A NUCLEAR INELASTIC SCATTERING STUDY OF THE DYNAMICS IN FILLED SKUTTERUDITES AND CLATHRATES

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The figure of merit of a thermoelectric material is inversely proportional to its thermal conductivity. Hence, the reduction of the lattice thermal conductivity is important and has been achieved through the inclusion of "rattlers" within the cages of various materials, specifically filled skutterudites and clathrates. An in-depth knowledge of the dynamics of these rattlers is thus required in order to improve the thermoelectric properties of materials. Among the various techniques, that can be used to study lattice dynamics, nuclear inelastic scattering has recently emerged as a feasible atom specific technique, because of both the increase in brilliance of synchrotron radiation and the development of high-energy monochromators. The recoil-free γ-ray resonance has been used for 50 years in Mössbauer spectroscopy to study various solid properties but the γ-ray absorption with exchange of energy with the lattice, i.e., the nuclear inelastic scattering, is also a very informative and currently available technique for several Mössbauer nuclides, a technique that provides an in-depth, atom specific, view of the lattice dynamics of a solid.

Fortunately, iron, europium, and antimony are Mössbauer active elements that are found in filled skutterudites and clathrates. First, we will show how the iron-57, europium-151, and antimony-121 nuclear inelastic scattering measurements[1,2] have provided a complete picture of the lattice dynamics in $EuFe_4Sb_{12}$. Because of the atom specific character of the technique, the contributions of the these three elements have been separated. The rattling europium contributes to the low energy vibrational states, at ca. 7 meV and show virtually no overlap with the iron vibrational states located between 27 and 35 meV. The absence of coupling between the europium and iron vibrations could not explain the low lattice thermal conductivity of $EuFe_4Sb_{12}$. The antimony-121 nuclear inelastic scattering measurements reveal the coupling between the rattler and the host framework. Upon filling of $CoSb_3$ there is a transfer of 10% of the vibrational states toward lower energy and an increase in the population of the vibrational states at ca. 7 meV. Second, we will show how a combination of neutron and nuclear inelastic scattering and Mössbauer spectral measurements[3,4,5] provide an excellent description of the dynamics of the caged guests in filled germanium clathrates and the first experimental observation of atomic tunneling.

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