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Magnetically Anisotropic Ni_2MnGa Thin films: Coating Glass and Si Micro-Cantilevers Substrates

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Abstract. Ni_2MnGa thin films, with thickness between 30 and 60 nm, were pulsed-laser deposited at room temperature on Si micro-cantilevers and glass substrates. Two different deposition processes were performed: normal deposition and off-normal. After annealing in an inert atmosphere, in-plane isotropic magnetic hysteresis loops were measured for the normal deposited films. In contrast, in-plane anisotropic hysteresis loops were obtained from the off-normal deposited ones. An in-plane easy direction for the magnetisation, perpendicular to the incidence plane of the plasma during deposition, was measured with an anisotropy field of ≈100 Oe and an easy coercive field of ≈24 Oe. The mechanical behaviour of the magnetically anisotropic coated micro-cantilevers and their response to a decreasing temperature permitted observing the martensitic transformation of the Ni_2MnGa thin films.

Introduction

Ferromagnetic shape memory alloys (FSMA) are ferromagnetic Heusler alloys exhibiting martensitic transformation as well as the property of being magnetically strained [1]. Such materials include NiMnGa [2] and other alloys [3–8] and are produced by a large variety of techniques, such as arc melting [9], melt-spinning [10, 11], sputtering [12–14] and Pulsed Laser Deposition (PLD) [15–17].

The NiMnGa system has attracted an especially great deal of interest because of its capability of producing magnetostrain on the order of 10% in bulk single crystals [18], which has high application potential for sensors and actuators [13 and references therein, 19, 20]. Recently, efforts have been made to grow FSMA films with thickness between 0.1 μm and several microns, either polycrystalline or single crystal [6, 21–23], because FSMA thin films are a prerequisite for miniaturised devices.

Micro-cantilevers are mechanical devices with very interesting applications and related phenomena. At first they were used to measure surface topographies in atomic force microscopes [24], and since then they have been largely utilised as high-sensitivity sensors in many different physical, chemical and biological applications [25, 26]. Previous works have also shown the deposition of thin films on micro-cantilevers for different applications, for instance, to perform non-contact current measurements [27], precise magnetic determinations in films [28] or for studying magneto-mechanical properties of soft magnetic Co-coated micro-cantilevers [29].

Some authors have studied the thermo- and magneto-elastic properties of FSMA with thicknesses on the order of 1 μm deposited on Si cantilever-shaped substrates [30]. NiMnGa/Mo composites, up to 10 μm in thickness, have also been used to fabricate single beam cantilevers of 1 mm in width and lengths in the range of 4-8 mm for studying the shape memory effect and magnetostriction [31], including the use of substrate release techniques to obtain free-standing FSMA films.

It has long been established that the growth of thin films by off-normal deposition allows for the induction of uniaxial in-plane magnetic anisotropy [32-34]. Currently, different methods using the oblique-incidence geometry, such as molecular beam epitaxy [35], vacuum deposition [36], e-beam evaporation [37], sputtering [38], and pulsed laser deposition [39], are suitable for growing of a large variety of thin films that exhibit such anisotropies.

In this work, we show the deposition of NiMnGa thin films with thicknesses between 30 and 60 nm onto glasses and Si micro-cantilevers. The controlled generation of in-plane magnetically anisotropic NiMnGa thin films will be shown. Furthermore, the mechanical behaviour of the coated micro-cantilevers and their temperature dependence can be used to study the martensitic transformation of FSMA thin films. These micro-scale substrates have the advantage of allowing the study FSMA thin films with thicknesses down to 100 nm.

