

Vibrational Density of States of Fe Thin Films

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Introduction

Low dimensional structures like thin films very often exhibit properties that differ considerably from those of corresponding bulk materials. Special deposition techniques allow preparation of materials that exhibit unique electronic, magnetic and vibrational properties. The latter are often influenced by the layer structure, especially in the case of propagating excitations like phonons. Due to the broken translational symmetry at boundaries of thin films and multilayers, changes in the vibrational spectrum are expected if the film thickness or multilayer period is comparable to the phonon mean free path. Size effects in the vibrational properties are of particular technical relevance if, for example, the thermal conductivity of thin films is considered, which can be an important issue for the thermal behavior of micromechanic and micro-electronic devices.

In this experiment we have determined the vibrational density of states (VDOS) of ⁵⁷Fe thin films via inelastic nuclear resonant scattering of synchrotron radiation. This method was introduced recently [1,2]. It relies on detection of time-delayed fluorescence photons emitted by decaying Mössbauer nuclei in the sample that were excited by synchrotron radiation pulses. If the energy of the incident radiation is off resonance, excitation of nuclei may be assisted by creation or annihilation of phonons in the sample. Therefore, the energy dependence of the yield of nuclear decay products (conversion electrons or subsequent K-fluorescence photons) gives a direct measure of the phonon density of states in the sample [2]. Due to the outstanding brilliance of undulator radiation, very small sample volumes are sufficient to measure phonon spectra of good statistical quality within a few hours. This allows to determine thermodynamic properties like lattice specific heat and vibrational entropy even for systems of reduced dimensionality.

Here we have investigated thin films of polycrystalline Fe in the thickness range of 10 - 30 nm. Since the typical wavelength of a 30 meV phonon in bulk Fe is approximately 1 nm, we expect an influence of the confined geometry on the phonon spectrum of these films.

Method

The experiments were carried out at the undulator beamline 3ID. Monochromatization to an energy bandwidth of 5.5 meV was achieved by a high-resolution nested monochromator. Delayed fluorescence photons were counted within a time range of 12 – 100 ns after excitation with a large area APD (avalanche photodiode) that was placed right above the film plane. The measurements significantly benefitted from the intensity enhancement in x-ray standing waves that form above total

reflecting boundaries due to superposition of incident and reflected waves. A substantial intensity enhancement inside a thin film can be achieved if the thin film is coated on a total reflecting substrate and the film thickness is an integer multiple of the standing wave period [3,4]. Depending on the film thickness, a certain number of guided modes can be excited, which show up as minima in the rocking curve of the layer system between the critical angles of the layer and the substrate material. At those angular positions the fluorescence yield can be significantly enhanced [5].

Two films were prepared, consisting of polycrystalline α -Fe (enriched to 95 % in ⁵⁷Fe) with thicknesses of 13 nm and 28 nm, respectively.

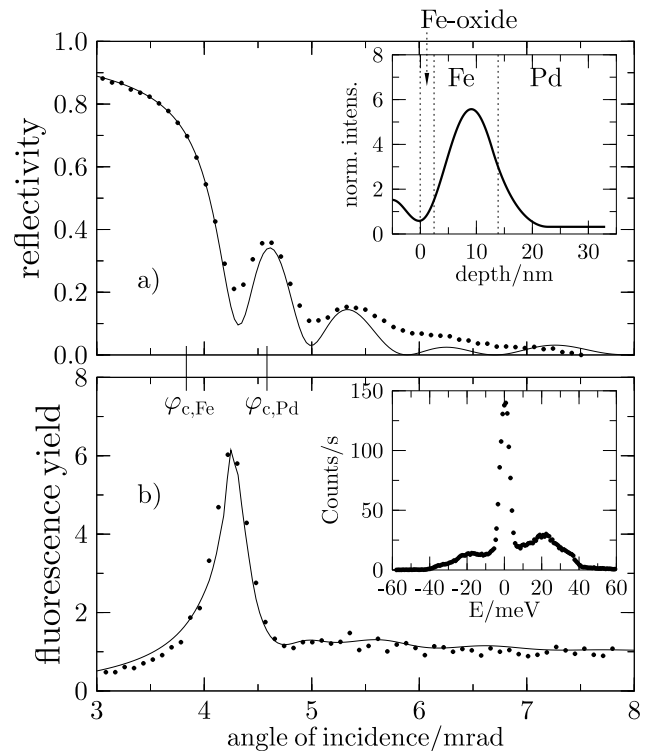


Figure 1: a) Electronic rockingcurve of 13 nm Fe on Pd. A guided mode is excited at an angle of 4.2 mrad that appears as minimum in the reflectivity between the two critical angles of Fe and Pd. The inset shows the depth dependence of the intensity inside the layer. b) Angular dependence of the yield of delayed fluorescence from the Fe film, with the photon energy 20 meV above the resonance. The yield peaks at the angle where the guided mode is excited. The inset shows the phonon spectrum that was recorded at this angle. Solid lines are theoretical fits.

