Mechanisms of Compositional Inhomogeneities in Bismide Films

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III-V semiconductor alloys containing Bi have attracted attention due to their novel properties, including a large reduction of bandgap [1], reduced temperature dependence of the bandgap [2], and an increase in spin orbit-coupling [3] with increasing Bi concentration. It has proven difficult to grow high Bi content films, as droplet formation and compositional inhomogeneities arise during growth. These phenomena are crucial to understand, because such fluctuations can cause carrier localization and degradation of device performance. Kinetic Monte Carlo growth simulations (Fig. 1a.) predict that the highest Bi incorporation rates occur when Ga droplets form on the surface [4]. This is because Bi incorporation requires a high availability of Ga [5]. However, growths exhibiting Ga droplets on the surface result in compositional fluctuations (Fig. 1b). We postulate this effect is caused by local variations in Bi incorporation rates due to the nonuniform Ga availability near the droplet. Indeed, high contrast related to high scattering from Bi on the surface near the droplet is seen in Fig. 1b. It was also observed that droplets led to degradation of film crystallinity, verified with X-Ray diffraction [6]. Growing under a higher As/Ga ratio eliminates Ga droplets, but not necessarily compositional inhomogeneities. Figure 1c. shows the formation of a lateral composition modulation. Nanometer-sized clusters of Bi-enriched GaAsBi also form [7]. Raising the growth temperature can mitigate lateral composition modulation and clustering, but results in lower Bi incorporation. In this talk we will also map out the droplet free-growth conditions to maximize the Bi incorporation and mitigate droplet-induced inhomogeneities.

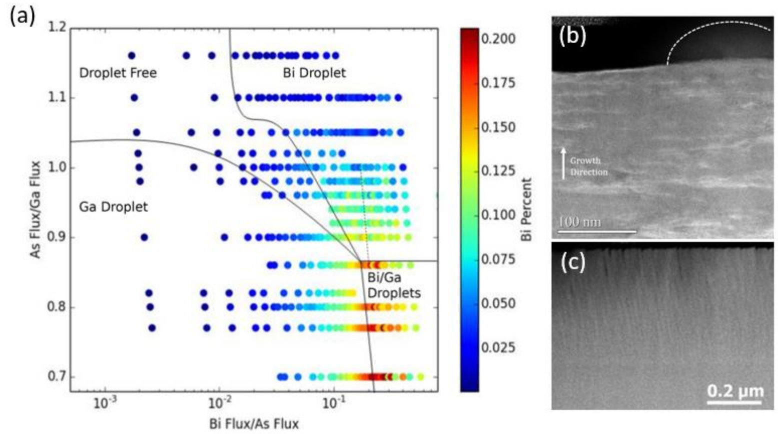

Figure 1: (a) Surface phase diagram and composition map of simulated GaAsBi growths at T=260C and various flux ratios. High angle annular dark field transmission electron micrographs of GaAsBi films grown at 315C and **(b)** As/Ga flux ratio= 1.7 **(c)** As/Ga flux ratio= 3.5

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[2] J. Yoshida, T. Kita, O. Wada, and K. Oe, Japanese J. Appl. Physics, vol. 42, no.2A, pp.371–374, 2003.
[3] B. Fluegel, S. Francoeur, A. Mascarenhas, S. Tixier, Phys. Rev. Lett., vol. 97, no. 6, pp. 11–14, 2006.
[4] G. V. Rodriguez and J. M. Millunchick, J. Appl. Phys., vol. 120, no. 12, pp. 0–6, 2016.
[5] C. R. Tait and J. M. Millunchick, J. Appl. Phys., vol. 119, no. 21, 2016.
[6] C. R. Tait, L. Yan, and J. M. Millunchick, Appl. Phys. Lett., vol. 111, no. 4, pp. 1–5, 2017.
[7] C. R. Tait, L. Yan, and J. M. Millunchick, J. Cryst. Growth, vol. 493, no. April, pp. 20–24, 2018.

