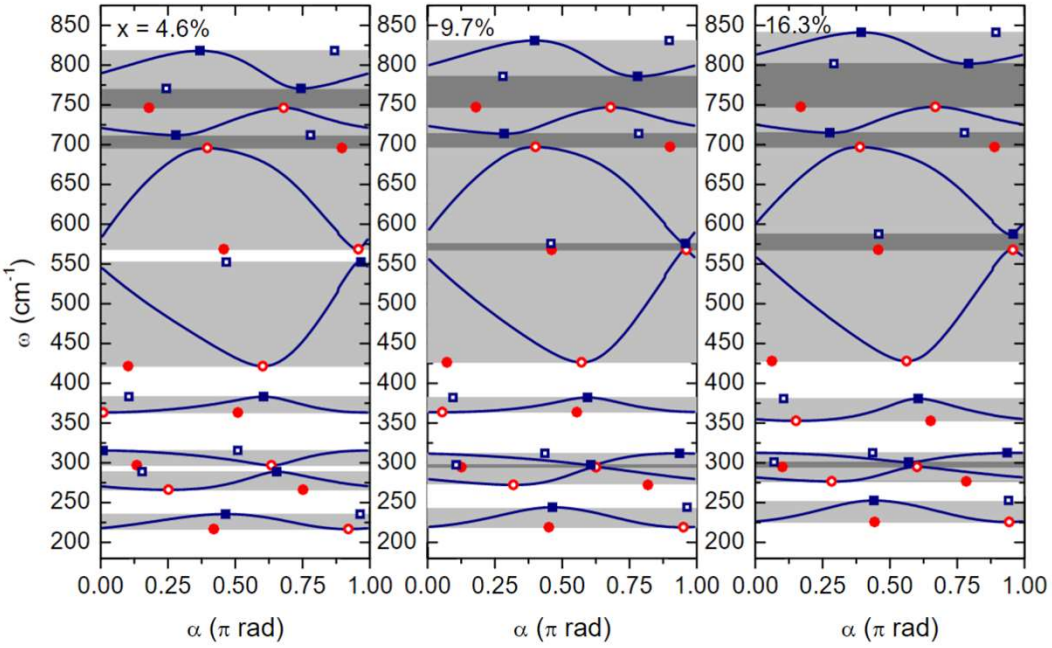

Figure 1: Ellipsometry-derived dispersion of limiting frequencies as a function of unit direction (α= cos(αx)+sin(αy)) in the monoclinic plane for beta phase $(Al_xGa_{1-x})_2O_3$. Symbols indicate frequencies and directions of transition vectors (solid symbols) and their normal directions (open symbols) of TO (circles) and LO (squares) modes. Grey areas indicate the frequency bands of outer (light grey) and inner (dark grey) mode pairs.

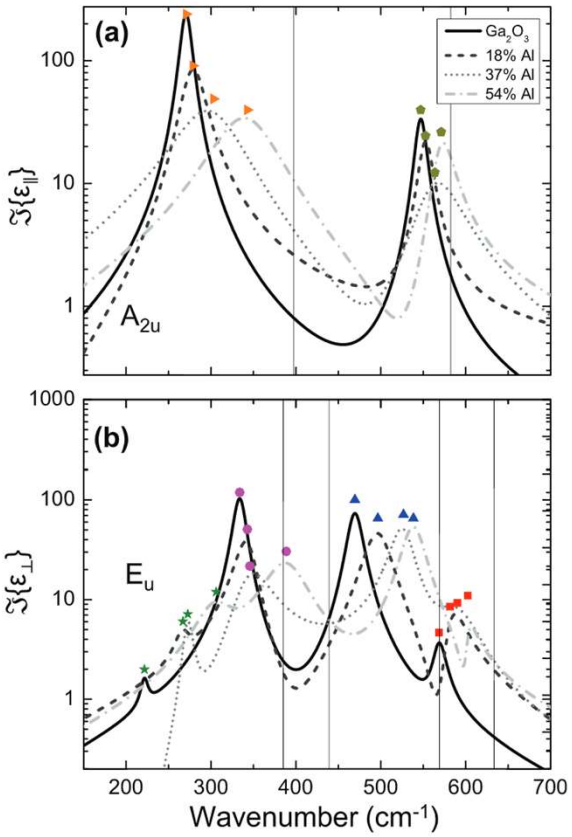

Figure 2: The imaginary part of the dielectric functions for alpha phase alloys studied here. (a) Im($\varepsilon_{||}$) and (b) Im($\varepsilon_{\perp}$). Data for α-Ga_2O_3 are included for comparison. Vertical lines indicate TO frequencies for α-Al_2O_3. Symbols indicate peak positions and thereby TO modes of the ternary alloys.

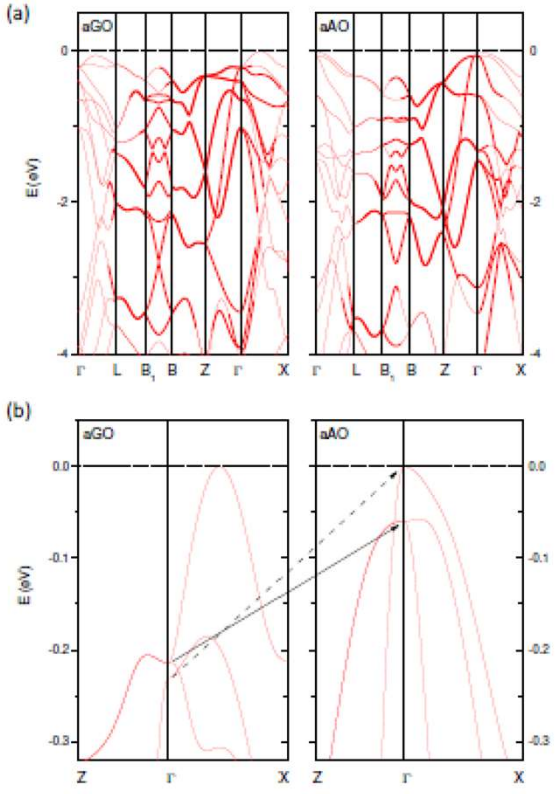

Figure 3: (a) Hybrid-level DFT structure of the valence bands for α-Ga_2O_3 and α-Al_2O_3 (b) an enlarged portion of the top of the valence band around the Γ point. Each plot is arbitrarily set so the energy of the valence band occurs at E = 0. Note that in α-Ga_2O_3 the valence band maximum occurs on the Γ-X line whereas for α-Al_2O_3 it occurs at the Γ point. Arrows in (b) mark the evolution of the sets of bands defining the band edge in the ordinary direction (solid arrow) and extraordinary direction (dashed arrow).