Experimental report

24/02/2016

Proposal: TEST-2556 **Council:** 4/2015

Title: Thermoelectric clathrate BaZnGe

Research area:

This proposal is a new proposal

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Samples: Ba*Zn6Ge40

Instrument	Requested days	Allocated days	From	To
IN5	2	2	24/11/2015	26/11/2015

Abstract:

This project is intended to establish the phonon dynamics in the compound BaZn6Ge40 of type I clathrate structure (space group Pm-3n). The present test experiment is the preparation for a continuation of spectroscopic measurements carried out on powder samples of different clathartes. Prior studyies have been published in the following papers:
Melnychenko-Koblyuk et al., J. Phys.: Condens. Matter 19, 216223 (2007)
Melnyczenko-Koblyuk et al., Phys. Rev. B 76, 144118 (2007)
Koza et al., Phys. Rev. B 82, 214301 (2010)

Clathrates are representatives of crystals formed by a fully bonded network of open-spaced polyhedra. Their structural complexity and phonon properties are conjectured to be the reason for an appreciably reduced thermal conductivity which is in the focus of our study. Detailed tudies of clathrates are made difficult for the difficult formation of single crystals. Recently our colleagues from the university of Vienna (Peter Rogl and coworkers) succeeded in growing single crystals of the nominal composition Ba8Zn6Ge40 large enough for precise measurements by inelastic neutron scattering. The goal of the project is the determination of (i) the low-energy dispersion of phonons, (ii) of the life-time of these phonons, and (iii) of their Gruneisenparameters. These would enable us to test recent models of the thermal conductivity in open-spaced compounds.

We have tested two crystals for their orientation and quality at the instrument OrientExpress. An example of a Laue pattern is shown in Fig. 1. Both crystals were confirmed to be of pure single-crystal quality. In total 105 Laue patterns at different orientations, sample to camera distances and collimations were recorded. The larger single crystal was mounted on a goniometer and prealigned to be tested for phonon intensities at the ToF spectrometer IN5.

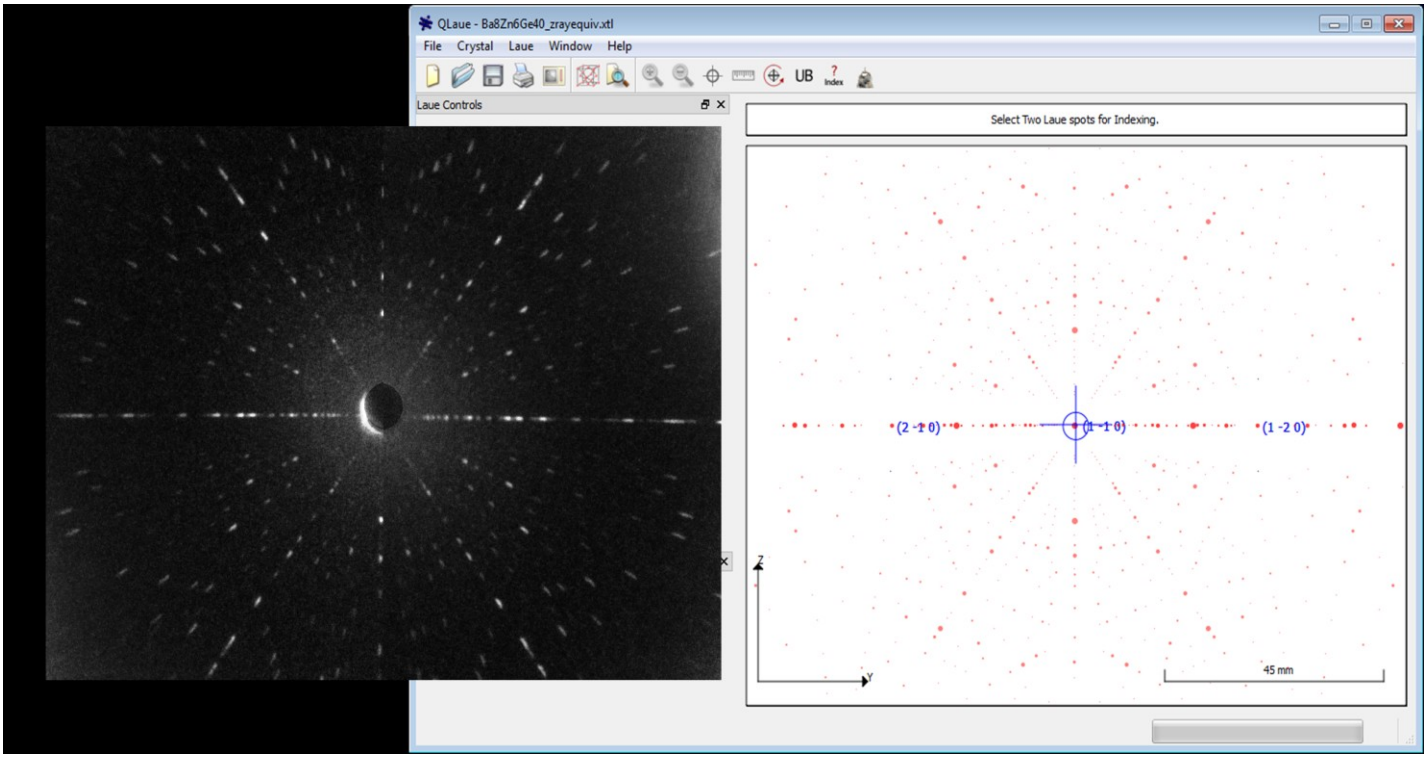

Figure 1: Left, example of one Laue pattern of a Ba8Zn6Ge40 crystal recorded at OrientExpress. Right, Laue pattern calculated for an equivalently scattering compound.

