ESRF	**Experiment title:** Lattice dynamics of $FeSi_2$ nanostructures by nuclear inelastic scattering and first principles theory	**Experiment number**: MA 4437
Beamline: ID18	**Date of experiment**: From 05/09/18 to 09/09/18	**Date of report**: 13/09/21
Shifts: 11	Local contact(s) Mirko Mikolasek	

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The aim of this experiment was a systematic study of the lattice dynamics and thereby the vibrational thermodynamics of epitaxial metallic α-phase and semiconducting β-phase $FeSi_2$ nanostructures by in situ nuclear inelastic scattering on ^{57}Fe and first-principles theory.

Report

The main aim of the experiment was a systematic study of the lattice dynamics of nanostructures of the metallic α-$FeSi_2$ phase and the semiconducting β-$FeSi_2$ phase. For this purpose, α-$FeSi_2$ islands and nanowires as well as β-$FeSi_2$ nanorods were prepared via reactive deposition epitaxy in our home lab at KIT, Karlsruhe. The crystal structure and the surface morphology of the nanostructures were characterized *in situ* via RHEED, XPS and AFM studies. Subsequently the $FeSi_2$ nanostructures were capped with 4 nm Si to prevent oxidation. The local atomic structure was determined by *ex situ* EXAFS measurements performed at KARA, KIT.

Figure 1 depicts the obtained results for the α-$FeSi_2$ (a) nanoislands measured along two orthogonal directions and nanowires measured (b) along (direction 1) and (c) across (direction 2) the nanowires orientation. The islands PDOS consists of three peaks with a sharp main peak located at 20 meV and two broader peaks located at 33 and 45 meV. Around 24 meV a small shoulder is visible and around 62 meV a minor high energy peak is present. No dependence of the shape of the PDOS on the direction of measurement is observed. For the nanowires the spectrum measured along direction 1 exhibits peaks at the same positions with different relative intensities, including a small peak at 24 meV. The peaks at 33 and 45 meV are the most pronounced ones, whereas the peak height at 20 meV is significantly reduced compared to the islands. In the PDOS measured along direction 2 the peak at 20 meV is missing. The intensity of the peaks at 24 and 33 meV is slightly increased whereas at 45 meV it is the same as for direction 1.
In order to attain a comprehensive understanding of the obtained results, first principles calculations of the *xy*- and *z*-polarized PDOS of the tetragonal α-$FeSi_2$ were performed and compared with the experimental data. For the nanoislands the experimental spectra can be explained by a superposition of the *xy*- and *z*-polarized PDOS [1]. The nanowires PDOS measured along direction 1 also consists of a superposition of the *xy*- and *z*-polarized PDOS with a significantly lower contribution of the *z*-polarized phonons. For the spectrum measured along direction 2, on the other hand, the experimental PDOS can be modelled by only taking into account the xy- polarized vibrational states [2].

