

Effects of *In-situ* UV Irradiation on the Uniformity and Optical Properties of MBE Grown GaAsBi Epi-layers

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The remarkable tunability of the band gap and spin orbit splitting in III-V alloys with only a small amount of bismuth makes the alloy potentially useful material for many applications. However, it is difficult to synthesize GaAsBi epi-layers with sufficiently high optical quality hampering the technological impact of the material. Spectral linewidths are typically exceedingly broad and band edge emission often suppressed at low temperatures by recombination at low energy states. In-situ UV irradiation of semiconductor alloys has been shown to lead to material quality improvements in the past and we apply it here to the growth of GaAsBi. The samples were irradiated by a pulsed 248nm laser focused to a 7x7mm spot. Rotation during growth results in the geometry shown in Fig. 1, where there is a uniformly illuminated central region and a periphery that is radiated by a fluctuating fluence. With irradiation GaAsBi was shown to be much improved[1], where luminescence linewidths as low as 14meV were demonstrated and band edge emission observed to low temperatures.

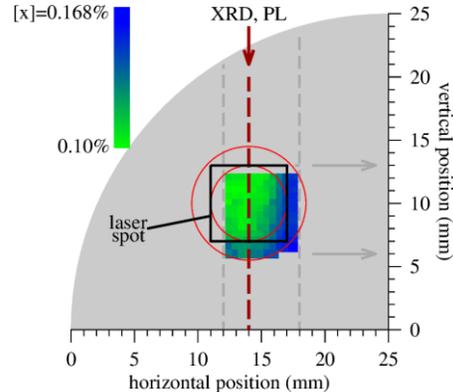


Fig. 1: Sample layout showing the central illuminated region and the vertical strip of the sample which was used for the PL spectra. The scale bar indicates the GaAsBi composition from XRD.

Using the inherent variation of the fluence across the sample we explore the role of the irradiation[2]. In the central uniformly lit region, steady state growth processes are achieved more quickly, yielding more abrupt interfaces, as well as uniform GaAsBi epi-layers.

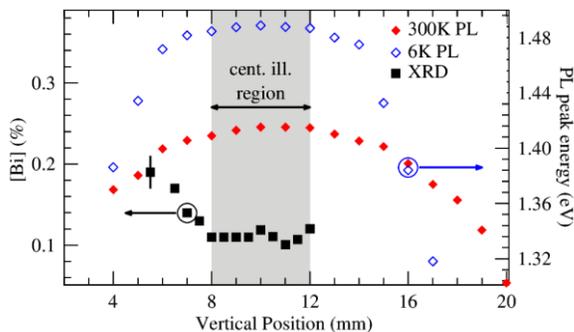


Fig. 2: Bismuth composition and PL peak energies as a function of position on the wafer, compositions found from XRD also shown. The extent of the central illuminated region of the sample is shown.

Comparing photoluminescence spectra at low (6K) temperature shows an increasing density of cluster related emission with decreasing fluence. This is observed in the shift in the peak emission energy away from the band edge, where the peak is shifted most dramatically far from the illuminated region, see Fig. 2. These results indicate a reduction in clustering of incorporated bismuth atoms with the use of the incident UV irradiation, and additionally the density of clusters may be controlled by the degree of irradiation.

[1] D.A. Beaton, A. Mascarenhas, and K. Alberi, J. Appl. Phys. **118**, 235701 (2015)

[2] D.A. Beaton, M. Steger, T. Christian, and A. Mascarenhas, J. Crys. Grow. **484**, 7-11 (2018)

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In-Situ UV irradiation of GaAsBi: Supplementary Pages

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Despite improvements in overall material quality GaAsBi epi-layers exhibit strong recombination from states within the band gap at low temperatures and exceedingly large spectral linewidths. Inhomogeneity in the distribution of bismuth atoms in the crystal are the root cause of this. Using in-situ UV irradiation of the as-growing surface, GaAsBi luminescence linewidths of 14 and 37meV at low temperature and room temperature were demonstrated. During growth samples are irradiated with light from a 248nm KrF excimer. Where sample rotation results in a circular uniformly illuminated central region, along with a small peripheral region that is non-uniformly illuminated. Initial observations indicate that, along with the UV irradiation, the amount of bismuth on the as-growing surface plays a critical role in the improved optical properties. The nature of the role of UV irradiation plays is explored by exploiting this inherent varied irradiance across a sample.

Using high resolution XRD, samples were mapped and a subset of these spectra are shown in Fig. A. The GaAsBi peak is a shoulder in the spectra, indicated in the figure along with the GaAs substrate main spectral feature. The composition of the sample was found to be roughly circularly symmetric around the 10mm vertical position, where this corresponds to the expected location of the laser spot on the sample. XRD spectra were fit with a simple bi-layer of GaAsBi; a thin initialization layer and a second thick GaAsBi epi-layer of uniform composition. The central region of the illuminated area is defined here by the observation of clear Pendellosung fringes in the XRD spectra, which span the vertical positions between 8.5 and 12mm. Observation of these

