

# Low frequency phonons in rare earths langasites

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Magnetic frustration is found in magnetic material when the individual spin moments cannot simultaneously minimize their magnetic interactions with their neighbored moments. As such, magnetically frustrated systems often support a highly degenerate ground state exhibiting a number of exotic physical phenomena such as spin ice [1] and spin liquid [2] states. Probing the spin and lattice dynamics in these systems provides an avenue to better understand the nature of these exotic states.

Rare-earth langasites are characterized by geometric magnetic frustration, exhibiting magneto-electric effects, high piezoelectric properties, and are seen as a possible candidate for a spin-liquid ground state [3]. Phonon and crystal electric field spectra provide important information to unravel the interplay between the structural and magnetic properties of the langasite family. The langasite structure crystallizes in the P321 space group with a general formula  $A_3BC_3D_2O_{14}$ , where magnetic rare-earth elements are situated at the A site of the structure.

Our study presents spectra of several rare-earth langasites  $RE_3Ga_5SiO_{14}$  ( $RE = La, Nd, Ho$ ) using Fourier-transform infrared (FTIR) reflection spectroscopy. Experiments have been performed with polarized radiation along the principal crystallographic axes and under different sample temperatures. Phonon excitations at unusually low frequencies are observed that brings the crystal structure of langasites close to a lattice instability. The results in the rare-earth langasite  $Nd_3Ga_5SiO_{14}$  (NGS) and in a holmium-substituted langasite  $Ho_{0.03}La_{2.97}Ga_5SiO_{14}$  (Ho-LGS) are compared with pure  $La_3Ga_5SiO_{14}$  (LGS) langasite compound that does not show magnetic frustration.

Spectra with polarization parallel to the  $b^*$ -axis of the crystal show a number of phonons between 100 and  $400\text{ cm}^{-1}$  with a weak temperature dependence indicating a stable crystalline structure. This behavior was found in all three investigated samples. Although they have the same crystalline structure, differences in the obtained dielectric function can be observed especially at low frequencies.

Phonon spectra in  $c$ -direction are dominated by a large excitation at unusually low frequencies (less than  $50\text{ cm}^{-1}$ ). In contrast to the other phonons modes, strong temperature effects are observed for the low frequency mode. For NGS the lowest phonon softens from  $50\text{ cm}^{-1}$  at room temperature to  $20\text{ cm}^{-1}$  at 10K that can be correlated with high static dielectric permittivity. This effect can be seen on the other crystals as well, but the effects are weaker. All phonons above a frequency of  $100\text{ cm}^{-1}$  behave like those of the  $b^*$ -axis, no substantial shift of the frequency can be detected.

Because of the observed phonon softening, an instability in the structure of the langasites along the  $c$ -direction can be expected. The intriguing prospect of how this abnormal phonon may affect the magnetic dynamics in the frustrated compounds is explored.

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## Refs

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