Inelastic nuclear resonant absorption of synchrotron radiation in thin terbium-iron films

T. Ruckert*, W. Keune*, W. Sturhahn◆, M.Y. Hu◆, J.P. Sutter◆, T.S. Toellner◆, E.E. Alp◆, and R. Roehlsberger◆
*Laboratorium für Angewandte Physik, Gerhard-Mercator-Universität, D-47048 Duisburg, Germany
◆Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439 USA

Introduction

Inelastic nuclear resonant absorption of synchrotron radiation is an efficient and unique method for the direct measurement of vibrational density of states (VDOS) of thin films that contain Mössbauer isotopes. This is demonstrated for the ^{57}Fe nuclear resonance in the case of amorphous and crystalline Tb-Fe alloy thin films. The vibrational properties of the amorphous thin films are characteristically different from that of the crystalline phase. Especially the phonon DOS of an amorphous $Tb_{0.33}Fe_{0.67}$ alloy film was found to be a broad and structureless hump, contrary to that of an epitaxial $TbFe_2$ film, which exhibits characteristic, sharp features.

Methods and Materials

Our samples were amorphous (a-) $Tb_{1-x}Fe_x$-alloy thin films with x = 0.30, 0.50, 0.67, 0.77, and 0.82. The thickness of the films was between 17.5–80 nm. For comparison, we also investigated an 80 nm-thick epitaxial (crystalline) $TbFe_2$(110) film (cubic Laves phase). All alloy thin films were prepared in ultrahigh vacuum by thermal co-evaporation of Tb (99.99% purity) and 95.5%-enriched ^{57}Fe metal [1]. The amorphous films were grown at 300 K on oxidized Si(001) wafers. For the 17.5 nm-thick films the Si wafers were initially coated with a 20 nm-thick Pt buffer layer. Reflection of the synchrotron beam at the Pt layer in grazing incidence enhances the 6.4 keV fluorescence signal of inelastic nuclear resonance absorption in the a-$Tb_{1-x}Fe_x$ overlayer due to a wave-guide effect ("GIAR" effect) [2]. The $TbFe_2$(110) film was deposited at 500°C on a sapphire (11-20) substrate which carried a 35 nm Nb(110) buffer layer and a 1.5 nm ^{57}Fe seed layer on the Nb surface [3, 4]. All samples were coated by ~ 5 nm Si for protection against oxidation

Fig. 1 exhibits typical x-ray diffraction (XRD) patterns of the epitaxial $TbFe_2$ film (top) and of the a-$Tb_{0.33}Fe_{0.67}$(17.5 nm) film (bottom). This result confirms the $TbFe_2$(110) epitaxial orientation and the amorphous structure of this Tb-Fe film, respectively; in the latter case only Bragg peaks of the Pt buffer layer and no Bragg reflections of the amorphous Tb-Fe film are observed. The amorphous structure of all a-$Tb_{1-x}Fe_x$ films was confirmed further by XRD and conversion electron Mössbauer spectroscopy (CEMS) [1].

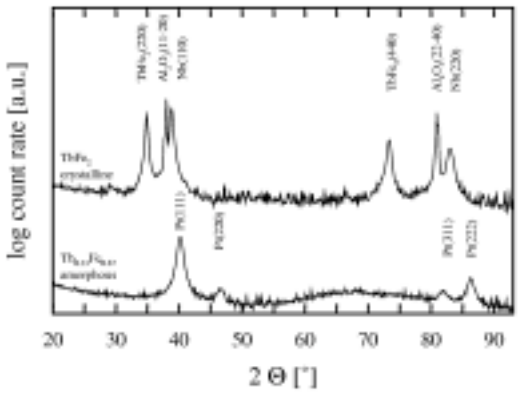

Figure 1: XRD of 800 Å-thick epitaxial $TbFe_2$ (110) film (top) and of 175 Å-thick amorphous $Tb_{33}Fe_{67}$ alloy film (bottom).

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 [5–9]. The synchrotron radiation had an energy bandwidth of 5.5 meV in the case of the amorphous alloys and 2.3 meV for $TbFe_2$ and a-$Tb_{0.33}Fe_{0.67}$.

Results and Discussion

The inelastic spectra for the $TbFe_2$ Laves phase measured at 300 and 10 K are shown in Fig. 2. The commonly feature is 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 phonon annihilation disappeared at low temperatures (10 K), as expected. The partial phonon DOS were extracted from the data by using the procedure described in [6]. Here we discuss only the 300 K results.

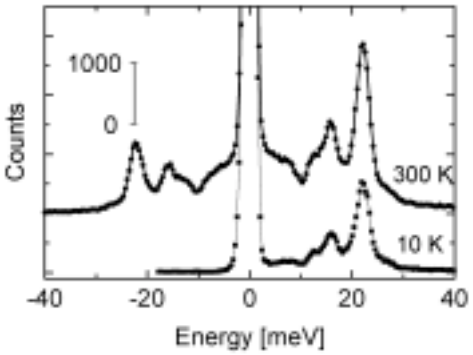

Figure 2: Experimental data for the $TbFe_2$ Laves phase at two temperatures (300 K and 10 K).

