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Phonon density of states in Fe/Cr(0 0 1) superlattices and Tb–Fe thin-film alloys

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Abstract

Inelastic nuclear scattering of X-rays from the 14.413 keV nuclear resonance of ^{57}Fe was employed to measure directly the Fe-projected phonon density of states (DOS) in MBE-grown Fe/Cr(0 0 1) superlattices on MgO(0 0 1). The Mössbauer-inactive ^{56}Fe isotope was used in the Fe layers. A 1 Å thick Mössbauer-active ^{57}Fe-probe layer (95% enriched) was placed at different locations within the Fe layers. This procedure permits to distinguish phonon density of states at the Fe–Cr-interface from that at the center of the Fe-film. Distinct differences have been observed in the DOS of our samples. The phonon DOS of an amorphous $Tb_{33}Fe_{67}$ alloy film was found to be a broad and structurless hump, contrary to that of an epitaxial $TbFe_2$ film, which exhibits characteristic features.

Keywords: Phonon density of states; Inelastic nuclear resonance scattering; Superlattices; Alloy films

Knowledge of the phonon density of states (DOS) is essential for our basic understanding of lattice vibrations. Extensive investigations of phonons in semiconducting superlattices have revealed novel phenomena, such as folding, confinement and interface modes [1]. For metallic multilayers (MMLs), on the other hand, only few reports of folded [2] or confined [3] phonons exist. Knowledge of vibrational properties of MMLs is highly desirable, also because their electronic properties may be affected by phonons [4]. In the present work inelastic nuclear scattering of X-rays from the 14.413 keV nuclear resonance of ^{57}Fe [5–7] was used to measure directly the Fe-projected phonon DOS in Fe/Cr(0 0 1) superlattices and in single films of epitaxial $TbFe_2$(1 1 0) (Laves phase) and amorphous $Tb_{33}Fe_{67}$ alloy.

Two types of Fe/Cr(0 0 1) superlattices (labeled Arg 03 and Arg 04, respectively) were epitaxially grown at 160°C by MBE on MgO(0 0 1) substrates carrying a 50 Å Cr buffer layer, which was grown at 670°C:

Arg 03:MgO(0 0 1)/Cr(50 Å)/[^{57}Fe(0.7 ML)/^{56}Fe(8 ML)/Cr(8 ML)]$_{200}$,

Arg 04:MgO(0 0 1)/Cr(50 Å)/[^{56}Fe(4 ML)/^{57}Fe(0.7 ML)/^{56}Fe(4 ML)/Cr(8 ML)]$_{200}$.

Isotopically depleted ^{56}Fe was used, which gives no nuclear resonance signal. In addition, 0.7 monolayers (ML) thick probe layers of 95.5% enriched ^{57}Fe were artificially placed either at the ^{56}Fe-on-Cr interfaces (Arg 03) or in the center of ^{56}Fe layers (Arg 04), thus providing a nuclear resonance signal either from this Fe/Cr interface (Arg 03) or from the Fe-film center (Arg 04). Fig. 1 shows X-ray diffraction (XRD) patterns of Fe/Cr-samples (Arg 03) and (Arg 04), respectively. Two symmetrical superstructure-satellite peaks demonstrate the high-quality superlattice structure of our Fe/Cr(0 0 1) multilayers.

The Tb–Fe alloy films were prepared in UHV by thermal co-evaporation of high-purity Tb and 95.5% enriched ^{57}Fe [8]. Films of 175 or 800 Å thick amorphous (a-)

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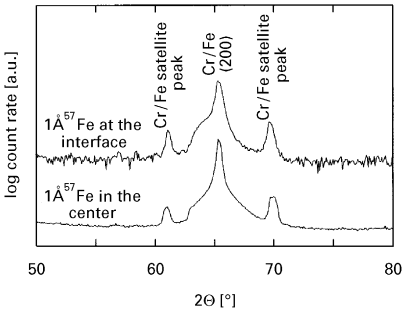

Fig. 1. XRD of Fe/Cr(0 0 1)-superlattice sample Arg 03 (top) and Arg 04(bottom) (Cu-K_α-radiation).

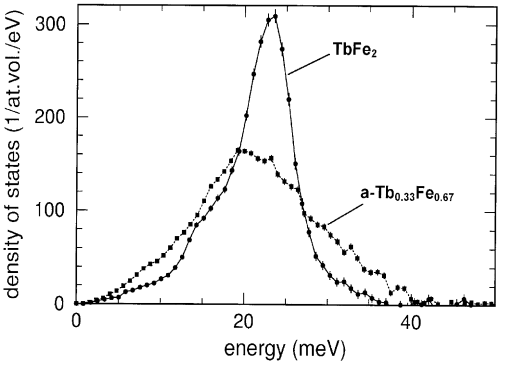

Fig. 2. Fe-projected phonon DOS of a-$Tb_{33}Fe_{67}$ alloy film (175 Å) (squares) and of epitaxial $TbFe_2$(1 1 0) film (800 Å) (circles).

