

FIG. 1: Temperature dependent Raman spectra of (001)BFO film on SRO/STO from 22-400°C (a) and 550-1000°C (b). The graphs in (b) were adapted from our earlier work [15]. The beginning of the dashed~arrows pointing up in (b) shows the beginning of the new phase. The existence of phonons in γ -phase in (b) are marked with the solid arrows pointing down. The α -phase extends up to 820°C; (c) and (d) temperature dependence (RT-500°C) of phonon frequencies variation and FWHM for 72 (peak 1), 140 (peak 2), and 171(peak 3) cm⁻¹, respectively.

ruled out. However, the Raman frequencies before and after thermal cycling remain unchanged makes this unlikely. This fact favors films over bulk or powder samples for very high temperature studies in the future. Reaching the tetragonal and cubic phases extrapolated from the powder study of Arnold et al. [2] does not seem impossible with thin films.

B. Phonon anomalies near T_N :

There are discrepancies in the literature regarding both the crystal structure of (001) BFO thin films e.g. with several reports claiming tetragonal [6, 28], rhombohedral [29, 30], and monoclinic [31, 32] structure, and its phonons. Of particular interest regarding phononmagnon coupling in BFO was the report [24] of a very