

Real-Time Optical Monitoring of the Epitaxial Growth of Zincblende Semiconductors

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Non-invasive optical probes such as reflectance anisotropy spectroscopy (RAS) are attractive for real-time monitoring of the epitaxial growth of cubic semiconductors given their high sensitivity and instrumental simplicity. RAS enhances the surface response against the bulk signal by taking advantage of the reduced symmetry of the near-surface region of cubic crystals. Nevertheless, although the surface specificity of RAS has been known for long time [1], its use for monitoring epitaxial growth had been hampered by the lack of both a rapid spectrometer to follow epitaxial growth and a full understanding of the physics underlining RAS line shapes.

In this talk we demonstrate the power of RAS to elucidate phenomena occurring during the very first stage of the MBE homoepitaxial growth of GaAs (first half monolayer (ML)). We present real-time RAS spectra acquired during growth, taking advantage of a recently developed rapid RAS spectrometer [2]. Typical time-dependent RAS spectra for growth under As-rich conditions (c(4x4) and (2x4) surface reconstructions) are shown in Fig. 1(a). A singular value decomposition (SVD) analysis of these spectra yields two physically independent components, termed B1 and B2 (Fig. 1(b)). Component B1 rises sharply upon starting growth and its amplitude changes sign as the surface reconstruction shifts from c(4x4) to (2x4) (Fig. 1(c)). All this takes place during the growth of the first half ML. Afterwards, the amplitude of B1 oscillates following layer-by-layer growth. In contrast, B2 shows relatively small changes along growth (Fig. 1(c)). Component B1 is associated with surface reconstruction that changes during growth, while the time evolution of B2 suggests that it is characteristic of As-terminated surfaces, regardless of their specific reconstruction.

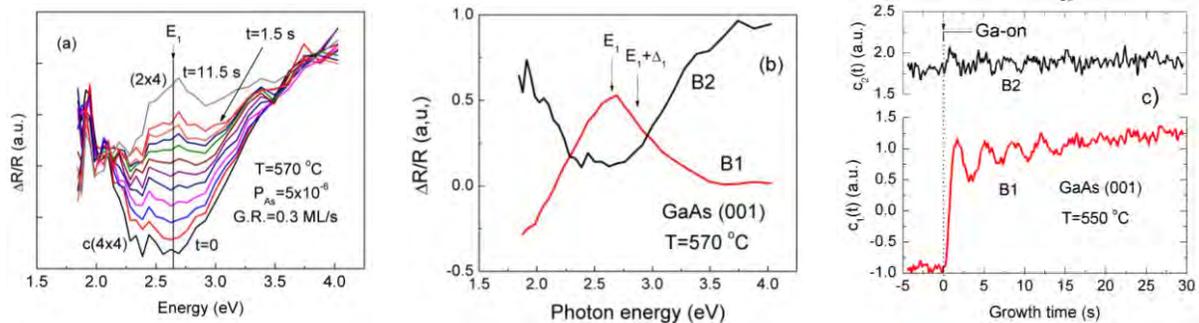


Figure. 1. (a) Time-resolved RAS spectra. (b) B1 and B2 basic spectral components. (c) Time-evolution during growth of the amplitude of B1 and B2 components. B1 amplitude rises sharply during the first half ML growth, while B2 amplitude remains relatively constant.

- [1] D.E. Aspnes, J.P. Harbison, A. Studna, and L.T. Florez, *J. Vac. Sci. Technol.* **A6**, 1327 (1988).
- [2] D Ariza-Flores, J Ortega-Gallegos, O Núñez-Olvera, R E Balderas-Navarro, L F Lastras-Martínez and A Lastras-Martínez, *Meas. Sci. Technol.* **26**, 115901 (2015).

Supplementary information

In terms of its basic, physically independent components the RAS spectrum may be written

$$\frac{\Delta R}{R} = \sum_i c_i(t) B_i(E)$$

where $B_i(E)$ is the i -th spectral component and $c_i(t)$ its corresponding time-dependent coefficient that describes the evolution of its amplitude during epitaxial growth. As pointed out in the abstract page, for epitaxial growth under As-rich conditions (c(4x4) and (2x4) surface reconstructions) the SVD analysis of the time-dependent RAS spectra yields two independent components termed B1 and B2. B1 component is associated to the orthorhombic strain induced by surface reconstruction in a near-surface region. This association is supported by Figure 2 where we show the line shape fitting (red continuous line) to the experimental B1 spectrum using the phenomenological model for strain-induced RAS line shapes reported elsewhere (L.F. Lastras-Martínez et al, Phys Rev. B **70**, 035306 (2004)).

To support our association of component B2 with As-rich surfaces, a series of RAS experiments were conducted with GaAs surfaces under arsenic flux but not gallium flux. We note that by decreasing the flux of gallium while maintaining a constant substrate temperature it is possible to vary the arsenic composition of the surface of GaAs and cause the surface reconstruction to change from c(4x4) to (2x4). In Figure 3 we show the measured RAS spectra varying the arsenic flux. We note that the RAS spectrum evolution mimics the changes observed in the RAS spectrum during epitaxial growth.

