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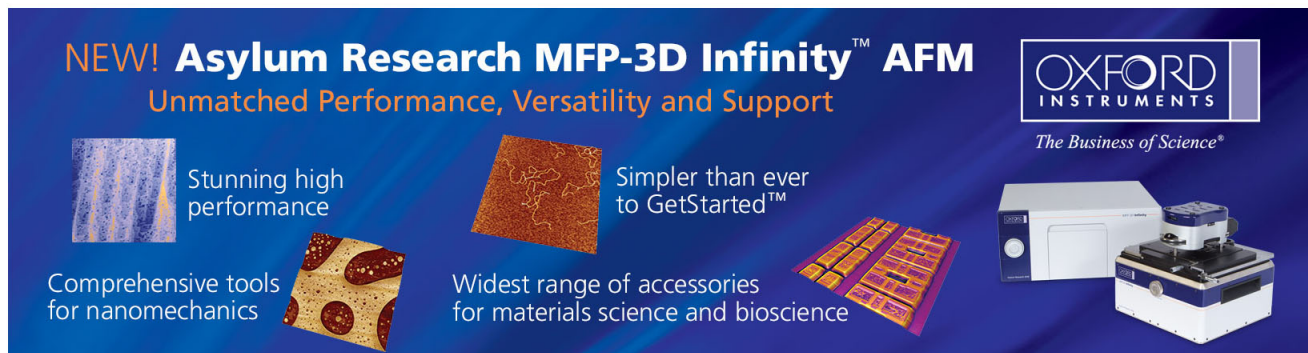
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## Discrete monolayer light emission from GaSb wetting layer in GaAs

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Distinct light emission peaks from monolayers of GaSb quantum wells in GaAs were observed. Discrete atomic layers of GaSb for the wetting layer prior to quantum dot formation give rise to transition peaks corresponding to quantum wells with 1, 2, and 3 ML. From the transition energies the authors were able to deduce the band offset parameter between GaSb and GaAs. By fitting the experimental data with the theoretical calculated result using an  $8 \times 8 \mathbf{k} \cdot \mathbf{p}$  Burt's Hamiltonian along with the Bir-Picou deformation potentials, the strain-free (fully strained) valence band discontinuity for this type-II heterojunction was determined to be 0.45 eV (0.66 eV). © 2007 American Institute of Physics. [DOI: 10.1063/1.2748087]

III-antimonide compounds have been regarded as potential materials for applications in ultrahigh-speed devices and long-wavelength photonic devices due to their high electron mobility and small band gap energies.<sup>1,2</sup> Moreover, heterostructures composed of antimonides and other III-V compounds, such as arsenides, have also been of physical interest because of the unconventional type-II and type-III band alignment.<sup>3-5</sup> There have been lots of theoretical predictions for interesting optical, electronic, and magnetic phenomena in nanostructures with antimonides.<sup>6</sup> Recently, with the advances in epitaxy technology, such nanostructures have been grown with high quality and from them many interesting experimental findings, either predicted or sometimes unexpected, have been observed. For instance, quantum wells (QWs) and self-assembled quantum dots (QDs) made by GaSb embedded in GaAs matrix, because of type-II heterostructures, can provide an opportunity for observation of optical transition between spatially separate electrons and holes around the heterointerfaces. However, despite a large amount of effort being put in this material system, one of the most important parameters, the band offset between GaSb and GaAs has yet to be accurately determined. The wide range of reported value in this parameter often leads to ambiguous interpretation of experimental results and theoretical predictions for nanostructures made from this system.

In this work, we report a study on optical transitions from monolayer-scale GaSb/GaAs QWs. Several distinct emission peaks were observed simultaneously from more than one sample by photoluminescence (PL) measurement. Each of the emission peaks lies at a definite position and can be well assigned to an optical transition from the QW of 1, 2, or 3 ML. Such definite transition energies allow us to determine the band offset between GaSb and GaAs by fitting the theoretical calculation to the measured data. Additionally, we also observed a subordinate emission peak that can be assigned to the optical transition in QDs. The QD emission can be observed only from the sample with a particular nominal GaSb thickness. Our study can therefore provide an insight into the mechanism for the formation of GaSb/GaAs QDs by molecular beam epitaxy (MBE).

