includes both strain and confinement effects, is found from PL studies of GaSb QD structures grown by substitution of $Al_{0.1}Ga_{0.9}As$ for the GaAs barrier layers.

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