Z. Kristallogr. Suppl. **26** (2007) 229-234

Texture of submicron Ni-Mn-Ga films studied by X-ray diffraction at the ANKA synchrotron source

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Keywords: X-ray synchrotron diffraction, texture, Ni-Mn-Ga/Mo thin film composites

Abstract. Texture measurements of two series of Ni-Mn-Ga thin films, sputter-deposited on Mo foils, are performed as a function of both film and substrate thickness. X-ray diffraction experiments were carried out at the SR source ANKA in Germany. In contrast to 1μm-thick films showing 220-fibre texture, both in-plane and out-of-plane texture components are found in submicron films which correlates with the enhanced magnetostrain effect in these films.

Introduction

Thin film technology utilising magnetostrained shape memory Ni-Mn-Ga alloys is currently under development in order to extend their functionality to the submicron scale. Since the magnetomechanical activation of polycrystalline films depends on their microstructure, the texture which develops during deposition/annealing has a crucial importance. Previous studies have shown that the martensitic Ni-Mn-Ga films deposited on Al_2O_3 ceramic have a 220-fibre texture which depends on the film thickness and substrate surface roughness [1,2]. In the case of the practically important Ni-Mn-Ga/Mo thin film composites showing a large increase of magnetostrain for decreasing film thickness [3,4], the texture features have not been clarified yet. In this work, the texture of Ni-Mn-Ga thin films sputter-deposited on Mo foils is studied by XRD at the PDIFF beamline at the ANKA synchrotron source [5]. The texture measurements of these films are performed in the cubic phase as a function of film composition, film thickness and substrate thickness.

Experimental method

Two series of Ni-Mn-Ga thin films with thicknesses of 0.1, 0.4 and 1.0µm, sputter-deposited on 5µm- and 10µm-thick molybdenum foils, were prepared using two targets with nominal compositions of $Ni_{49.5}Mn_{28}Ga_{22.5}$ (Ni49 film) and $Ni_{52}Mn_{24}Ga_{24}$ (Ni52 film). These film composites were vacuum annealed at 800°C for 1hr. The film compositions and heat treatment ensured the formation of tetragonal (Ni49 film) and orthorhombic (Ni52 film) modulated martensitic phases stable at room temperature [1,2,4] and a grain size typically twice the film thickness [1], which is commonly found for single-phased metallic thin films [6-8]. Commercially available Mo foils have a 100 out-of-plane texture as they are prepared by rolling [9].

X-ray diffraction measurements were conducted at the ANKA synchrotron source. The divergency of the beam was 0.05mrad, the wavelength was 0.15434nm with a standard deviation of 10^{-5}nm. The beam had a cross section of 1mm x 2mm and covered about 10^6 to 10^8 grains, assuring statistically significant sampling. Measurements were carried out using a scintillation detector with a 0.1° Soller collimator providing angular resolution. The goniometer was equipped with an in-house designed heating stage. The sample layout is shown in figure 1. The full 2θ-scans at room temperature showed in the range of 40 – 50° one cluster of peaks produced by a corresponding martensitic phase.

In the cubic $L2_1$ ordered phase which is stable at the selected temperature 150°C only the 220_C and 400_C peaks were found. These peaks were located at the 2θ-range of 43.5 – 44.5° and 63 – 66°, respectively. The integral intensities of these peaks were studied as a function of rotation angle Φ with steps of 10° in the range 0 – 90° at various constant tilting angles χ varied with steps of 10° in the range 0 – -90°. For χ = -90°, diffracting planes are parallel to the film plane. Data were corrected for the decay of synchrotron beam intensity. All texture measurements were made at 150°C. In addition, one representative cooling ramp with χ = -90° was recorded to confirm the martensitic transformation (MT) and accompanying lattice distortion. Since the data of the diffraction peak were collected during about 30s and the cooling rate was 5K/min, the uncertainty of the sample temperature is 5K.

For the magnetostrain measurements, single-beam cantilevers of 2mm width and 10mm length were fabricated by laser cutting of the Ni52/Mo film composites. A bending type deformation perpendicular to the length of the cantilever was determined as a function of the magnetic field applied along the length of cantilever using the method of laser beam reflection.

