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Ni–Mn–Ga films on Si, GaAs and Ni–Mn–Ga single crystals by pulsed laser deposition

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Abstract

We report the deposition of thin films of the magnetic shape-memory (MSM) alloy Ni–Mn–Ga on different substrates by pulsed laser deposition (PLD). Both standard semiconductors – Si and GaAs – and Ni–Mn–Ga single crystals were used. The films on silicon had a relatively smooth surface and showed large saturation magnetizations, up to 60% of the bulk value, when deposited at substrate temperatures between 500 and 600 °C. The films on GaAs, on the contrary, were non-ferromagnetic with a granular surface. In addition, preliminary results of films deposited on Ni–Mn–Ga single crystals indicate that the Ni–Mn–Ga film exhibits the same 6% strain in the magnetic field as the bulk.

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Keywords: Magnetic shape-memory effect; MSM; Ferromagnetic shape memory; FSMA; Ni–Mn–Ga; Pulsed laser deposition; Thin film

1. Introduction

Magnetic shape-memory (MSM) alloys are promising candidates for providing new functionalities in microelectronics and magnetic devices [1] due to the large strain they exhibit in a magnetic field [2–4]. So far, the best MSM materials are Ni–Mn–Ga alloys which can undergo up to 10% shape changes in an applied field [4].

To realize microscopic structures in a small volume, MSM materials have to be in the form of thin films. Ferromagnetic Ni–Mn–Ga films have already been produced by molecular beam epitaxy (MBE) [5–8], magnetron sputtering [9,10], and pulsed laser deposition (PLD) [11,12] but to our knowledge the MSM effect has not yet been observed in such films.

We have prepared thin Ni–Mn–Ga films by pulsed laser deposition on silicon, GaAs and single-crystalline pieces of Ni–Mn–Ga. The aim was to select an optimal material to produce films with a large saturation magnetization, a smooth surface, and the desired crystal structure. In addition, Ni–Mn–Ga single crystals were used in order to demonstrate the production of martensitic Ni–Mn–Ga films. Such films are good candidates to exhibit the MSM effect since the substrate allows free shape changes of the film material.

2. Experimental

The films were deposited in an ultra-high-vacuum chamber by ablating single-crystalline Ni–Mn–Ga

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targets (diameter 30 mm, thickness 3 mm) with 300 mJ laser pulses at 248 nm (Lambda Physik COMPex 205 KrF excimer laser). In these experiments, the laser-spot size on the target was approximately 3 mm^2 and the fluence was adjusted to ≈ 3 J/cm^2. A typical film thickness was 200–300 nm, which required $\sim 40\,000$ laser pulses. The targets were mounted in a rotating holder to ensure uniform wear of the surface. Throughout the experiments, we used a fixed target–substrate distance of 50 mm.

In the case of silicon and GaAs substrates, small pieces were cut from 500 μm thick Si(1 0 0) or 625 μm thick GaAs(1 0 0) wafers. The substrate temperatures ranged from 450 to 650 °C. The depositions were carried out both in vacuum (base pressure $< 5 \times 10^{-6}$ mbar) and at various argon background pressures (3×10^{-4}–5×10^{-3} mbar). When using Ni–Mn–Ga single crystals as substrates, the deposition temperature was kept between the martensitic transformation temperature (≈ 40 °C) and the Curie temperature (≈ 100 °C). All these experiments took place in vacuum.

The surface quality of the films deposited on Si and on GaAs was studied with a LEO-1450 scanning electron microscope (SEM). The chemical composition of these films was determined using energy-dispersive spectroscopy (EDS) and the crystal structure was studied with a Philips X'pert X-ray diffractometer using Kα radiation from a Co source. The magnetic properties of the films were measured with a vibrating sample magnetometer (VSM) in fields up to 1.43 T. The films on Ni–Mn–Ga single crystals were analyzed with an optical microscope to study the motion of the twin variants.