The GaSb/GaAs nanostructures were grown by a VECCO Gen-II solid source MBE system with valve cracker sources of antimony and arsenic on (001) GaAs substrates. Two samples were prepared with different nominal GaSb film thicknesses: (1) sample A has a 1 ML GaSb film and (2) sample B has a 2 ML GaSb film. Here, 1 ML GaSb film means the number of GaSb molecules that is needed to form a continuous GaSb layer pseudomorphically matched to GaAs. In both samples, the GaSb films were sandwiched between a 150 nm GaAs buffer layer and a 150 nm GaAs capping layer. After the capping layer growth, a GaSb film with the same thickness was grown on the surface for atomic force microscope (AFM) measurement. We have monitored the reflection high-energy electron diffraction patterns to make sure that the morphology of GaSb layer on the surface is the same as that buried in the sample. The growth temperature for GaSb was 500 °C, the V/III flux ratio was about 5, and the growth rate was 0.2 ML/s. After the growth of the GaAs buffer layer, the As shutter was closed, the Sb shutter was opened, and the GaSb film was immediately deposited. Following that, a 1 min interruption under Sb flux was performed. We used As<sub>4</sub> as the arsenic source instead of As<sub>2</sub> to avoid intermixing of Sb and As atoms during the growth process.<sup>7</sup>

Figures 1(a) and 1(b) show the AFM images of samples A and B, respectively. The smooth surface of sample A clearly shows that 1 ML of GaSb is not enough for quantum dot formation. However, on the surface of sample B, which had a 2 ML GaSb film, we can see clear images of quantum

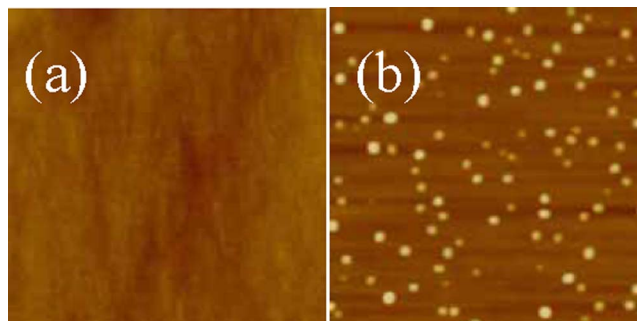


FIG. 1. (Color online) AFM surface images of (a) sample A (with 5  $\mu\text{m}$  square image area) and (b) sample B (with 1  $\mu\text{m}$  square image area).

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dots. The density of the dots is about  $1.04 \times 10^{10} \text{ cm}^{-2}$  and the height and the diameter of QDs are in the ranges of 5–10 nm and 50–60 nm, respectively. The formation of dots implies that the strain of the GaSb film cannot be sustained pseudomorphically in GaAs when more than 1 ML of GaSb is deposited. This is obviously due to the large lattice mismatch (8%) between GaSb and GaAs. However, as will be described in the following, because of the nature of epitaxial growth, 1 ML GaSb deposition does not mean that there is only one kind of quantum wells with thickness of 1 ML. Actually what we found in this work is that the maximum thickness of GaSb wetting layer (or quantum well) is 3 ML.

The PL measurement of the two samples was performed at 15 K with a cw Ar laser as the excitation source. The excitation power was varied from 1 to 100 mW and the beam spot size was around 200  $\mu\text{m}$  in diameter. The measured spectra of samples A and B are shown in Figs. 2(a) and 2(b), respectively. For sample A [see Fig. 2(a)], the dominant peak is  $A_1$  at  $\sim 1.4$  eV. At low excitation levels, this is the only peak observed. However, when the excitation exceeds 100 mW, two additional peaks  $A_2$  and  $A_3$  at about 1.3 and 1.22 eV are observed along with the main peak  $A_1$ . For sample B, the situation is more complicated. From Fig. 2(b), one can see that the emission is dominated by the peak  $B_2$  at  $\sim 1.3$  eV at all excitation levels. With the excitation level increasing, the side peaks  $B_1$  and  $B_3$ , at 1.22 and 1.4 eV, respectively, become more obvious. It is noticed that the three peaks  $B_i$  for sample B appear exactly at the same positions of the peaks  $A_i$  for sample A ( $i=1, 2, 3$ ). At the excitation of 100 mW, we also found a broad peak  $B_{\text{QD}}$  around 1.05 eV in the spectrum for sample B. This peak is absent from the spectra for sample A and should be attributed to optical transition in QDs.