Experimental

NiMnGa thin films were grown by PLD onto commercial Si micro-cantilevers that are 450 μm long, 50 μm wide, and 2 μm thick –on their face, 450x50 μm^2, opposite to the tip of the micro-cantilever– and on glass substrates that are 7 mm in diameter. The target, with a nominal composition of $Ni_{52}Mn_{24}Ga_{24}$, was prepared starting from the pure elements, Ni and Mn powders and Ga pressed at 450 kg/cm^2. The pressed mixture was melted for alloying in an induction furnace in an inert atmosphere. For PLD, a Nd-YAG laser beam with λ = 1024 nm, 20 Hz repetition rate, 4 ns pulses and energy per pulse of 220 mJ, delivered only to the target, was introduced into the chamber through a quartz window. The chamber was at a base pressure of 10^{-6} mbar. The area of the laser beam on the target was ≈13 mm^2. The incidence angle of the laser beam on the target was 45°. All substrates were at room temperature during deposition. The target-to-substrate distance was 75 mm. Two different PLD processes were performed: normal deposition and off-normal (oblique) deposition. For the oblique deposition, the micro-cantilevers and the glass substrates were placed onto the lateral surface of a rotating cone to allow the deposition at an off-normal angle θ; in this work, θ = 50°. Micro-cantilever holders were designed so that two micro-cantilevers were simultaneously coated at the same θ, one with its longitudinal axis positioned parallel to the cone generatrix and the other perpendicular to it, that is, parallel or perpendicular to the incidence plane of the plasma respectively. The deposition time was varied such that films with different thickness, between ≈30 and 60 nm, were grown.

The as-deposited samples were annealed in 99.999 pure Ar for different times, typically for 5 and 20 min, at different temperatures between 653 and 723 K.

The temperature dependence of the resistance of the films was measured between 200 and 300 K using the four-probe technique. The magnetic hysteresis loops of the coated glass circles were determined by vibrating sample magnetometry (VSM). The in-plane magnetic field was applied parallel or perpendicular to the direction of the incidence plane of the plasma. Also, the Magneto-Optical Kerr Effect (MOKE) was used to magnetically characterise the films by applying the in-plane magnetic field in the two directions described before. Apart an optical system was used for measuring the intensity of the polarised transmitted light through the films deposited on glass as a function of the angle between the polarised light plane and the incidence plane of the plasma during PLD.

A special, N refrigerated chamber was built for simultaneous cooling and optical observation of the micro-cantilever deflection using a monocular microscope with a CCD, with a final magnification of 1000X. This deflection was determined by measuring the vertical deviation from the horizontal position of the micro-cantilever's free end.

Results and Discussion

The optical and magnetic properties of these PLD Ni_2MnGa thin films are shown below. Fig. 1 shows the optical behaviour of two PLD Ni_2MnGa thin films deposited on glass. In previous works, we have demonstrated the controlled generation of nano-strings during off-normal PLD of thin films

[39]. These nano-strings generated an optical anisotropy, as well as electrical and magnetic anisotropies [40].An optical anisotropy was generated in the off-normal deposited thin films.

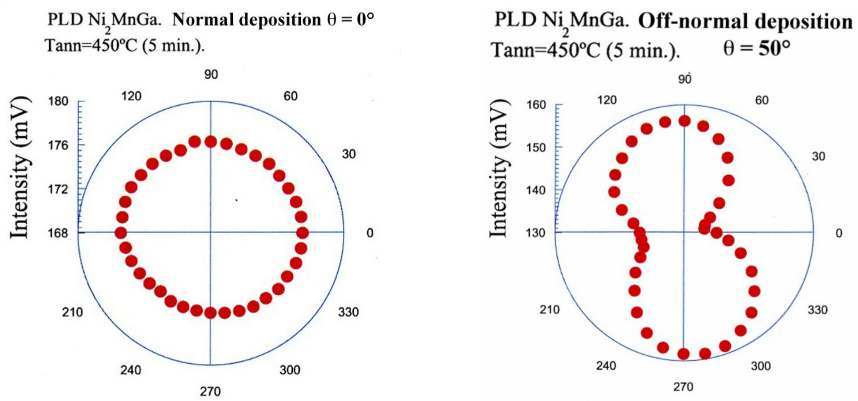

Fig. 1: Intensity of the transmitted polarised light through two Ni_2MnGa annealed films deposited on glass as a function of the angle between the polarised light plane and the incidence plane of the plasma during PLD: left, sample deposited at normal PLD (optically isotropic). Right, sample deposited at off-normal PLD (optically anisotropic).