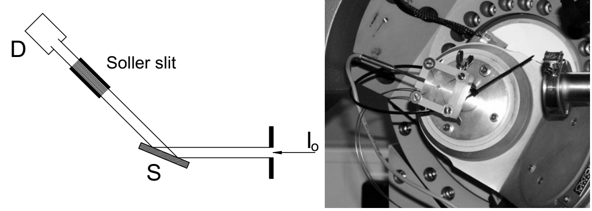

Figure 1: (left) schematic of scattering setup at ANKA, (right) the sample mounted on the heating-stage on the Kappa-goniometer: the sample measures approx. 2 x 2mm

Results and discussion

Figure 2 shows the results of 3D-presentation of integrated X-ray intensities of 220_C and 400_C peaks for Ni52/Mo(10µm) film composite series as a function of χ and Φ.

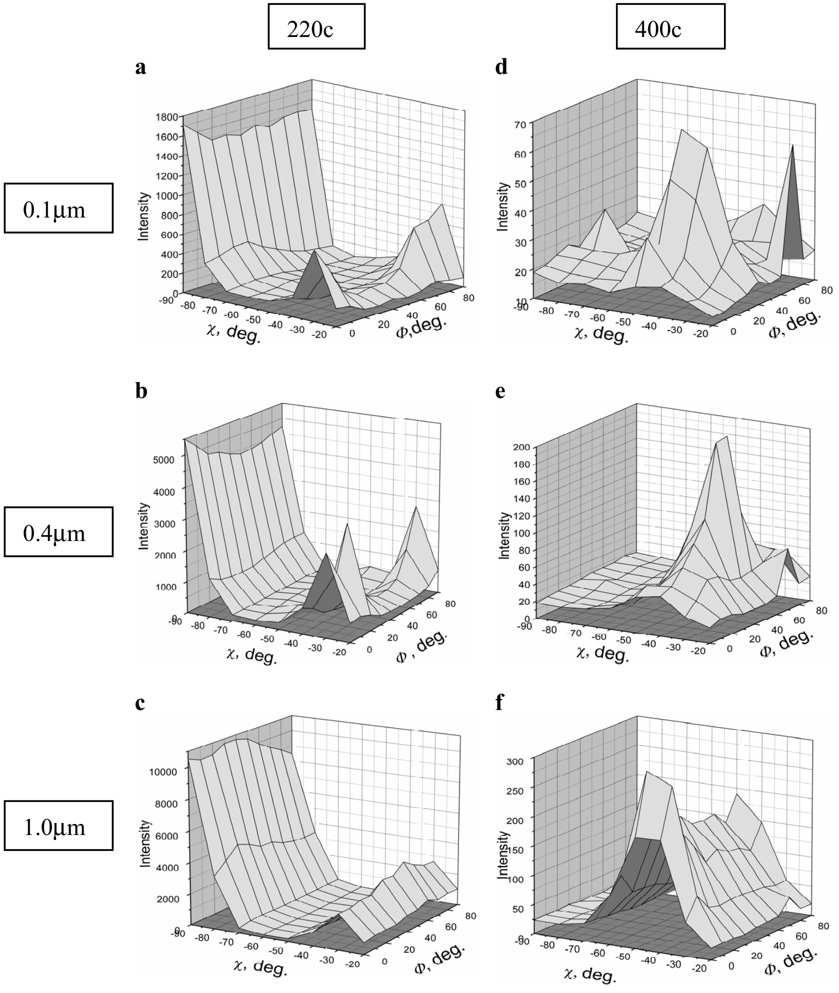

Figure 2. Typical set of 3D-diagrams of the integrated intensity calculated from the 220_C (left column) and 400_C peaks (right column) of 2θ-scans measured at 150 °C for 0.1 (a,d), 0.4 (b,e) and 1µm (c,f) Ni52/Mo thin film composites as a function of tilt angle χ and rotation angle Φ. The thickness of the Mo foil substrate is 10µm.