3. Results and discussion

Fig. 1 shows SEM images of Ni–Mn-Ga films deposited on silicon (Fig. 1a) and on GaAs (Fig. 1b). The film prepared on silicon has a relatively smooth surface whereas in the case of GaAs substrate, the film surface is granular. This observation implies that chemical reactions occur in the GaAs/Ni–Mn–Ga interface. To prevent this, chemically inert lattice-matched buffer layer is required between GaAs and Ni–Mn–Ga as suggested in Refs. [5–8]. However, Fig. 1a and b show that the density of micron-sized droplets on the surface is low enough such that micrometer-sized structures can be patterned in the films. EDS measurements gave Ni/Mn/Ga compositions of the order of 46/31/23, which proves that stoichiometric material transfer from the target (composition 48.9/30.8/20.3) has taken place.

Fig. 2 presents in-plane magnetization curves of Ni–Mn–Ga films deposited in vacuum on silicon at 550 °C and on GaAs at 450 °C. In the case of silicon, the curve is square-like and narrow suggesting the presence of a single magnetically soft phase. On the other hand, the film prepared on GaAs is strongly paramagnetic. Using an approximate film thickness of 300 nm, an area of 0.5 cm^2, and the density of bulk Ni–Mn–Ga (8 g/cm^3), we obtain a saturation magnetization of approximately 34 emu/g ($\approx 60\%$ of the

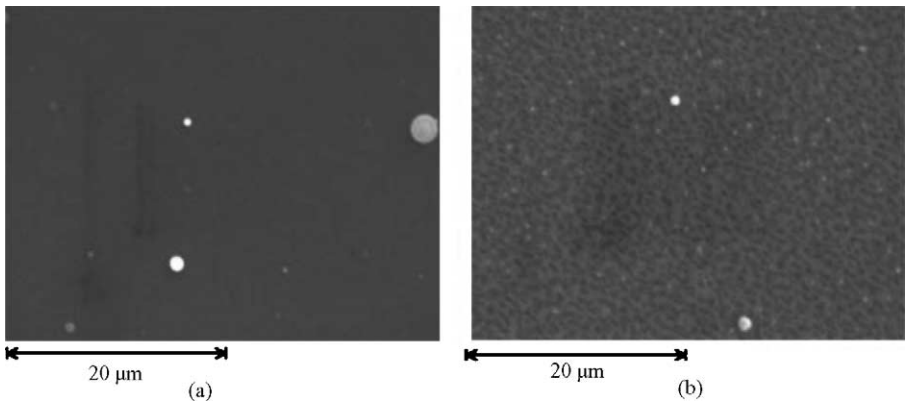

Fig. 1. Scanning electron microscope images of the surface of Ni–Mn–Ga films deposited on (a) Si and (b) GaAs.

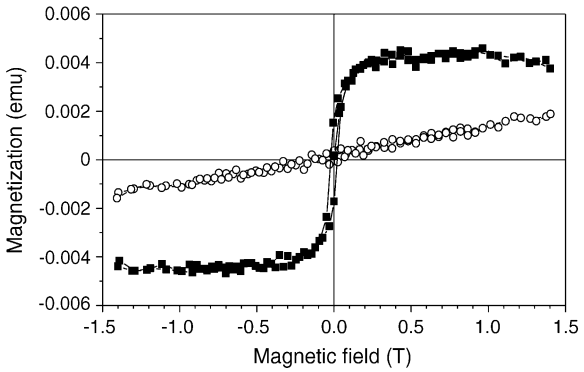

Fig. 2. Magnetization as a function of the applied magnetic field B for a Ni–Mn–Ga film deposited on Si (solid squares) on GaAs (open circles). The curves are measured at room temperature by applying the field in the film plane.