No ferromagnetic property was detected in the Ni_2MnGa as-deposited thin films. However, after annealing, all films exhibited ferromagnetic behaviour. In-plane isotropic magnetic behaviour was measured for all the normal PLD films. Conversely, the off-normal PLD films exhibited a clear in-plane magnetic anisotropy (Fig. 2). An easy direction for the magnetisation was present perpendicular to the plasma incidence plane during deposition. Anisotropy fields, $H_k \approx 100$ Oe, and easy coercive fields, $H_c \approx 25$ Oe, were measured.

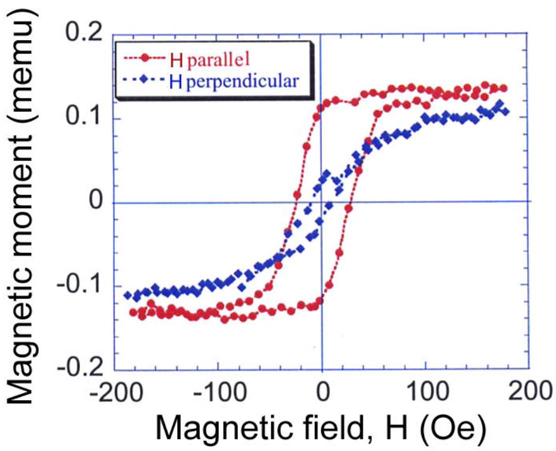

Fig. 2: VSM in-plane hysteresis loops, at room temperature, austenite state, of an off-normal, heat-treated PLD Ni_2MnGa thin film. The magnetic field was applied parallel and perpendicular to the easy direction for the magnetisation. A magnetic anisotropy was generated in the direction perpendicular to the plasma incidence plane during deposition. The anisotropy field H_k was ≈100 Oe, and the easy coercive field H_c was ≈25 Oe.

Fig. 3 (left) shows a picture of an off-normal coated, transverse to the plasma incidence plane, micro-cantilever. It experienced a *positive* deflection with respect to the uncoated micro-cantilever, which was strictly straight. These deformations have never been observed when coating micro-cantilevers with other materials, such as Co, W or Au, where the micro-cantilevers always maintained their straight geometry. This as-coated micro-cantilever also exhibited an optical anisotropy, see white inset in this picture. After annealing (see right picture), the deflection dramatically changed, undergoing an opposite *negative* deflection. This noteworthy variation of the deflection evidenced the longitudinal strain, elongation, that the deposited film produced on the micro-cantilever and the contraction of the film produced by the annealing. However the optical anisotropy, both white insets in Fig. 3, remained after this heat treatment..As the deposited films likely had nano-crystalline structures, the growth of the Ni_2MnGa crystalline phase during annealing is probably the origin of this extremely high contraction. The ability of these micro-devices to detect the change in geometry of these nano-thick Ni_2MnGa films can be proven. The right picture also shows the high contraction of the micro-cantilever when it was cooled to –46 °C. The change of the micro-cantilever deflection by annealing was concurrent with the appearance of the ferromagnetism. The other inset in the right picture of Fig. 3 shows the in-plane MOKE hysteresis loops of a coated glass piece that underwent deposition and post-annealing simultaneously with the micro-cantilever. The maximum applied magnetic field was 40 Oe. It confirmed the generated in-plane magnetic anisotropy perpendicular to the plasma incidence plane.