The fact that the positions of the emission peaks from two different samples are identical indicates that they originate from optical transitions in QWs with the same thickness. The fact that the QW in sample A has a nominal thickness of 1 ML allows us to assign the main peak  $A_1$  to the optical transition in 1 ML QWs. Consequently, the lower peaks  $A_2$  and  $A_3$  at lower energies can be assigned to optical transitions in 2 and 3 ML QWs, respectively. As will be seen later, this assignment is in reasonable agreement with our calculation result. Based on a similar argument, the main peak  $B_2$  of sample B, which was deposited with 2 ML of GaSb, is due to the optical transition in 2 ML QWs. The two side peaks  $B_1$  and  $B_3$ , therefore, naturally come from optical transitions in 1 and 3 ML QWs, respectively. The emission peak from the quantum dots  $B_{\text{QD}}$  is obviously wider than those from the quantum wells. This is caused by the nonuniformity in the size of the QDs. In contrast, the emission peaks from the thin QWs are narrower. The discreteness of the atomic layers in these monolayer QWs gives no contributions to the linewidth broadening due to thickness fluctuations, which is normally seen in wide QWs.

The emission spectra described above have revealed that thin QWs with different number of atomic layers can coexist when a few monolayers of GaSb are deposited in GaAs. Such peculiar emission spectra have not been seen in the more popular InAs/GaAs system. The InAs wetting layer prior to the quantum dot formation usually shows only one broad emission peak. While it is unlikely that the thin InAs layer is atomically smooth without steps, the single emission peak is probably due to an average effect of the transition

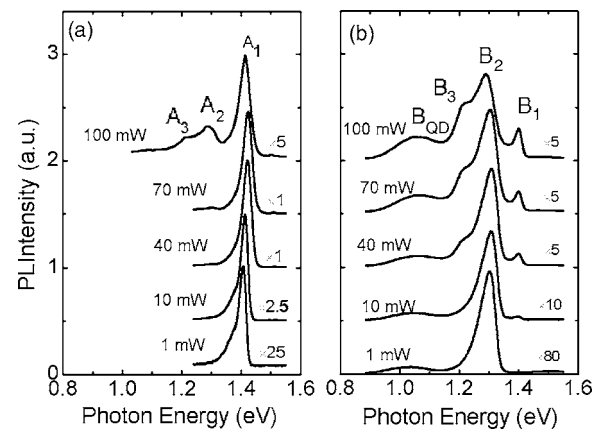


FIG. 2. Photoluminescence spectra of (a) sample A and (b) sample B at 15 K.

energy caused by some kind of intermixing between the In and the Ga atoms after the quantum dot formation. On the other hand, the coexistence of 1, 2, and 3 ML QWs in the GaSb films indicates that the morphological pattern with atomic steps remains intact after they are covered with GaAs. In this work, we used  $\text{As}_4$  instead of  $\text{As}_2$  as the arsenic source for GaAs growth, which may have contributed to the reduction in intermixing between As and Sb.<sup>7</sup>

Because of the spatial separation between electrons and holes in the type-II GaSb/GaAs heterostructures, the carrier recombination lifetime is long and the carrier density can significantly increase with the excitation power. It follows that the electric field, which is induced by the spatially separated electron-hole space charges, increases around the heterointerface with the excitation power. As illustrated in the inset of Fig. 3, the increased electric field causes an upward shift of the energy level in the triangular potential well on the GaAs side. The observed blueshift for the emission peaks from GaSb/GaAs heterostructures is usually proportional to the 1/3 power of the excitation level, which is consistent with the triangular well approximation for electrons in GaAs.<sup>2,3</sup> Figure 3 shows the energy positions of the emission peaks  $A_1$  and  $B_2$  as functions of the cubic root of the excitation power density. The upper and lower lines in the figure, corresponding to  $A_1$  and  $B_2$ , respectively, show how the transition energies in the 1 and 2 ML QWs change with excitation power. As seen in the figure, the energy shift follows the 1/3 power dependence quite well. However, it should be

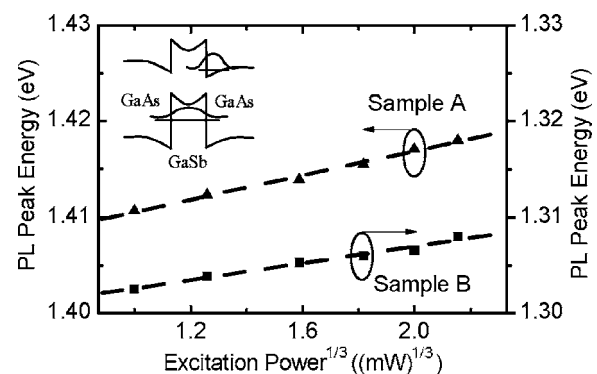


FIG. 3. Measured transition energy vs the cubic root of the excitation power. The intercepts of the extrapolated lines with the y axis give the transition energies at thermal equilibrium. The carrier transition diagram for a GaSb/GaAs quantum well is shown in the inset.



