Effects of lattice expansion play an important role for the thermal properties of nanostructures like thin films and small particles. In general, lattice-expanded states are often found near surfaces and solid/solid boundaries where the atomic environment abruptly changes. In case of iron, the most prominent system is the interface between the Fe and the W(110) surface. It is generally accepted that the first layer of Fe on W(110) grows in a pseudomorphic fashion, i.e., with the Fe atoms aligned in registry with the W atoms of the substrate [1, 2]. This means a relative stretching of the Fe lattice constant by about 10% compared to its bulk value, implying significant changes of the elastic properties in this layer and the adjacent layers. With increasing number of layers one expects a gradual transition of the thermal properties towards those of bcc Fe, the nature of which, however, has not been revealed so far.

One route to determine elastic properties and thermal quantities is the measurement of the density of phonon states (DOS). These quantities are then calculated by a weighted integration over the DOS. Recently, nuclear inelastic scattering of x-rays has been applied to determine the DOS of nanostructured Fe like nanocrystals, precipitates and thin films, see, e.g., ref.[3] and references therein.

In the experiment described here, this method was employed under ultra-high vacuum (UHV) conditions to determine the DOS of ultrathin crystalline Fe films on W(110) down to thicknesses of one monolayer (ML) and to determine the evolution of their elastic properties. The experiments were performed at the nuclear resonance beamline ID18 [4] of the ESRF where a chamber dedicated for preparation and measurements under UHV conditions was installed. Fe films with an isotopic enrichment of 95% in $^{57}$Fe with thicknesses of 1, 2, 3, 10, and 40 ML (thickness equivalent: 1 ML = 0.2 nm) were epitaxially grown on the (110) surface of a tungsten single crystal. Energy spectra were obtained with a high-resolution (3.3 meV) monochromator scanning around the 14.4125 keV nuclear resonance, collecting the delayed Fe K-fluorescence. Fig. 1 displays the DOS that were extracted from the energy spectra by using a quasiharmonic model. The DOS of the 10 ML and 40 ML films resemble closely the DOS of bulk Fe but the spectral features are shifted to slightly lower values, e.g., the high-energy peak appears at 34 meV. With decreasing thickness one observes a further increase of low-energy modes while spectral features at high energies are significantly reduced.

The pronounced softening of the vibrational excitations close to the tungsten surface has a significant impact on the elastic properties, as shown in the right panel of Fig.1. To a good approximation one observes a linear dependence on $1/N$ over the range from bulk Fe to 2 ML thick films. Thus, one tends to attribute the deviations from the bulk behavior for films thicker than 1 ML to the contributions from the two atomic layers at the boundaries (the surface and the Fe/W interface). In a simple model, a given elastic quantity $f$ for a film consisting of $N$ atomic layers can be expressed as $f(N) = f_b(1 - n/N) + f_n n/N$, where $f_b$ is the value of the elastic quantity for bulk material and $f_n$ is the effective value for both interfacial regions that sum up to a total of $n$ atomic layers. The solid lines in Fig.1 (right panel) are plots according to the equation above with $n = 2$, i.e., the values for $f_2$ are those extracted from the measurement at the 2 ML film. Consequently, this linear decomposition of elastic quantities in contributions from the boundaries and the bulk should manifest also in a similar decomposition of the DOS. In fact, the DOS of the 3 ML film appears...
Figure 1: Left panel: DOS of single-crystalline Fe films on W(110) for thicknesses ranging from the monolayer to a 40 ML thick film. The curves are displaced from another by 0.04 meV$^{-1}$. The solid line in the upper graph represents the density of phonon states of polycrystalline bulk $\alpha$-iron, i.e., the ambient temperature bcc phase of iron as calculated from neutron data and convoluted with the energy resolution function of this experiment. The dashed and dotted line in the DOS of the 3 ML film represent the smoothed DOS of the 2 ML film and the DOS of bulk bcc Fe convoluted with a damped harmonic oscillator (quality factor $Q = 7$), respectively, out of which the DOS of the 3 ML film seems to be composed of. The solid lines for the 3 ML, 10 ML, and 40 ML DOS result from the weighted sum of these two contributions in each case.

Right panel: Elastic properties (mean displacement, average force constant) and the Debye enhancement factor $\alpha$ of Fe films on W(110) as function of $1/N$ where $N$ is the number of atomic layers. These quantities are derived from the DOS shown in the left panel. Points at $1/N = 0$ mark the bulk values. The solid lines are calculations according to the model described in the text.

to be a superposition of the 2 ML DOS and the DOS of bulk bcc Fe. In a similar fashion, also the DOS of the other films can be described, as indicated by the solid lines in Fig. 1 [5].

In conclusion, the phonon density of states of thin epitaxial Fe films on W can be described as being composed of contributions from the interfacial atomic layers and the interior of the film (see also Ref. [6]). Quite remarkably, the contribution from the inner part resembles closely the DOS of bulk bcc Fe (including a significant phonon lifetime broadening), while the boundaries contribution basically corresponds to the DOS of a 2 ML thick Fe film.

References