The partial vibrational DOS of the 175 Å thick a-$Tb_{33}Fe_{67}$ film (Fig. 3) extends up to ~ 40 meV and shows a maximum at ~ 20 meV. It represents a structureless broad feature, as anticipated for such an atomically disordered material. By contrast, the partial DOS of the crystalline $TbFe_2$ film (800 Å) exhibits two sharp maxima at ~22.5 meV and ~16 meV. The DOS extends to ~ 35 meV. Comparison with phonon DOS and dispersion curves for other (bulk) Laves-phase compounds obtained by inelastic neutron scattering [10] shows that the peak near 16 meV is related to the high DOS at the Brillouin zone boundary of acoustic modes (longitudinal and transverse), while the main peak near 22.5 meV is due to optical modes.

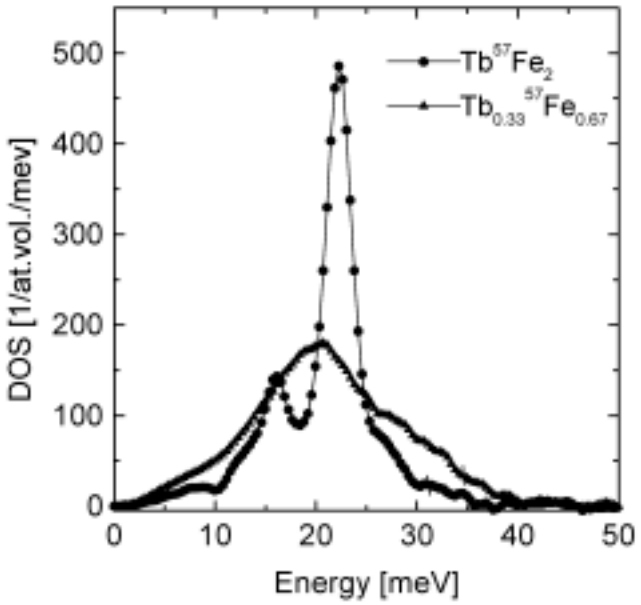

Figure 3: Fe-projected phonon DOS of a-$Tb_{33}Fe_{67}$ alloy film (175 Å) (triangles) and of epitaxial $TbFe_2$(110) film (800 Å) (circles), measured at 300 K (2.3 meV energy resolution).

For the other amorphous $Tb_{1-x}Fe_x$ films, the DOS represents a structureless broad feature, as anticipated for such atomically disordered materials (Fig. 4). The maximum energy of the DOS and the position of the main peak in Fig. 4 shift to lower energies with decreasing Fe content x of the amorphous films (i.e., the Fe vibrations soften with decreasing iron content). The same DOS were measured on 80 nm-thick and 17.5 nm-thick a-TbFe films [9] (i.e., the DOS does not depend on the thickness in this thickness regime).

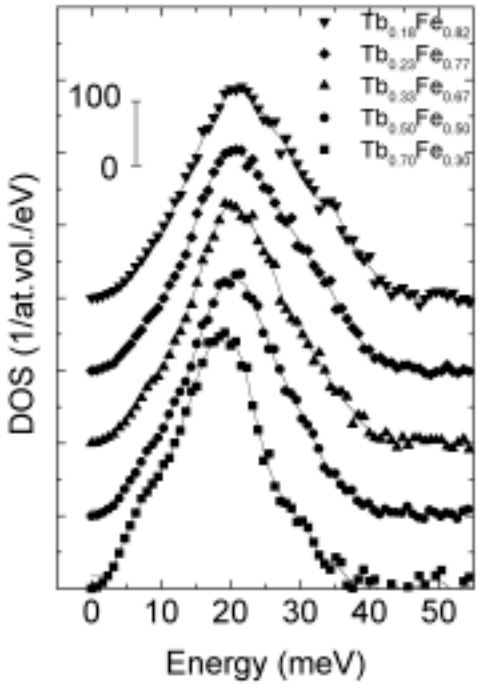

Figure 4: Partial density of states (DOS) versus energy of 17.5 nm-thick amorphous $Tb_{1-x}Fe_x$ films with decreasing iron content x (from top to bottom) (5.5 meV energy resolution). The drawn lines are a guide for the eye only.

It is possible to determine directly the temperature-dependent probability for recoilless absorption, i.e., the Lamb-Mössbauer factor (f-factor) [11, 12], from the data. The f-factor at 300 K is significantly lower for the alloy films than for bulk iron ($f_{bulk\ Fe}$ = 0.8). We found $f_{amorp.}$ = 0.4–0.65 for the amorphous alloys. The probability for recoilless absorption increases with the iron concentration x of the amorphous alloys. The f-factor for the Lavers phase is somewhat higher than for the amorphous alloys ($f_{Laves.}$ = 0.68).

Summarizing, we have demonstrated that inelastic nuclear resonant absorption of synchrotron radiation via the 14.4125 keV ^{57}Fe nuclear transition is a unique method for the direct measurement of Fe-projected VDOS in crystalline and amorphous thin films. At the Advanced Photon Source, practical measurement times are achieved with an effective total ^{57}Fe thickness in the samples of the order of only ~ 5 nm. This opens vibrational dynamics as a new domain in thin-film research.

Acknowledgments

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