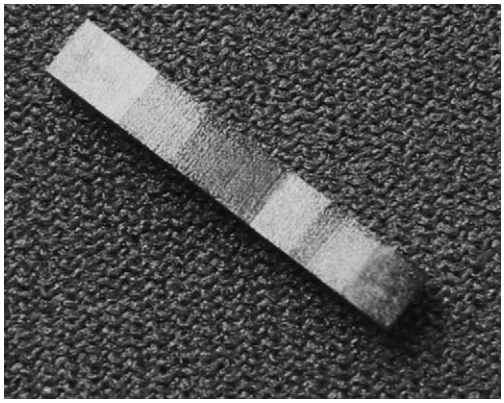

Fig. 4. Optical microscope image of a Ni–Mn–Ga film deposited on a Ni–Mn–Ga single crystal. Dark and light areas are two different twin variants, the proportions of which change when applying stress.

bulk value) for the film prepared on Si. This number is almost six times higher than the values reported in Refs. [11,12].

X-ray diffractograms of two Ni–Mn–Ga films prepared on silicon with different thicknesses (100 and 300 nm) are shown in Fig. 3a. The peaks labelled as austenite in the pattern allow one to identify an $L2_1$ cubic phase with a lattice constant of 0.580 nm – close to that of bulk austenite (0.584 nm) [13]. The additional peaks in the diffractogram suggest that an intermediate $L2_1$ layer with a lattice constant of 0.539 nm has formed between the austenite and Si possibly due to the non-perfect lattice match.

Despite a better matching between the austenitic Ni–Mn–Ga and the GaAs lattices, no traces of the austenite were observed in the films deposited on GaAs. The diffractogram in Fig. 3b shows large peaks which may be ascribed to a cubic phase with a lattice constant of 0.62 nm and a strong (1 0 0) texture. The diffraction pattern supports the notion that some reactions occur between the substrate and Ni–Mn–Ga.

Optical microscope images of Ni–Mn–Ga films deposited on Ni–Mn–Ga substrates indicate that the material has transformed to the martensitic state when cooled to room temperature after the deposition. When applying a magnetic field of 0.5 T on the sample, the full 6% strain corresponding to a change from one variant state to another was observed indicating that the film exhibits the same behavior as the bulk. Fig. 4 shows an intermediate state where both of the variants are visible. These preliminary results suggest that it is possible to produce martensitic Ni–Mn–Ga films by PLD. We will continue analyzing these films and report detailed results later.

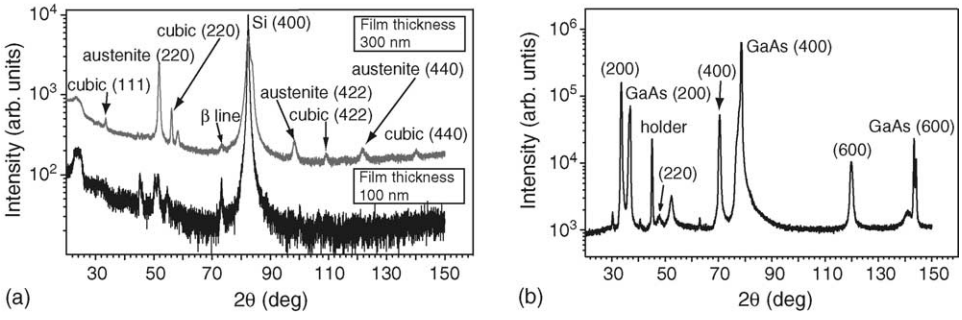

Fig. 3. X-ray diffractograms of (a) two Ni–Mn–Ga films on silicon with thicknesses 100 and 300 nm and (b) a Ni–Mn–Ga film on GaAs.

In conclusion, we have prepared thin Ni–Mn–Ga films on different substrates by pulsed laser deposition. We have found proper values for all the necessary deposition parameters to produce ferromagnetic films with a good surface quality and a large saturation magnetization, up to 60% of the bulk value, when using silicon as a substrate. In addition, we have discovered that a lattice-matched buffer layer is needed between GaAs and Ni–Mn–Ga to prevent chemical reactions in the film–substrate interface. The results obtained with Ni–Mn–Ga films on Ni–Mn–Ga single crystals reveal that the film exhibits the same 6% strain in the magnetic field as the bulk. This gives us a reason to assume that the magnetic-field-induced strain could also occur in the film when the substrate is lithographically removed.

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