Introduction

Incoherent nuclear resonant absorption (INRA) of synchrotron radiation at the 14.4125 keV nuclear resonance of $^{57}$Fe [1–3] in combination with the $^{57}$Fe-isotope tracer-layer technique provides a unique method for the depth-selective direct measurement of the Fe-projected (partial) phonon density of states (DOS) in nanoscaled multilayers/superlattices. By artificially placing monolayer (ML)-thick $^{57}$Fe probe layers directly at the interfaces or at a certain distance from the interfaces in a multilayer/superlattices that otherwise contains nonresonant $^{56}$Fe layers, the phonon DOS of buried interfaces or buried layers can be conveniently measured. Using this technique, we have investigated Fe/Cr (001) superlattices and epitaxial $\text{Fe}_{x}\text{Cr}_{1-x}$ (001) single-layer alloy films.

Methods and Materials

The following types of Fe/Cr films (labeled FeCr1–FeCr5, respectively) were epitaxially grown in ultra-high vacuum (UHV) at 160°C by molecular beam epitaxy (MBE) on MgO(001) substrates carrying a 50Å Cr buffer layer, which was grown at 670°C:

- FeCr1: $[^{56}\text{Fe}(4\text{ML})/^{57}\text{Fe}(0.7\text{ML})/^{56}\text{Fe}(4\text{ML})/\text{Cr}(8\text{ML})]_{200}$,
- FeCr2: $[^{57}\text{Fe}(0.7\text{ML})/^{56}\text{Fe}(8\text{ML})/\text{Cr}(8\text{ML})]_{200}$,
- FeCr3: $[^{57}\text{Fe}(0.7\text{ML})/\text{Cr}(8\text{ML})]_{200}$,
- FeCr4: $[^{57}\text{Fe}_{0.03}\text{Cr}_{0.97}(001)]$, 7000 Å thick,
- FeCr5: $[^{57}\text{Fe}_{0.01}\text{Cr}_{0.99}(001)]$, 7000 Å thick,

The pressure during growth was $< 7 \cdot 10^{-10}$ mbar. Samples FeCr1 and FeCr2 were superlattices with (001) orientation. The 0.7 ML-thick probe layers (1 Å) of 95.5% enriched $^{57}$Fe were artificially placed either at the $^{56}$Fe-on-Cr interfaces (FeCr2) or in the center of the $^{56}$Fe layers (FeCr1), thus providing a nuclear resonance signal either from this Fe/Cr interface (FeCr2) or from the Fe-film center (FeCr1). Isotopically depleted $^{56}$Fe was used, which gives no nuclear resonance signal. In addition, we prepared an epitaxial film (FeCr3) that contains no bcc-$^{56}$Fe layers, but only 0.7 ML-thick $^{57}$Fe probes embedded on both sides between 8 ML-thick Cr(001) layers. The fourth sample was a 7000 Å-thick epitaxial $^{57}$Fe$_{0.03}$Cr$_{0.97}$ alloy film (FeCr4), that was prepared in UHV by thermal coevaporation of high-purity Cr and $^{57}$Fe, under the same conditions as the other samples. The last sample (FeCr5) was also an epitaxial alloy with a lower Fe concentration (1 at. % $^{57}$Fe). This sample was prepared under the same condition as the sample FeCr4.

Figure 1 shows high-angle ($\Theta$-2$\Theta$) x-ray diffraction patterns of multilayers FeCr1, FeCr2 and FeCr3, and alloy films FeCr4 and FeCr5. (Cu-K$\alpha$-radiation)

The INRA experiments were performed at the undulator beamline 3-ID of SRI-CAT at the Advanced Photon Source. The method of inelastic nuclear resonant absorption of 14.4125 keV x-rays is selective to the $^{57}$Fe resonant isotope and provides the Fe-projected (partial) phonon DOS rather directly with a minimum of modelling [2]. Details of the technique are described in ref. [1–3]. The monochromatized synchrotron radiation was incident onto the thin films under a grazing angle of $\approx 4$ mrad and had an energy bandwidth of 2.3 meV. The energy was tuned around the nuclear resonance of $^{57}$Fe. The measurements were performed at 300 K with collection times of ~10–24 hours per spectrum.