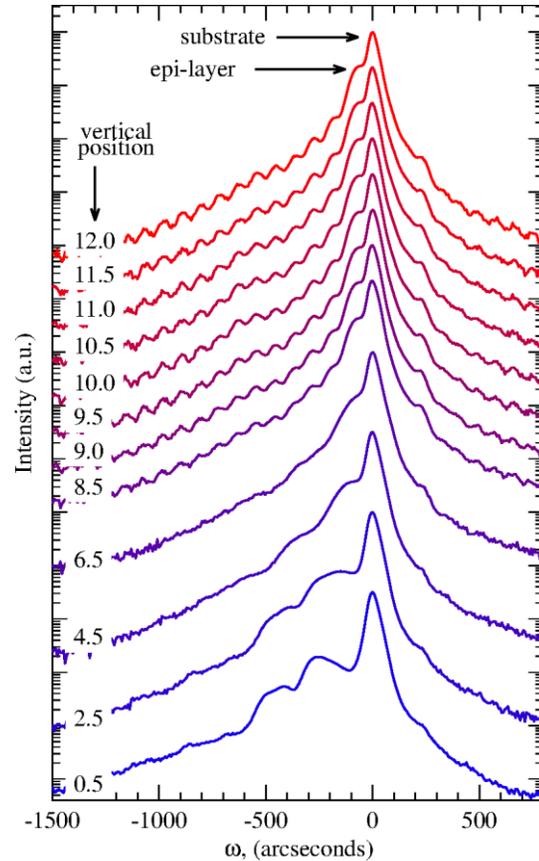


Fig. A: XRD spectra along through the central illuminated region. Only in this region are fringes observed, indicating the film is uniform and has abrupt interfaces. Away from the central region the sample shows increasing non-uniformity.

fringes indicate that within the central illuminated region of the sample i) the interface between the GaAsBi initialization layer and substrate is abrupt, and ii) the GaAsBi epi-layers are of uniform composition.

Figure B shows a series of low (6K) temperature and room temperature PL spectra taken along a similar vertical trace of the sample. The same trend is again found. The PL spectra within the central region exhibits a single intense peak positioned very close to the expected band gap energy; E_g based on the XRD measurements of the composition. The

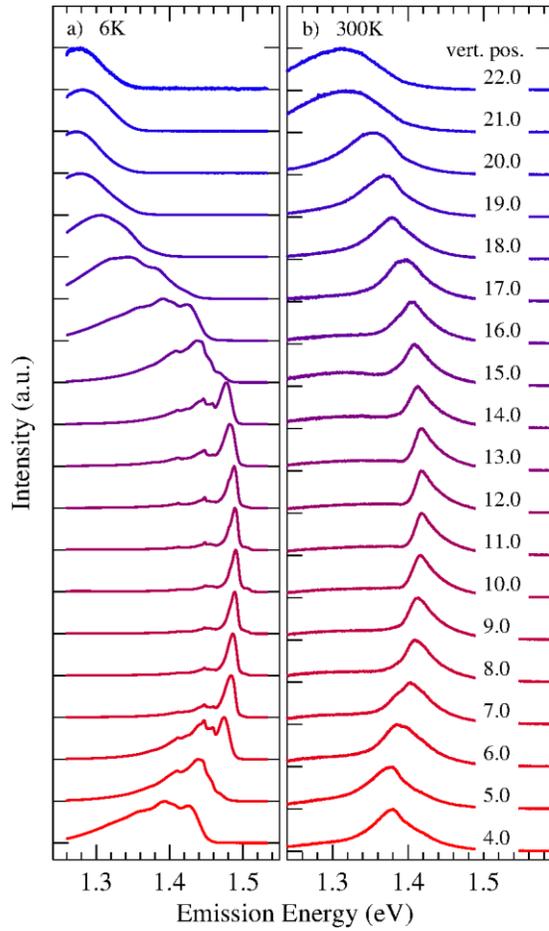


Fig. B: Luminescence spectra taken vertically along a sample. The central illuminated region of the sample shows the narrowest peak linewidth and least low energy emission. Away from the illuminated region broadened emission is observed, along with increased emission from low energy states at low temperatures.

low temperature spectra from areas far from the central region show increasing emission from states well below the band gap energy. This relatively deep low temperature PL emission is commonly observed and often attributed to recombination at bound states associated with bismuth pairs and clusters. The trend in the low temperature PL spectra suggests that the density of bismuth-related cluster states is reduced within the illuminated area of the sample. At 300K, the shift of peak energies to lower energy is less pronounced as many carriers thermalize

out of the lowest energy trap states. Emission from the pair and cluster states continues to contribute to PL linewidth broadening at room temperature, which is also commonly observed in GaAsBi.

To verify the observed effects are related to incident fluence, PL spectra from the central illuminated region from three samples are compared Fig. C, along with a spectrum from the peripheral area. The spectra from the central illuminated region all show a dominant recombination from a band edge related state, as well as weaker emission from recombination at pair and cluster related states. The spectra from the peripheral is dominated by emission from recombination from pair and cluster related states. Note that the spectral shape of the lower energy emission is much broader for the periphery spectrum due to a distribution of clusters involved in recombination; this is another indication that the illumination plays a significant role in the incorporation of bismuth adatoms into the host lattice.

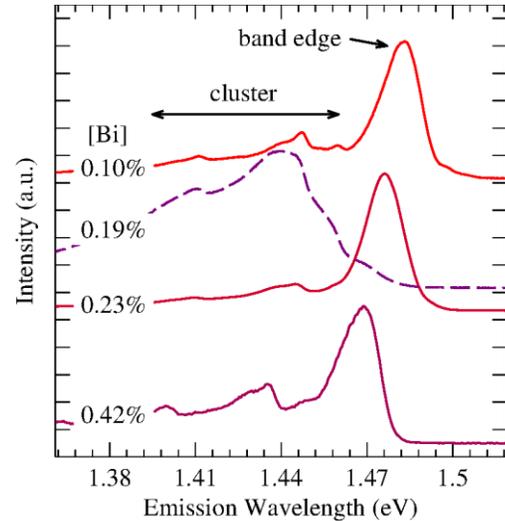


Fig C: Comparison of 6K PL from the central illuminated region (solid curves) and from the periphery (dashed curve). Central illuminated region spectra show band edge related emission, whereas the spectrum from the peripheral region is dominated by pair and cluster related states.