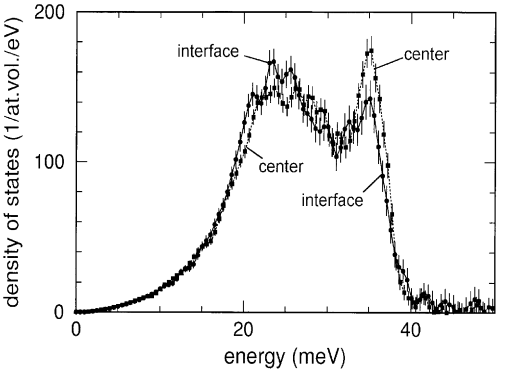

Fig. 3. Fe-projected phonon DOS of Fe/Cr(0 0 1)-superlattice sample Arg 03 (interface signal) (circles) and Arg 04 (signal from center of Fe layers)(squares).

$Tb_{33}{}^{57}Fe_{67}$ films were grown at 300 K and on Si(0 0 1) wafers initially coated with a 200 Å thick Pt buffer layer. The 800 Å thick epitaxial $TbFe_2$(1 1 0) film was grown at 500°C on a sapphire (11-20) substrate after deposition of a 350 Å thick Nb(1 1 0) buffer layer followed by a thin (15 Å) ^{57}Fe seed layer [9]. The samples were coated with $\sim$ 50 Å Si for protection. XRD patterns of the epitaxial $TbFe_2$ film and of the a-$Tb_{33}Fe_{67}$(175 Å) film confirmed the $TbFe_2$(1 1 0) epitaxial orientation and the amorphous structure of this Tb–Fe film, respectively. The inelastic nuclear resonant absorption experiments were performed at the undulator beamline 3-ID of SRI-CAT at the Advanced Photon Source. Details of the technique are described in Ref. [6]. The incident monochromatized synchrotron radiation had an energy band width of 5.5 meV (2.5 meV) in the case of the Tb–Fe alloy films (Fe/Cr superlattices). The energy was tuned around the 14.413 keV nuclear resonance of ^{57}Fe. The measurements were performed at 300 K with collection times of $\sim$ 10 h to $\sim$ 24 h per spectrum.

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 [5–7]. The partial phonon DOS were extracted from the measured excitation probabilities by using the procedure described in Ref. [6].

The partial vibrational DOS of the 175 Å thick a-$Tb_{33}Fe_{67}$ film (Fig. 2) extends up to $\sim$ 40 meV and shows a maximum at $\sim$ 20 meV. It represents a structureless broad feature, as anticipated for such an atomically disordered material. The same result was obtained on a 800 Å thick a-$Tb_{33}Fe_{67}$ film (not shown). By contrast, the partial DOS of the crystalline $TbFe_2$ film (800 Å) exhibits a sharper maximum at $\sim$ 23 meV and extends to $\sim$ 35 meV. Moreover, a distinct shoulder appears near 15 meV. Comparison with phonon DOS and dispersion curves for other (bulk) Laves-phase compounds obtained by inelastic neutron scattering [10] shows that the shoulder near 15 meV is related to the high DOS at the Brillouin zone boundary of acoustic modes (longitudinal and transverse), while the main peak near 23 meV is due to optical modes.

The partial phonon DOS of Fe/Cr-samples Arg 03 (interface site) and Arg 04 (center site) show distinct differences (Fig. 3). The DOS of the center site is very similar to that of bulk Fe [6], exhibiting peaks near 23, 28 and 36 meV. Compared to the DOS of the center site (Arg 04), the DOS-peak of the interface site (Arg 03) near 36 meV is remarkably reduced in intensity, while the lower energy feature is notably enhanced. As a possible explanation for the reduction of 36 meV peak intensity one should notice that its position nearly coincides with the deep minimum at $\sim$ 34 meV in the bulk-phonon DOS of Cr [10]. Because of this lack of phonon DOS in Cr, part of the Fe phonons near 34 meV remain confined in the Fe layer and are suppressed at the Fe/Cr interface. This

confinement affects longitudinal [0 0 ζ] phonons of Fe near the H point of the Brillouin zone [10].

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