Results

The measured normalized data (not shown), i.e., the resulting phonon excitation probabilities per unit of energy, commonly feature a dominant elastic peak in a narrow energy range around the nuclear transition energy and side bands at lower and higher energy due to phonon annihilation and phonon creation, respectively [1–3]. The partial phonon DOS were extracted from the measured excitation probabilities by using the procedure described in [2].
The partial phonon DOS of our Fe/Cr-samples show distinct differences with each other (Figure 1) [4, 5]. The phonon DOS of the center site (FeCr1) is found to be similar to that of bulk bcc Fe [2], exhibiting peaks near 23 and 28 meV (transverse phonons) and 36 meV (longitudinal phonons). The phonon DOS of the other samples are different. Compared to the phonon DOS of the center site (FeCr1), the DOS peak of the interface site (FeCr2) near 36 meV is remarkably reduced in intensity, while the lower-energy feature (near 23 and 26 meV) is notably enhanced. Moreover, the lower-energy double-peak structure for the center site starts to change to a single-peak structure at the interface. The suppression of the longitudinal atomic vibrations and the enhancement of the transverse atomic vibrations at the interfaces is even more pronounced in sample FeCr3, where only 0.7 ML-thick $^{57}$Fe probes are on both sides in contact with 8 ML-thick Cr(001) layers. Sample FeCr3 clearly exhibits a rather strong single peak at lower energy (near 23 meV). The evolution of this 23 meV DOS peak is even more pronounced for the epitaxial 3 at.% and 1 at.% $^{57}$Fe-Cr alloy films (samples FeCr4 and FeCr5). The peak at 23 meV shares features of a vibrational resonance mode of Fe in the Cr matrix, i.e. Cr modes coupling to the Fe modes. Nevertheless, there is still a weak peak observable near 36 meV for these samples.

Discussion

The observed similarity of the DOS for bulk bcc Fe and sample FeCr1 (center side) demonstrates that phonon confinement (if it exists at all) within 8 ML-thick bcc-Fe films embedded between Cr layers is not detectable in the DOS. As a possible explanation for the reduction of the 36 meV peak intensity at the interface (sample FeCr2 and FeCr3), one should notice that its position nearly coincides with the deep minimum at ~ 34 meV in the bulk-phonon DOS of Cr [6]. Because of this valley in the phonon DOS of Cr, longitudinal phonons of Fe near 36 meV are partially suppressed at the Fe/Cr interface. An inspection of phonon dispersion curves of bulk Fe and Cr shows that this suppression affects longitudinal [00 $\xi$] phonons of Fe near the H point of the Brillouin zone [6]. Concerning the enhancement of the lower-energy DOS feature at the interface, one should notice that the number of Cr neighboring atoms around an $^{57}$Fe probe atom tend to increase from sample FeCr1 to sample FeCr4. This may be concluded by assuming ideally flat interfaces in the multilayer and a binomial distribution of atoms in the alloy. In this case, the number of nearest (n1) and next-nearest (n2) Cr neighboring atoms (configuration (n1,n2)) is (0,0), (4,1), (8,2), for samples FeCr1, FeCr2, FeCr3, respectivley, and approximately (8,6) for samples FeCr4 and FeCr5. These numbers are lower limits, valid only for the above assumptions. However, this tendency will even remain for moderately intermixed interfaces, as in our case. The peak intensity in the phonon DOS near 23 meV apparently correlates with the number of Cr neighbors. An Fe phonon mode near 23 meV with a low dispersion seems to develop with increasing number of Cr neighbors, implying decreasing average force constants. Summarizing, we have demonstrated that INRA of the 14.4125 keV $^{57}$Fe nuclear transition is sensitive for measuring the Fe-projected vibrational density of states in thin films, multilayers, and isotopically-enriched buried interfaces.

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References