Qualitatively similar behaviour is obtained for the Ni49/Mo(10μm) composites as well as for the films deposited on 5μm-thick Mo foil. In correspondence to figure 2(c), the 1μm-thick Ni–Mn–Ga film shows in the cubic phase a fairly strong 220-fibre texture: the maximum of (220)-close-packed peak intensity is found at $\chi = -90^o$ (with (220) planes parallel to the film plane) independently on the angle Φ. In addition, a peak occurs at inclination to the film surface of about 30^o ($\chi \approx -30^o$), which is independent of Φ. This peak stems from the (202)-close-packed planes which are inclined by 60^o towards (220).

In the thinner films, 3D-maps of intensity distribution are more complicated, see figures 2(a) and (b). Again a Φ-independent maximum of 220 peak intensity is found at $\chi = -90^o$ evidencing a preferable orientation of 220-close-packed planes in plane of film. Contrary to 1μm-thick films, an additional maximum of 220 peak intensity at $\chi \approx -30^o$ shows a strong Φ dependence. This fact suggests an in-plane texture component in submicron Ni-Mn-Ga/Mo films. The details of latter texture can be studied by observations of the full pole figures. According to the 3D-maps shown in figures 2(d),(e) and (f), both an angular position of the 400 peak intensity maximum, which is about $\chi \approx -45^o$ with regard to the <220> directions, and its χ and/or Φ dependence is in line with the aforementioned interpretations.

Figure 3(a) shows the temperature dependence of the X-ray diffraction peak ($\chi = -90^o$) measured during cooling from the cubic phase to the martensitic phase (C → M). Because the c/a < 1 type of lattice distortion proceeds in the textured film, a splitting of the cubic 220_C peak is accompanied by the appearance of one broad $202_M/022_M$ maximum in a tetragonal/orthorhombic martensitic phase while 220_M reflection is not observed indicating the preferential formation of martensite variants with the long lattice parameter *a* parallel to the film plane and the short lattice parameter *c* inclined to the film plane by 45^o. This result indicates that the film is under tensile stress while undergoing the martensitic transformation. Furthermore, there are two twin-related variants with *c* inclined by 45^o to the film plane. The twinning planes of these variants are parallel to the film plane as found experimentally in [1]. The inset to figure 3(a) yields a martensitic transformation by the anomalous decrease of lattice parameter calculated assuming cubic unit cell. This is the first direct experimental evidence of MT in the Ni-Mn-Ga films attached to a metallic substrate.

Since the martensitic transformation is diffusionless, a martensitic substructure inherits the texture features of the cubic phase [2]. Therefore it is worthwhile to compare the texture characteristics of Ni-Mn-Ga/Mo films measured in the cubic phase with those of Ni-Mn-Ga/Al_2O_3 films obtained in the martensitic phase. From this comparison, a qualitatively different behavior is deduced, namely, in contrast to films on alumina, the films on Mo show (i) an additional in-plane texture component, which was mentioned above, and (ii) a reverse dependence of the major out-of-plane texture component on the film thickness. The latter feature follows from figure 3(b) where the quantity $\Delta\chi$, characterizing spread of the inclination angle of out-of-plane component of texture in Ni-Mn-Ga/Mo films, tends in average to increase as a function of film thickness. In the case of films on alumina, this quantity demonstrates a substantial descending thickness evolution [2]. A reasonable explanation of the difference (ii) stems most probably from the considerable different quality of substrate surface: as compared with Mo foil, the surface of alumina ceramic has 2 times larger both RMS roughness and maximum peak-to-valley range (see [2] and references therein).

