Experimental report

09/02/2017

Proposal: 7-01-427 **Council:** 4/2015

Title: Study of lattice dynamics and anharmonicity of Al6Ge5 with same structure as free defect Zn4Sb3

Research area: Materials

This proposal is a new proposal

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Samples: Al6Ge5

Instrument	Requested days	Allocated days	From	To
IN6	3	3	18/06/2015	19/06/2015
			23/06/2015	25/06/2015

Abstract:

We propose an experimental study of the vibrational dynamics of Al6Ge5 (same structure as free defect Zn4Sb3) by neutron time-of-flight spectroscopy. Our interest is the establishment of the details in the low-energy inelastic responses supposed to be responsible for the reduced thermal conductivity of this compound. The effect of temperature on the mode frequencies is of importance in order to study the anharmonicity of the vibrations modes. This work will also permit to improve our understanding of the mechanisms behind the low thermal conductivity of Zn4Sb3 and notably the importance of anharmonicity vs defects. Our experimental efforts are guided and supported by ab-initio lattice dynamics calculations.

Al_6Ge_5 structure is isomorph to the ideal structure of Zn_4Sb_3 [1] which has a very high ZT at high T (ZT = 1.3 at 670 K) due to its low $k_{lattice}$ (about 0.3 W/m.K at 670 K) and might be one of the most promising thermoelectric compounds currently investigated [2]. Actually, there are still some controversies about the origin of the low thermal conductivity of Zn_4Sb_3. Different scenarios have been proposed: the scattering from different kinds of point defects [3-5] and to the coupling to low-energy dumbbell modes at about 5 meV [6], the large anharmonicity of the vibrations of Sb atoms coordinated with a prism of 6 neighboring atoms [7]. As Al_6Ge_5 has much less defects and lighter atoms than Zn_4Sb_3, but similar thermal conductivity, it is a very suitable system not only for testing the different scenarios proposed for explaining the low thermal conductivity of Zn_4Sb_3 but also merits to be studied by itself. Experimental conditions for the synthesis of Al_6Ge_5 with the highest purity reported so far in our previous article [1] has shown that Al and Ge remain as secondary phase.

The measurements were performed on IN6 with high-resolution option to monitor vibrational properties of powdered Al_6Ge_5 sample from 50 to 400K and a wavelength of 4.14Å and 5.12Å. The features observed in the Generalized Density Of States (GDOS) at 100K (figure 1) are in accordance with the calculation, there is no feature from Al and Ge secondary phases. We observed a low energy mode at about 8.5 meV mainly due to Ge atom vibrations, which is predicted by the calculations. When decreasing the temperature down to 50K, we did not succeed to obtain measurements with low noise whereas by increasing the temperature all the features are smearing out and at 200K low energy modes are almost not visible anymore. At room temperature it is almost impossible to see the features. This modification of the GDOS could be due to broadening related to the large anharmonicity of the lattice vibration of Al_6Ge_5. It can be also due to a large Debye Waller factor, which would decrease also the intensity. However, no shifting are clearly observed between 100 and 150K, further investigation are needed for the 200K case has possible shifts are observed for the different modes.

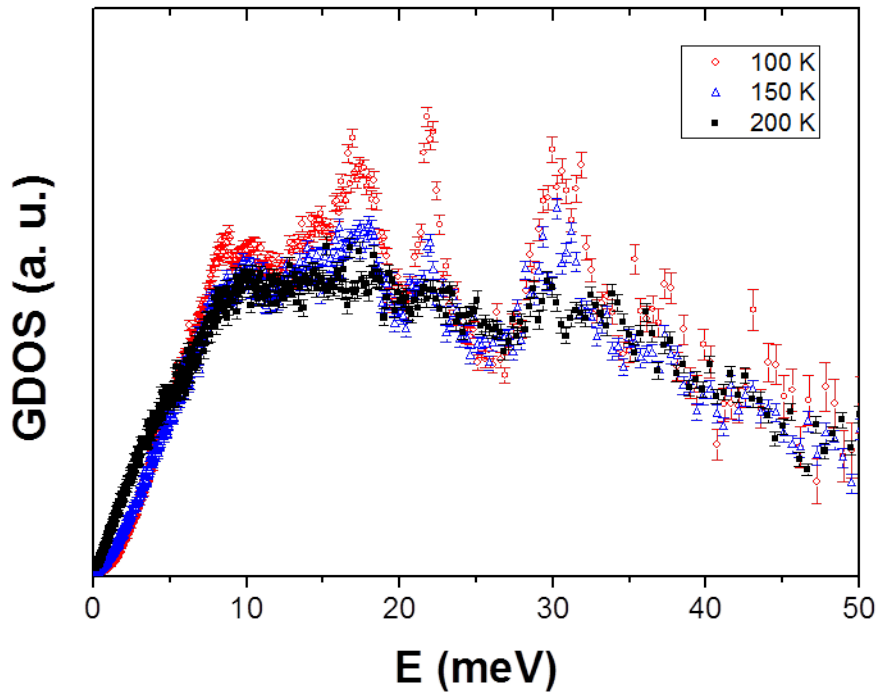

Figure 1: Experimental GDOS of Al_6Ge_5 at different temperature

References

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