Experimental report

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Proposal: **7-01-439** **Council:** 4/2016

Title: Spin-phonon coupling in FeSi

Research area: Physics

This proposal is a new proposal

Main proposer: **Matthias MOERTTER**

Experimental team: Dominic BOLL
Frank WEBER

Local contacts: Alexandre IVANOV

Samples: FeSi

Instrument	Requested days	Allocated days	From	To
IN8	8	7	15/12/2016	22/12/2016

Abstract:

We recently reported evidence that FeSi exhibits an intrguing coupling between its magnetic and lattice degrees of freedom using inelastic neutron scattering performed at LLB, ILL and MLZ (Krannich et al., Nat. Commun. 2015)). Here we propose to investigate the behavior of phonon modes in FeSi featuring such a strong spin-phonon coupling under an applied magnetic field up to B = 10 T. The magnetic field will strenghten the (already in zero field present) magnetic fluctuations leading to a ferromagnetic-like state, for which spin-phonon coupling was calculated to be very sensitive with regard to the energies of magnetic excitations. If the scenario of a direct coupling between the magnetic and lattice degrees of freedom is correct, we expect a magnetic field dependence of the phonon life time and its linewidth in energy if we tune the magnetic properties with the field. This would be direct evidence of spin-phonon coupling and not biased by other possible, e.g., temperature dependences.

EXPERIMENTAL REPORT

INSTRUMENT IN8 – thermal triple-axis spectrometer	DATES OF THE EXPERIMENT 15. - 22.12.2016	NUMBER OF THE EXPERIMENT 7-01-439
TITLE OF THE EXPERIMENT Spin-Phonon coupling in FeSi		
AUTHORS AND THEIR LABORATORY (full address) Dominic Boll, Frank Weber *Institute for Solid State Physics, Karlsruhe Institute of Technology, Karlsruhe, Germany*		
Local contact Alexandre Ivanov		

Here, we report on our investigation of the lattice dynamics in FeSi in high magnetic fields up to $B=10\,\mathrm{T}$. Former investigations on this material have shown that the lattice degrees of freedom are linked with the evolution of the magnetic properties [1]. For low temperatures, FeSi is a semiconductor with a small bandgap of approximately $60-70\,\mathrm{meV}$ and has a non-magnetic ground state. A cross-over to a paramagnetic metal occurs between $T=150\,\mathrm{K}$ and $200\,\mathrm{K}$ far below the expected temperature range with respect to the bandgap of $60\,\mathrm{meV}$. The temperature-induced magnetism in FeSi involves a strong phonon renormalization which effects the phonon energy, linewidth and intensity of specific modes. The strongest changes have been observed for two longitudinal phonon modes at the R point and one at the zone center, i.e., the zone boundary along the [111] direction in reciprocal space at energy transfers of about $24\,\mathrm{meV}$ and $34\,\mathrm{meV}$. The phonon patterns of these modes involve strongly varying Fe-Fe distances. Additionally, signatures of short-range ferromagnetic correlations in FeSi via paramagnetic scattering were observed and indeed, closely related materials such as MnSi and $\mathrm{Fe_{1-x}Co_xSi}$ show helical magnetic order between neighbouring Fe moments.

We used the thermal triple axis spectrometer IN8 at the Institute Laue Langevin (ILL) with a strong magnet to study the phonon properties of specific modes in a temperature range between $T=2\,\mathrm{K}$ and $300\,\mathrm{K}$ and in magnetic fields up to $B=10\,\mathrm{T}$. We did energy scans at the wave vectors from to $Q=(2,2,0.5)$, at $Q=(1.5,1.5,1.5)$ and $Q=(1.75,1.75,2)$. These Q points were used because the associated phonon modes were known to show a strong renormalization in zero field.

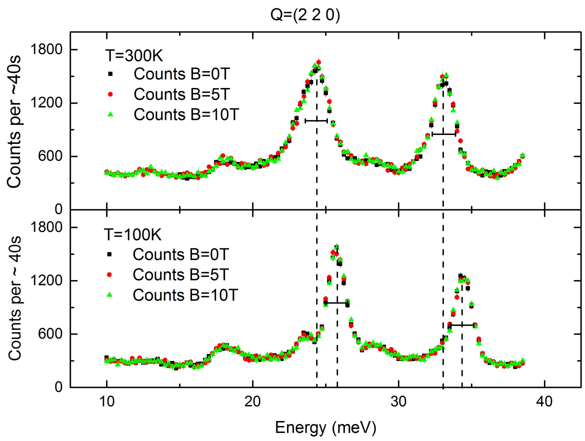

Figure 1: Inelastic neutron scattering data of FeSi for $Q=(2,2,0)$ at $T=100\,\mathrm{K}$ (bottom) and $T=300\,\mathrm{K}$ (top) for different magnetic field strengths. The vertical dashed lines mark the position of the phonon energy; the horizontal error bars indicates the linewidths of the low-temperature phonon modes.

Typical scans for $Q=(2,2,0)$ at $T=100\,\mathrm{K}$ and $T=300\,\mathrm{K}$ are shown in Figure 1. For all energy scans we used Cu(200) crystals both as monochromator and analyzer in order to achieve the best energy resolution possible and measured at a fixed final momentum of $k_f=2.662\,\mathring{A}$. From our previous measurements we knew that the phonon profiles observed at $T=100\,\mathrm{K}$ in zero field do not yet show any broadening and, hence, can be used to define the experimental resolution. Therefore, we fitted Gaussians to the phonon peaks observed at $T=100\,\mathrm{K}$ (zero field) and used the determined linewidth of the Gaussian in fitting a Voigt profile for the high-temperature data where the broadened intrinsic phonon lineshape is represented by Lorentzian. In the setup with Cu(200) monochromator and analyzer we achieved an energy resolution down to $1.35\,\mathrm{meV}$ near 25 meV in energy transfer. This is a huge improvement to previous measurements using PG(002) for monochromator and analyzer with a resolution of about 3meV for the same phonon modes.

