Dumping Soft Modes in Raman Spectra of Antiferroelectric (Pb-La)(Zr-Sn-Ti)O_3 Ceramics

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Since their discovery in 1950s, PZT-type materials have been the most widely used piezoelectric ceramics[1]. With increasing Ti content above $x = 0.06$, the solid solution changes from antiferroelectric to ferroelectric. The six structural phases in the solid solution $Pb(Zr_xTi_{1-x})O_3$ have been observed at ambient pressure are an antiferroelectric phase for compositions near $PbZrO_3$ ($x < 0.06$), ferroelectric low-temperature and high-temperature rhombohedral phases for most Zr-rich alloys, a monoclinic ferroelectric phase near (50% Zr)/(50% Ti) composition, a tetragonal ferroelectric phase for Ti-rich alloys, and a cubic paraelectric phase for all compositions at sufficiently high temperature. Recently it was found by inelastic and diffuse X-ray scattering techniques[2] that in pure lead zirconate phase transition from cubic into antiferroelectric phase is driven by phonon soft mode condensation. Here we report low frequency Raman scattering study of phase transition from cubic into antiferroelectric phase in $(Pb_{0.97}La_{0.02})(Zr_{0.864}Sn_{0.04}Ti_{0.096})O_3$ composition.

Raman spectra have been obtained with Jobin Yvon – Horiba T-64000 triple spectrometer with N2 cooled CCD matrix. Spectra were excited by Ar+ laser (514.5 nm, 500 mW). Data collection time was 600 s for one spectrum, temperature stabilization better than 0.1 K.

The spectrum of higher temperature cubic phase consists of weak and wide bands, as assumed to be in a highly disordered lattice and agrees with previous data[3]. Under cooling a set a new lines appears (Fig. 1); their frequencies show only slight anomalies aside from normal temperature drift, while their widths increase very quickly coming to the transitions point from below.

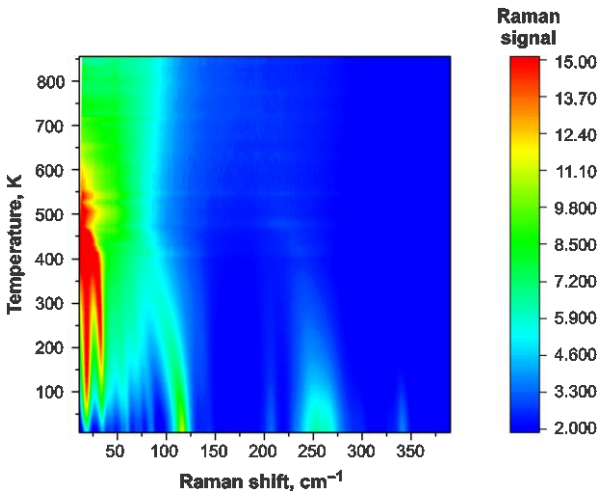

Figure 1. Raman intensity vs. temperature. Softening of the lower frequency modes is clearly seen.

Results are discussed in terms of overdumped soft modes interacting via their dumpings at the phase transition.

References

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