The IN5 test was carried out at room temperature with a wavelength of 3.2A. The orientation of the crystal was chosen according to our ab initio lattice dynamics calculations which indicated the strongest intensity for acoustic phonons around the Bragg peak (060). The phonon dispersions based on these calculations are reported in Koza et al., Phys. Rev. B 82, 214301 (2010). A prescan of 80 degrees with a step of 2 degrees and a final scan of 45 degrees with 1 degree step were conducted. The acquisition time varied between 10 and 20 minutes.

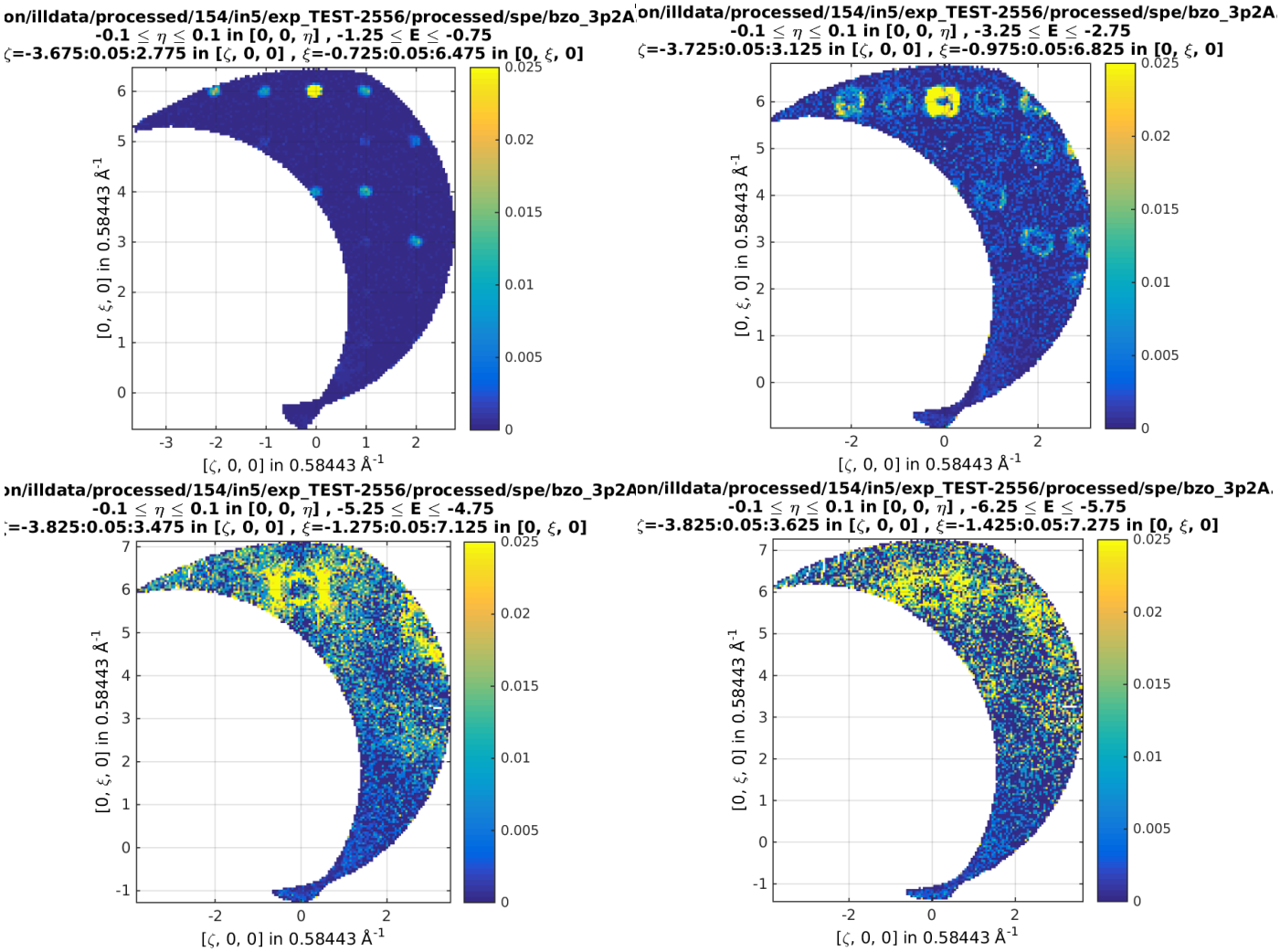

Figure 2: Constant energy slices through the xx0 plane. Energies correspond to 1, 3, 5, and 6 meV on the energy gain side of neutrons.

Fig. 2 shows constant energy slices through the xx0 plane. At energies of 1 and 3 meV the dominant intensity of acoustic phonons around the Bragg peak (060) is obvious. At higher energies the pronounced localization of acoustic branches towards the zone boundaries and additional low-energy optic modes give rise to the diffuse signal.

The test confirmed the excellent quality of the cystal, the reliability of the ab initio lattice dynamics calculations and the feasibility of deriving the essentials of the phonon dispersion from an IN5 experiment on the Ba8Zn6Ge40 crystal. The test showed as well that for a detailed qualitative analysis of the phonon spectra a mapping with a step of 0.5 and a wider scanning range would be highly appreciable.