As a first result we can conclude that there are no field-induced changes of the phonon properties at the investigated wavevectors in an applied magnetic field up to $B=10\,\mathrm{T}$ heating up to room temperature (see Figure 1). This result was verified after 4 days of beam time. Therefore, we used the remaining time for a different scientific question. Our new goal was to study the momentum dependence of the phonon renormalization in zero field.

Figure 2 shows the momentum dependence of the relative energy difference of the two phonon modes at $Q=(2,2,l)$ and $l=0-0.5$ between $T=2\,\mathrm{K}$ and $T=300\,\mathrm{K}$. For both phonon modes a strong softening up to 7% is observed in this temperature range. This is approximately seven times stronger than a normal softening expected due to thermal expansion. This observation confirms our former investigation on the phonon energies in FeSi.

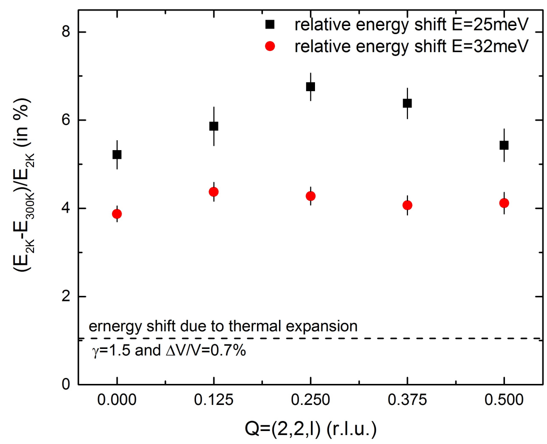

Figure 2: Momentum dependence of the relative energy difference in FeSi for the two phonon modes at $Q=(2,2,l)$ *.*

In Figure 3, the temperature dependence of the linewidth for the lower-energy phonon mode at $Q=(2,2,l)$ is shown. Here, we show only the values up to $l=0.25$ because of some artificial peaks in the spectra which prevented us from determining the linewidth of the other phonon modes accurately. In general, we found a very pronounced background in the spectra, as it can be seen in Figure 1. Hence, we decided to perform some empty-can scans to investigate the background. Although the empty-can scans did show some of the structures, we could not assign all background variation to scattering from the magnet walls and/or the sample holder.

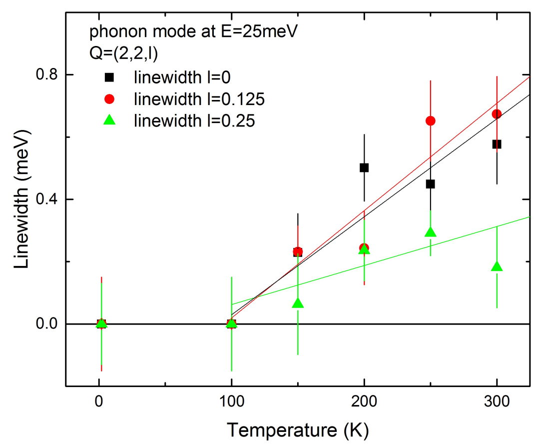

Figure 3: Temperature dependence of the intrinsic linewidth for the low-energy phonon mode at $Q=(2,2,l)$ *. The solid lines represent a linear fit in order to guide the eye.*

As a consequence, we were not able to determine the linewidths of the phonon modes near the zone boundary. Nevertheless, we observe a broadening of the phonon peaks closer to the zone center starting at approximately $T = 150\,\mathrm{K}$. This was the first time we could see this onset with a normal triple axis spectrometer due to the very good energy resolution of IN8. Previously, this was only observed in TAS measurements using the neutron-resonant spin-echo technique. Obviously, the normal TAS measurements are much faster and, hence, preferable.

We confirmed our former observation on the phonon renormalization and broadening of the phonon mode at the zone center [1]. Our new results show a strong softening of the energies and a broadening of the linewidth for all phonon modes from $Q = (2,2,0)$ to $Q = (2,2,0.25)$. These results for the behavior at finite l values will be used to benchmark calculations predicting momentum-dependent electron-phonon coupling. From this we will learn whether the large linewidth in FeSi can be qualitatively understood by electron-phonon coupling effects, which are only activated if magnetic moments are present at elevated temperatures. Another possibility would be a direct spin-phonon coupling which in general might have a different momentum dependence. which results in a strong softening of the energies and a broadening of the linewidth for all phonon modes from $Q = (2,2,0)$ to $Q = (2,2,0.25)$.

In summary, our inelastic neutron scattering measurements on IN8 at temperatures between $T = 2\,\mathrm{K}$ and $300\,\mathrm{K}$ and magnetic fields up to $B = 10\,\mathrm{T}$ have revealed no detectable changes in the phonon properties depending on the magnetic field. On the other hand, we could use the very good energy resolution of the setup of about $1.35\,\mathrm{meV}$ in order to investigate the momentum dependence of the linewidth of the phonon modes from $Q = (2,2,0)$ to $Q = (2,2,0.25)$. Here, we found a broadening starting at $T = 150\,\mathrm{K}$ which appear stronger near the center of the Brillouin zone. To confirm our results theoretical analysis have to be done to check whether there are no magnetic field effects or if the magnetic field used in this experiment is not sufficiently strong to cause changes in the phonon properties.

[1] Krannich, S. *et al.* Magnetic moments induce strong phonon renormalization in FeSi. *Nat. Commun*. 6:8961 doi: 10.1038/ncomms9961 (2015).