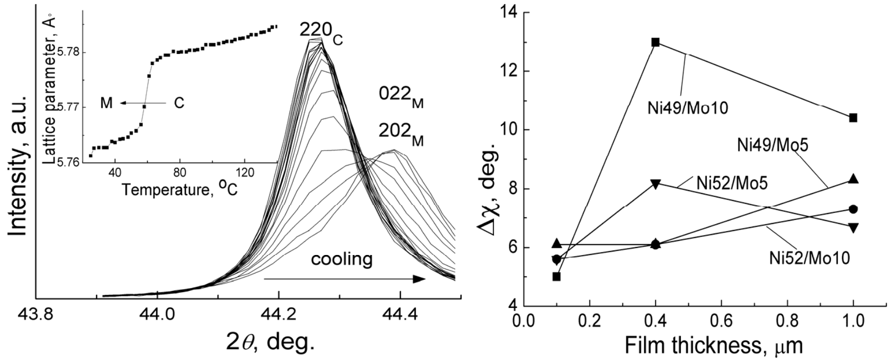

Figure 3(left): 220_C X-ray peak of cubic phase, C, for 1μm-thick Ni52/Mo(10μm) composite film exhibiting a drastic change in shape and position at MT into martensitic phase, M, during cooling from 150 ^oC ($\chi = -90^o$). Inset shows the corresponding temperature dependence of lattice parameter.
Figure 3(right): Spread of the inclination angle of out-of-plane texture at $\Phi = 0$ in Ni-Mn-Ga/Mo films as a function of film thickness. The lines are guides for the eye.

An additional in-plane crystallographic anisotropy of the submicron Ni-Mn-Ga/Mo film composites found in the present work is assumed to be a consequence of the in-plane rolling texture of the Mo substrate mentioned in the section 'Experimental method'. The strong rolling texture of the substrate [9] combined with the elastic anisotropy of Mo (anisotropy ratio 0.775) imposes an elastic anisotropy of the substrate. When the film is thin compared to the substrate (as for the 0.1μm-thick film), the substrate anisotropy causes anisotropic in-plane stresses in the film during deposition leading to an in-plane texture. As far as the in-plane texture can enhance an in-plane unidirectional magnetic field actuation of such composites, the increased magnetostrain response measured in [4] for thinner Ni-Mn-Ga/Mo films can be linked to the rolling texture of the substrate. In figure 4, the experimental magnetostrain curves for Ni52/Mo(10μm) composite films are depicted.

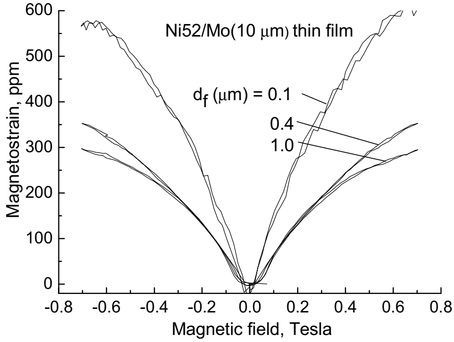

Figure 4. Magnetostrain curves for Ni52/Mo(10μm) thin films composites showing evolution with film thickness

As is evident in figure 4, the maximum magnetostrain is observed for 0.1μm-film. This finding correlates with the texture evolutions shown in figures 2 and 3. Thus, the magnetic-field-induced strain increases with increasing degree of crystallographic anisotropy in the Ni-Mn-Ga film composites. Furthermore, the deflection of a substrate is proportional to the film thickness and the film stress (in the Stoney approximation which is valid for the 0.1μm-thick film), the results of the magneto-mechanical experiments indicate that the 0.1μm-thick film is able to support a stress several times larger than the 1μm-thick film. Thus, much larger specific output of the submicron films in comparison with the thicker ones can be obtained.

Summary

The texture evolution in Ni-Mn-Ga thin films sputter-deposited on Mo foils was studied as a function of film thickness using X-ray diffraction. The texture measurements were performed in a cubic phase at 150°C. It is shown that the films are transforming martensitically while cooling to room temperature. The magnetostrain effect of Ni-Mn-Ga/Mo film composites correlates with both the spread of the inclination angle of the habit plane to the film plane and an existence of the in-plane texture component.

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Acknowledgements. VAC is grateful to Fondazione Cariplo (project 2004.1819-A10.9251) as well as to the IMT and the ANKA facility at Forschungszentrum Karlsruhe for providing financial support and synchrotron beam time in a framework of proposal MS-12. The authors thank K. Seemann and A.Agarval for valuable help in magnetostrain measurements and M. Beiser for laser cutting the specimens. PM acknowledges partial support through the National Science Foundation (NSF) under project No. 0502551 and through DARPA under contract No. N660001-01-8034.