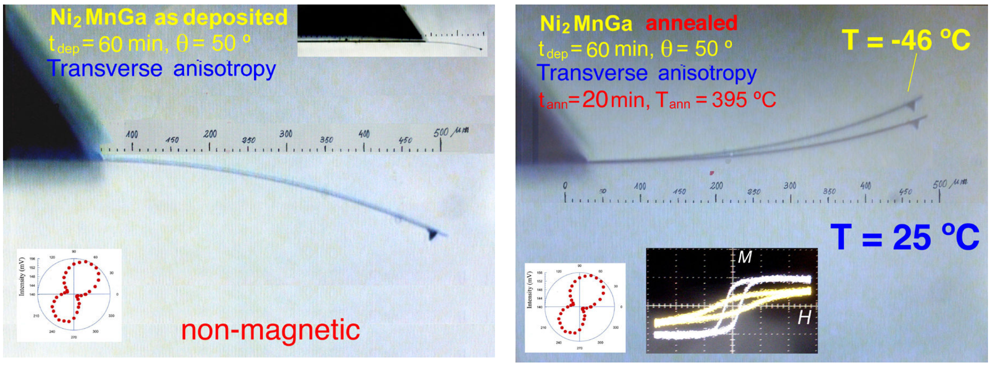

Fig. 3: Left: image of an off-normal coated micro-cantilever. A *positive* deflection with respect to the virgin micro-cantilever, strictly straight, is observed. Right: the annealed micro-cantilever that has undergone a *negative* deflection. Two different situations of the annealed coated micro-cantilever are shown, the micro-cantilever at T_{room} and at –46 °C. Inset: in-plane anisotropic MOKE hysteresis loops of a glass substrate coated by PLD and annealed simultaneously with the micro-cantilever. $H_{max.}$= 40 Oe. The optical anisotropies of this as-coated and post annealed glass are shown in both white insets.

Fig. 4 (left) shows the cooling-heating dependencies of the deflection for an off-normal coated micro-cantilever after annealing. It revealed the possibility of measuring structural deformation of FSMA thin films, on the order of 60 nm in thickness, by using micro-cantilevers as substrates without removal. Fig. 4 (right) displays the cooling-heating curves of the resistance for a coated glass substrate with a Ni_2MnGa film that was grown by PLD and post-annealed simultaneously with the micro-cantilever.

It was observed that the deviation of the linear response of the resistance between ≈230 and 275 K when heating was practically concomitant with the deviation from linearity of the micro-cantilever deflection. These hysteretic behaviours, which revealed the starting and final temperatures for the martensitic transformation of these thin films, indicated that the mechanical behaviour of the micro-cantilevers can be used for studying the structural transformations of these FSMA thin films.

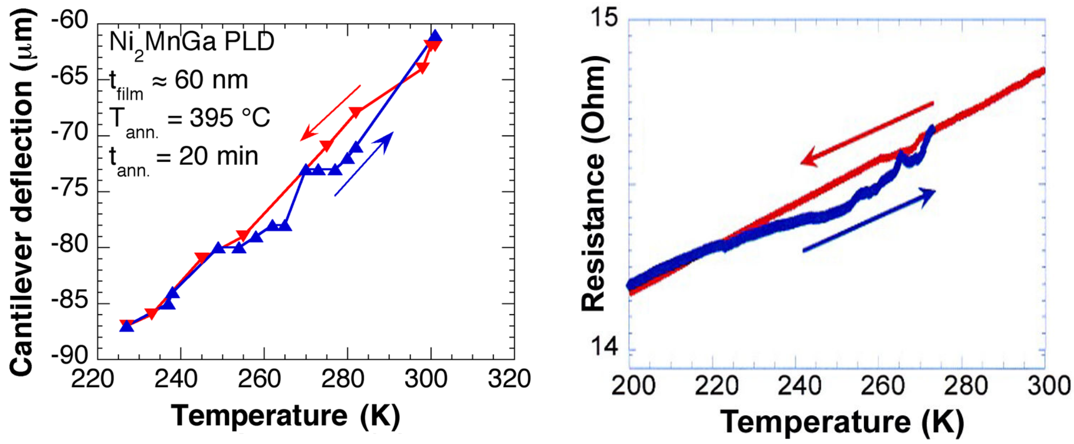

Fig. 4: Left: cooling-heating dependencies of the deflection for a coated and post-annealed micro-cantilever. Right: cooling-heating curves of the resistance for a glass substrate coated by PLD and post-annealed simultaneously with the micro-cantilever.

Summary

It has been shown the PLD of NiMnGa thin films with thicknesses between 30 and 60 nm onto Si micro-cantilevers and glass substrates. The generation of a controlled in-plane magnetic anisotropy has been demonstrated in these glasses and micro-cantilevers coated with NiMnGa FSMA thin films. The mechanical behaviour of these coated and post-annealed micro-cantilevers permitted observing the martensitic transformations of these FSMA thin films. It has been shown that the use of these micro-cantilevers as substrates, with geometrical parameters bound to their mechanical ones, is an advantage for studying martensitic transformations of thin films less than 100 nm in thickness

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