In addition, since the substrate is at a relatively high temperature, allowing a Ga flux in addition to the As flux it is possible to obtain a (4x) As-rich surface reconstruction. Figure 3(c) shows the RAS spectra for different Ga fluxes. We note that an SVD analysis of the spectra of Figures 3a and 3c together yields three independent spectral components shown in Figure 3c. B1 and B2 components coincide with those found for real-time RAS spectra. The coefficients c_i corresponding to each of these components are plotted in Figure 3d as a function of As flux. We can see that the amplitude of component B2 is constant to the extent that the surface is As-rich. In contrast, this amplitude decreases when the surface loses arsenic and becomes Ga-rich. This result supports our association of B2 to As-rich surfaces.

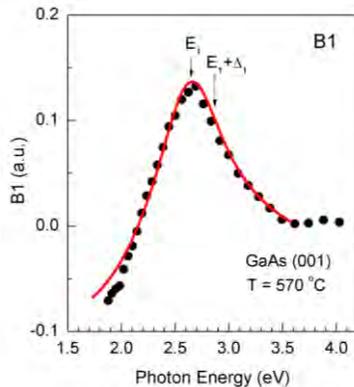


Figure 2. Line shape fitting (red continuous line) of B1 spectra shown in Figure 1(b). Fitting was carried out following the phenomenological model for strain-induced RAS line shapes reported previously. The close fitting of the experimental RAS spectrum supports our conclusion that the B1 component is associated to the orthorhombic strain induced by both c(4x4) and (2x4) surface reconstructions.

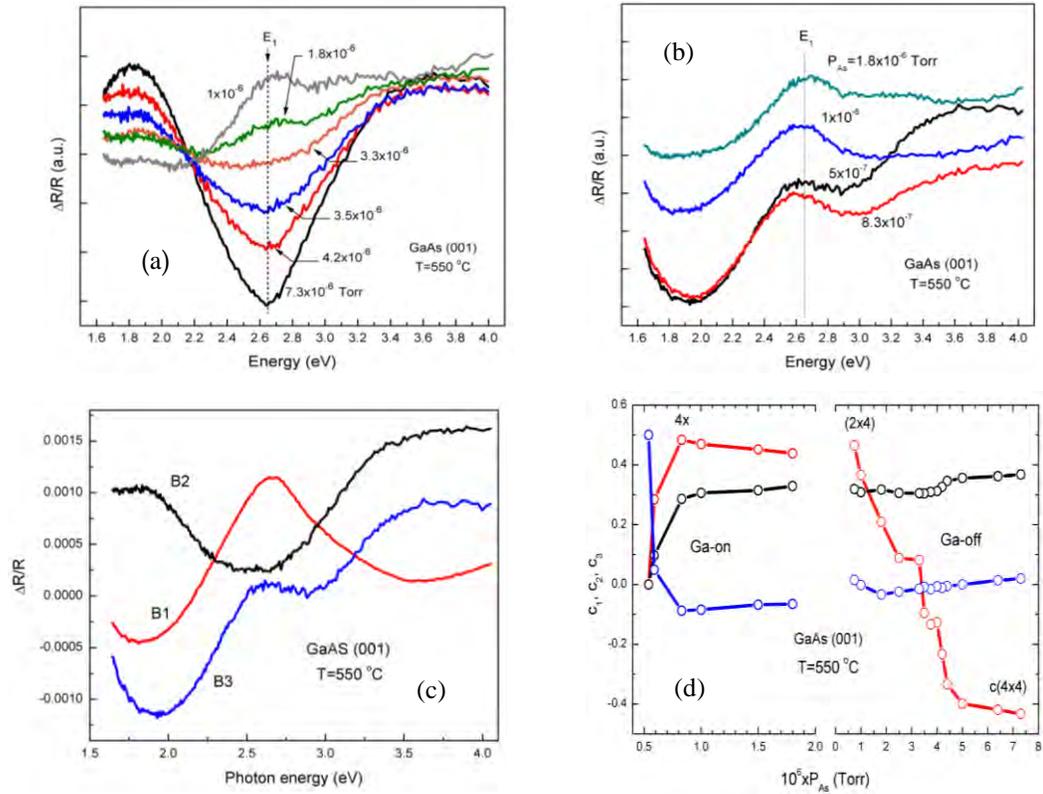


Figure 3. (a) RAS spectra for As-rich GaAs surfaces as a function of As-flux. (b) RAS spectra for Ga-rich surfaces as a function of As-flux. (c) Spectral line shapes for the three independent RAS components B1 (red), B2 (black) and B3 (blue) corresponding to spectra of Figures 3(a) and 3(b). (d) Dependence of c_i coefficients corresponding to B_i components as a function of As-flux, for both As-rich and Ga-rich surfaces. Colors correspond to those of Figure 3(c).