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Ultrasound Studies of Thermoelectric Materials

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ABSTRACT

Motivated by the search for improved thermoelectric materials, several compounds have attracted attention that combine the high electron mobilities found in crystals with a low thermal conductivity κ, approaching κ-values typical for glasses. The common structural feature of these "electron crystal phonon glass" (ECPG) materials is that they contain loosely bound atoms that reside in a large crystalline "cage". These "rattlers" scatter phonons and greatly reduce the thermal conductivity of the material, but generally not to quite glasslike levels. A particular class of ECPGs is formed by the Ge-clathrates, with $Sr_8Ga_{16}Ge_{30}$ and $Eu_8Ga_{16}Ge_{30}$ having a truly glasslike thermal conductivity while maintaining the crystalline electronic properties.

Here we report Resonant Ultrasound Spectroscopy measurements for filled and unfilled skutterudites and for Ge-clathrates. The presence of the rattler significantly softens the elastic behavior of all measured materials, but its influence is most dramatic in the case of $Eu_8Ga_{16}Ge_{30}$, where a remarkable "dip" is observed in the shear modulus c_{44}. This response, combined with results from low-temperature ultrasonic attenuation, neutron-scattering, thermal conductivity and microwave absorption measurements, provides clear evidence for the existence of a new type of four-well tunneling states, closely linked with the fourfold split positions of Eu known from neutron diffraction density profiles.