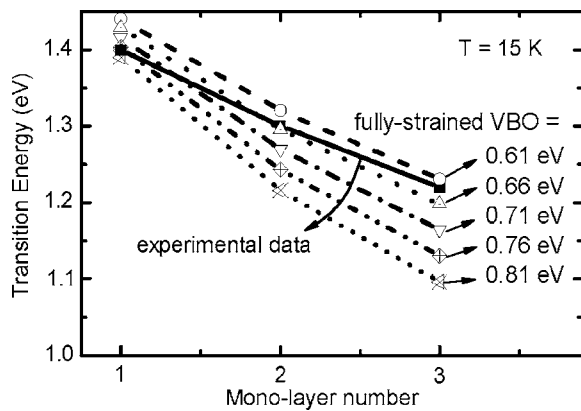


FIG. 4. Transition energies of quantum wells with 1, 2, and 3 ML of GaSb. The measured result is compared with the theoretical result.

noted that the excitation power was kept below 10 mW in this plot. At high pumping powers, because of local heating, the 1/3 power law dependence will no longer be valid.

The plot in Fig. 3 can be used to obtain the transition energies of 1 and 2 ML GaSb/GaAs QWs in the absence of the excitation by linear extrapolation of the lines to the y axis. The obtained transition energy can be considered as the band gap energy of GaSb/GaAs QWs at thermal equilibrium. The resulting band gap energy is 1.404 eV for 1 ML QW and 1.298 eV for 2 ML QW. For the 3 ML QW, we cannot adopt the extrapolation method described above since we lack a series of appreciable emission signals at low excitation levels. Instead, we estimate the band gap energy at thermal equilibrium for the 3 ML QW to be about the position in energy of the peak  $B_3$  obtained from sample B at excitation of 70 mW. This may overestimate the band gap energy according to the band filling effect mentioned above, but because the heating effect can compensate part of the energy shift, we expect the error is reasonably small compared to the difference from the band gap energy of 1 or 2 ML QW.

Now that the band gap energies of 1, 2, and 3 ML QWs have been obtained by the experiment, we can determine the band offset between GaSb and GaAs by fitting the theoretically calculated band gap to the experimentally obtained data with the band offset as an adjusting parameter. To this end, we use the eight-band  $\mathbf{k}\cdot\mathbf{p}$  model to calculate the valence band structures of GaSb/GaAs QW in the flatband approximation for various values of band offset.<sup>8</sup> The strain effect is considered, assuming the GaSb is pseudomorphically grown on strain-free GaAs, using the Bir-Picus deformation potential theory. The parameters taken for calculation can be found in Ref. 9. The calculated transition energy is then the difference between the GaAs conduction band edge and the first heavy-hole subband edge in the GaSb QW. Figure 4 shows the calculated transition energy of 1, 2, and 3 ML GaSb QWs for a series of valence band offset (VBO) values, along with the data obtained from measurement, where VBO is the difference in valence band edge between fully strained GaSb and strain-free GaAs. As can be seen, the variation of the

transition energy with the QW thickness is quite consistent between the calculation and the measurement, proving the correctness of our previous assignment of the emission peaks to optical transition in QWs with definite monolayer-scale thickness. Comparison between the calculated and the measured data suggests that the VBO should lie in the range of 0.61–0.81 eV. The strain-free VBO should lie in the range of 0.4–0.6 eV. For the best fitting, the fully strained VBO is 0.66 eV and the strain-free VBO is 0.45 eV. The slight deviation in the calculated data is attributed to the emission of band bending in our calculation, which is particularly important to the structure with the 3 ML QW. There is a wide range of VBO values (from 0.12 to 0.9 eV) reported in the past.<sup>10–13</sup> Our study has narrowed significantly the VBO range. Previously, Ledentsov *et al.* have compared the measured transition energies of thin GaSb layers with their theoretical calculations, and found a large discrepancy.<sup>3</sup> This is most likely due to a choice of a large VBO.

In conclusion, we have observed distinct PL peaks from monolayer GaSb layers in GaAs. Such peaks have been identified to be originated from optical transitions in 1, 2, and 3 ML GaSb/GaAs type-II QWs. The observed phenomenon is peculiar to the monolayer-scale GaSb/GaAs layers and has never been found in the more popular InAs/GaAs structures. A range of valence band offset for the strain-free GaSb/GaAs heterojunction from 0.4 to 0.6 eV is suggested according to the fitting of the measured data to our eight-band  $\mathbf{k}\cdot\mathbf{p}$  calculation.

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