Supplementary Information:

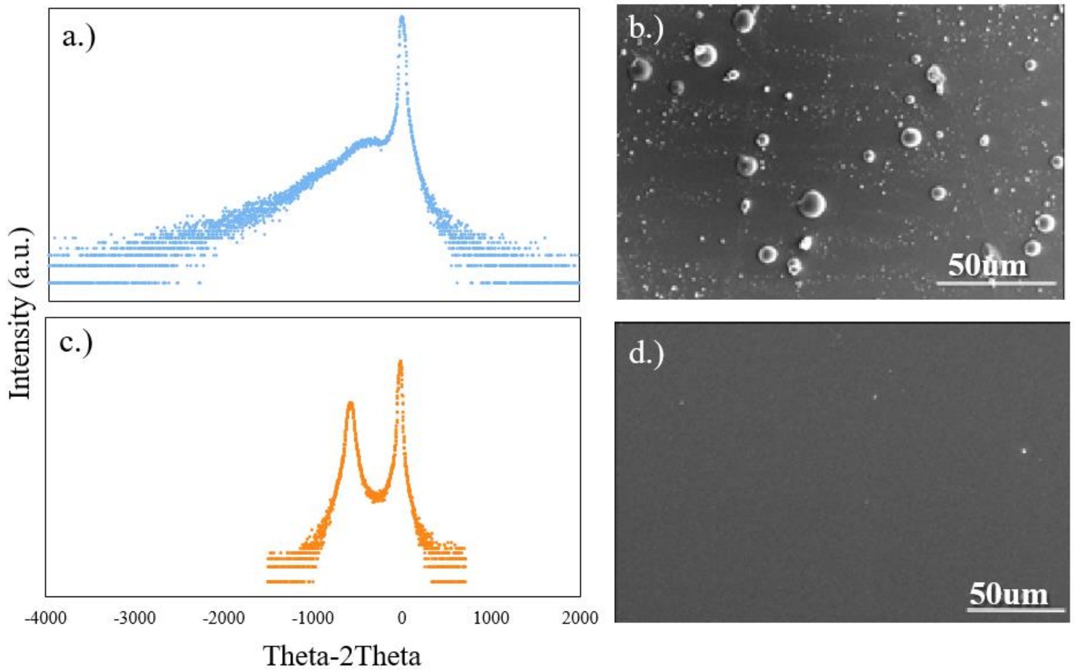

Figure 1S. a, b) XRD and SEM respectively of a sample grown at As / Ga flux ratio of 1.7 and 315C demonstrating a very broad film peak indicating poor crystalline quality of film when Ga droplets form on the surface. **c, d)** XRD and SEM respectively of a sample grown at As / Ga flux ratio of 3.5 and 315C shows much sharper film peak demonstrating higher crystalline quality films occur when Ga droplets do not form on the surface

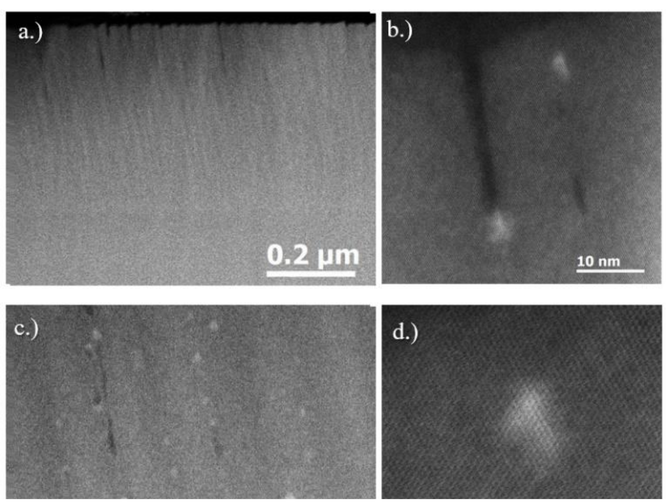

Figure 2S. High angular annular darkfield TEM demonstrates compositional inhomogeneities in a film grown at As/Ga flux ratio of 3.5 and 315C, in a regime without droplets. **a.)** demonstrates lateral composition modulation. **b, c)** show both the lateral compositional modulation and Bi precipitate formation at different length scales. **d.)** shows nanometer scale Bi precipitate formation