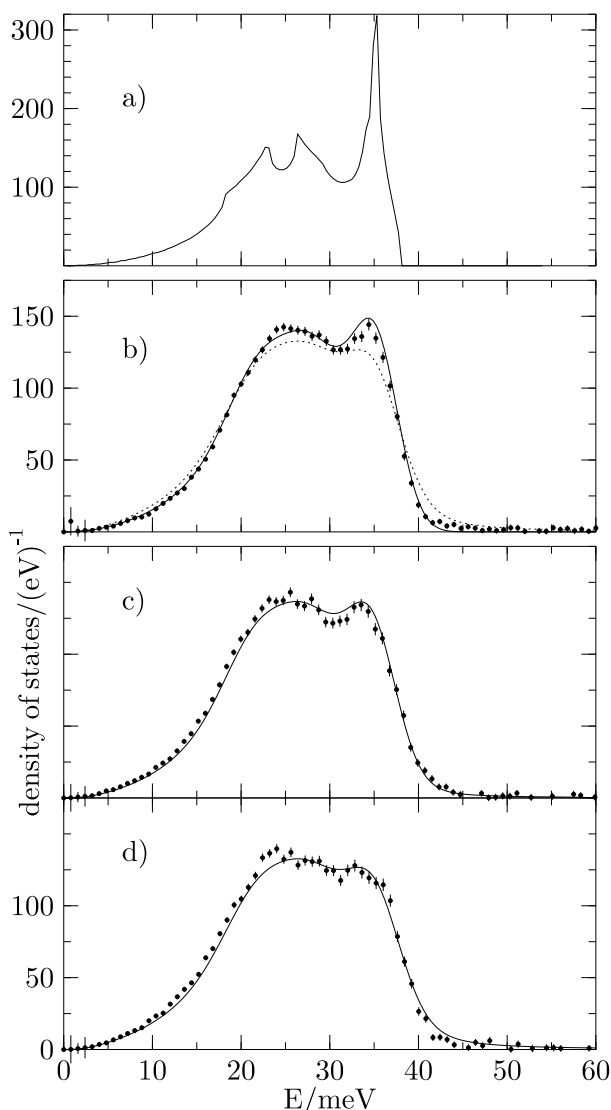


Figure 2: a) DOS of bulk Fe, from which the calculations (solid lines) for the following experimental data were derived: b) DOS of bulk Fe, c) DOS of 28-nm Fe on ZERODUR, d) DOS of 13-nm Fe on 20-nm Pd on ZERODUR. The dotted line in b) corresponds to the solid line in d). The energy resolution in the experiments was 5.5 meV [5].

The films were deposited at room temperature by rf-sputtering in an Ar atmosphere onto a superpolished glass-ceramics substrate. The 13-nm-thick film was deposited onto a 20-nm-thick Pd layer to create an x-ray waveguide structure. The intensity enhancement obtained through the waveguide effect is illustrated in fig. 1.

Results and Discussion

The DOS for both films is shown in fig. 2c,d. For comparison, the DOS of bulk α - ^{57}Fe , obtained from a 10- μm -thick foil under the same experimental conditions, is shown in fig. 2b. Fig. 2a shows the DOS of bulk bcc Fe as calculated from neutron data.

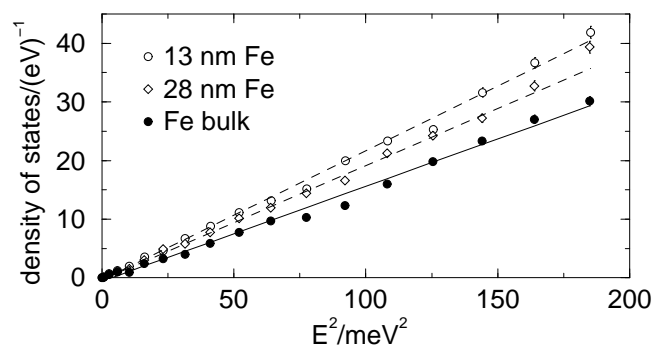


Figure 3: Low energy region of the phonon DOS of the Fe thin films and bulk Fe from fig. 2 plotted vs. E^2

The peak at 35 meV corresponds to longitudinal phonons close to the boundary of the first Brillouin zone, while the two peaks at 23 meV and 27 meV mainly belong to the van-Hove singularities of transverse phonons. The most obvious feature in the film DOS is the shape of the peak at 35 meV, which suggests phonon damping in the Fe films. In order to describe the measured DOS of the films, $g(E)$, we have applied the model of a damped harmonic oscillator (DHO) as it was used in a similar study on Fe nanocrystals [6]. This model contains as the only adjustable parameter the quality factor Q that describes the damping of vibrational modes. The DOS of the 28-nm-thick Fe film shown in fig. 2c was described with $Q = 25(2)$, while for the 13-nm-thick Fe film a value of $Q = 13(1)$ was obtained [5]. For a direct comparison with bulk bcc Fe, the calculated DOS of the 13-nm Fe film is drawn as dotted line in fig. 2b. The discrepancies between both calculated DOS reveal the features that are introduced by the damping: The high-energy tail is smeared out over a wide range of energies, the peaks due to the van-Hove singularities are smoothed, and there is a slight increase of the DOS at low energies, caused by the long tails of the DHO function.

An enhancement of low energy modes is often found in systems with structural irregularities and is attributed to softening of transverse phonons. The phonon DOS in this energy region for both samples is shown in fig. 3, plotted versus E^2 . The DOS is to a good approximation quadratic in energy, as expected from the Debye model. The slope indicates the sound velocity in the Fe films to be larger than in the bulk.

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