

## Ba<sub>2</sub>LnTi<sub>2</sub>Nb<sub>3</sub>O<sub>15</sub> (Ln=La, Nd) - a new relaxor ferroelectric system

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### Abstract

A new lead-free relaxor ferroelectric system  $x\text{Ba}_2\text{LaTi}_2\text{Nb}_3\text{O}_{15} - (1-x)\text{Ba}_2\text{NdTi}_2\text{Nb}_3\text{O}_{15}$ , with the tetragonal tungsten bronze structure, has been synthesized. Two end member compositions ( $x = 0, 1$ ) and one intermediate composition ( $x = 0.5$ ) have been prepared by solid state ceramic synthesis. The powders were calcined at  $1300^\circ\text{C}$  for 8 hours and uniaxially pressed pellets were sintered at  $1450^\circ\text{C}$  for 4 hours. Sintered pellet densities were  $\approx 95\%$  theoretical. The powders were checked by XRD and the data sets were fully indexed on a tetragonal tungsten bronze unit cell with no evidence of any secondary phase being present. At room temperature,  $\text{Ba}_2\text{LaTi}_2\text{Nb}_3\text{O}_{15}$  was assigned the centrosymmetric space group  $P4/mbm$  and  $\text{Ba}_2\text{NdTi}_2\text{Nb}_3\text{O}_{15}$  the non centrosymmetric space group  $P4bm$ .  $\text{Ba}_2\text{LaTi}_2\text{Nb}_3\text{O}_{15}$  exhibits a smeared maximum of permittivity at 220 K (at 1 MHz),  $\text{Ba}_2\text{NdTi}_2\text{Nb}_3\text{O}_{15}$  undergoes a first order ferroelectric phase transition near 400 K and  $\text{Ba}_2\text{La}_{0.5}\text{Nd}_{0.5}\text{Ti}_2\text{Nb}_3\text{O}_{15}$  exhibits both ferroelectric phase transition at 270 K and relaxor ferroelectric behaviour at higher temperatures. All three ceramic systems were investigated in the broad frequency (100 Hz - 100 THz) and temperature (10 - 500 K) range. The temperature dependence of the optical phonon modes was studied by means of time domain THz spectroscopy, infrared reflectivity and Raman spectroscopy. A softening of the dielectric relaxation from the THz and microwave regions below 100 Hz was observed on cooling in the relaxor ferroelectric  $\text{Ba}_2\text{LaTi}_2\text{Nb}_3\text{O}_{15}$ , across the whole investigated temperature range. In other two compounds the relaxations soften only in the paraelectric phase down to GHz (in  $\text{Ba}_2\text{NdTi}_2\text{Nb}_3\text{O}_{15}$ ) or down to 100 MHz (in  $\text{Ba}_2\text{La}_{0.5}\text{Nd}_{0.5}\text{Ti}_2\text{Nb}_3\text{O}_{15}$ ) range and below  $T_c$  the relaxations vanish. Origin of the dielectric relaxations is discussed on the basis of structural investigations.