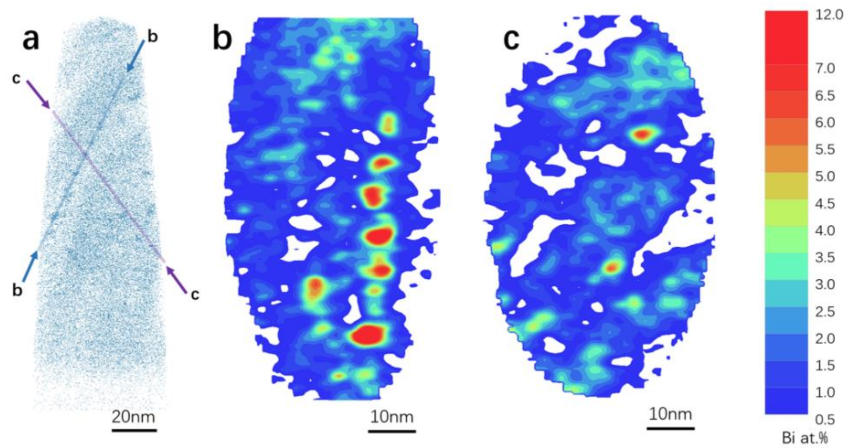

Figure 3S. Atom Probe Tomography showing high Bi content modulations and precipitate formation, from a sample grown at 315C and As / Ga ratio of 3.5, in a regime without droplets

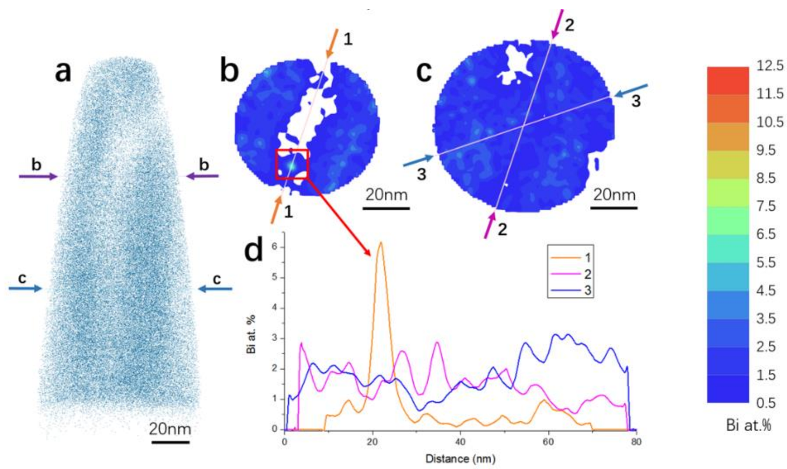

Figure 4S. Atom Probe Tomography of sample grown at 325C, As / Ga flux ratio of 3.2, and Bi incorporation of 2.2% demonstrates smoother regions of film have more uniform incorporation

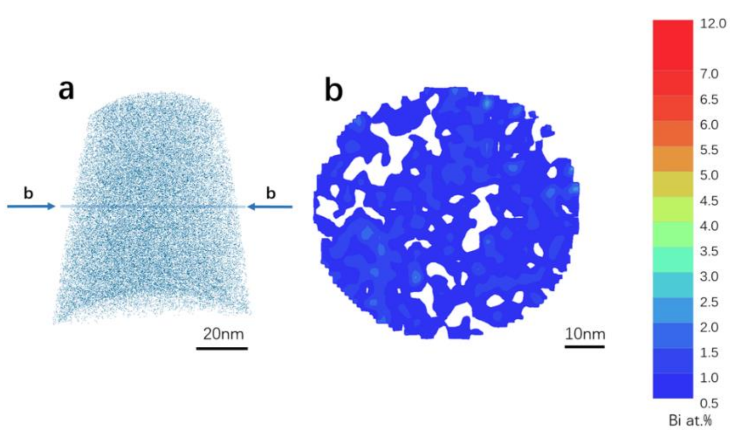

Figure 5S. Atom Probe Tomography of a sample grown at 350C and As /Ga flux ratio of 3.2, with Bi incorporation of 1.1% shows that at higher temperatures compositional inhomogeneities are relieved however Bi incorporation is decreased