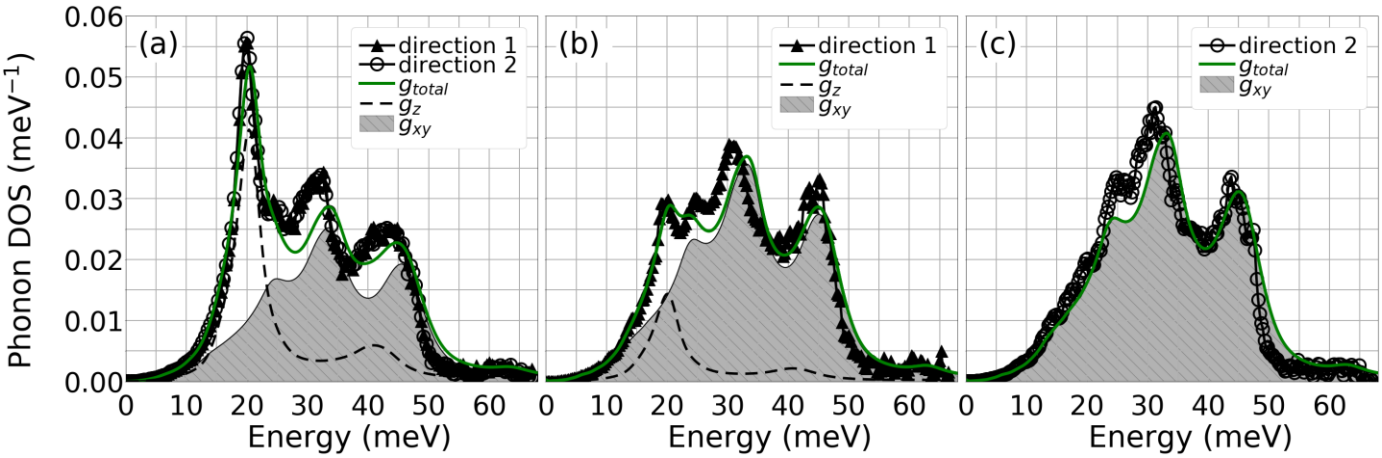

Figure 1. *The Fe-partial phonon DOS of α-phase $FeSi_2$ of (a) nanoislands measured along two orthogonal directions, (b) nanowires measured along the structure, and (c) nanowires measured across the structure. The ab initio calculated PDOS of α-phase $FeSi_2$ in xy-projection (g_{xy}, grey/hatched) and z-projection (g_z, dashed line) are compared with the experimental data.*

In Figure 2 the PDOS of bulk β-phase $FeSi_2$ (determined in the previous experiment MA-3725) is compared to the results obtained for β-$FeSi_2$ nanorods with average widths and lengths of 52 nm and 167 nm (S1), and 40 nm and 129 nm (S2) measured along (direction 1) and across (direction 2) the rods. The bulk PDOS consists of two major peaks at 26 and 36 meV and a minor peak at 43 meV. No vibrational anisotropy is observed for the spectra measured along the two orthogonal directions. Furthermore, a good agreement with the *ab initio* calculated PDOS of β-$FeSi_2$ is observed. For the nanorods of S1 and S2 the maxima at 36 meV and 43 meV are still present with a slight increase of intensity at 43 meV compared to the bulk sample. In the low energy part of the spectrum a shift of intensity from the peak at 26 meV to new vibrational states around 20meV is observed. This effect is more pronounced in the spectra measured along direction 1, in particular in case of S1 [3].

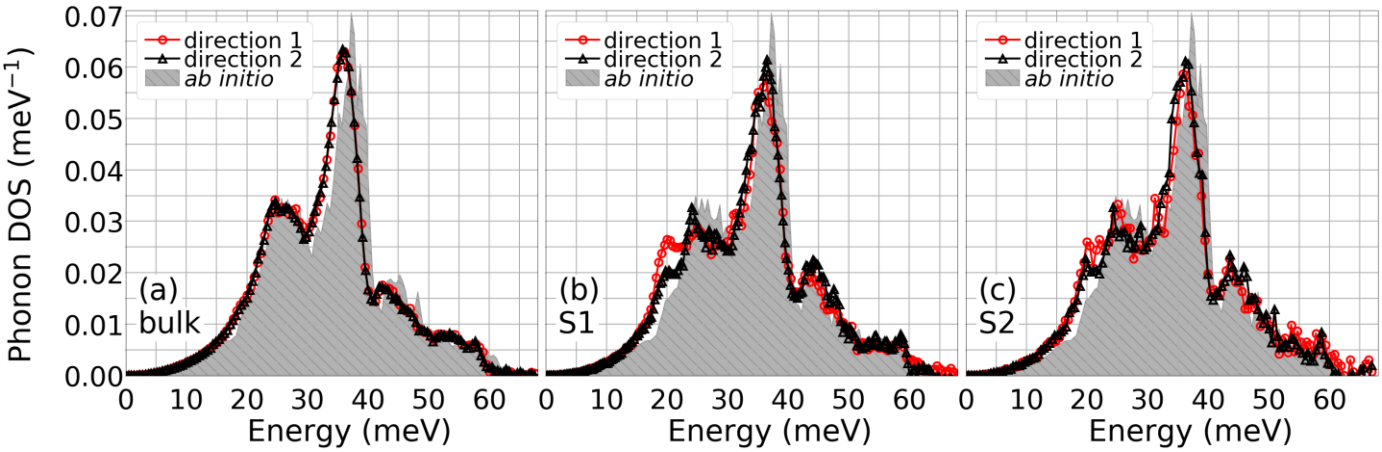

Figure 2. *The Fe-partial PDOS of β-phase $FeSi_2$ of (a) bulk film measured along two orthogonal directions, (b) S1 and (c) S2 measured along (direction 1) and across (direction 2) the nanorods. The grey/hatched spectrum denotes the ab intio calculated PDOS of β-phase $FeSi_2$.*

References

[1] Kalt et al., Phys. Rev. B 101, 165406 (2020)
[2] Kalt et al., Phys. Rev. B 102, 195414 (2020)
[3] Kalt al., to be submitted (2021)