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Pulsed Laser Deposition Of Thin Films Of Various Full Heusler Alloys Co_2 mnx (X = Si, Ga, Ge, Sn, SbSn) At Moderate Temperature

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Abstract: *This work presents pulsed laser deposition of cobalt-based Heusler thin films Co_2MnX (X = Si, Ga, Ge, Sn, SbSn) on different substrates (Si, GaAs, InAs). The deposition processes developed in vacuum (about 10^{5} Pa) to avoid oxidation of the films and targets. The temperature of the substrates during the depositions was kept below 500 K to minimise interface inter diffusion. From X-ray diffraction, we found that the films are crystalline and slightly oriented. The stoichiometric composition of the films was further checked by EDS, while the size and density of droplets were determined by SEM. The magnetic properties of the films are consistent with those of the bulk material used as target.*

Keywords: Pulsed laser deposition; Ferromagnetic alloys; Crystallised Heusler thin film

1. INTRODUCTION.

The Heusler and half-Heusler alloys X_2MnZ and respectively XMnZ [1] build a family of magnetic materials, with particularly interesting properties as regards to spin device applications [2,3].

Important requirements for spintronic devices are the efficient injection of spin-polarised carriers into a semiconductor and at least 300 K operation. Efforts are directed to find the appropriate combination ''material-technique'' to satisfy these requirements. One of the attractive choices would be half-metallic ferromagnets lattice-matched to semiconductor structures. The Coseries of Heusler alloys have attracted considerable interest because of their high Curie temperatures and the possibility that some of them may also be half-metallic [4]. Many of those alloys have not been fully explored and only a very limited number have been grown on semiconductor substrates, by MBE [5–7] and by sputtering [8]. It is important that good stoichiometry, smooth interfaces, and a high crystalline quality of the films are all achieved by an appropriate deposition technique. We attempted to grow a series of Co_2MnX by pulsed laser deposition (PLD). Despite the high ablation thresholds and low growth rates, PLD offers a range of advantages over the conventional deposition techniques used with non-equilibrium metallic alloys: the opportunity to easily act on the deposition parameters or on target composition. Our work presents experiments to deposit thin films of Co_2Mn(Si, Ga, Ge, Sn, SbSn) on different semiconductor substrates (Si, GaAs, InAs) using a KrF excimer laser Lambda Physik LPX 220 I (l = 248 nm, t= 20 ns) delivering up to 400 mJ/pulse at frequencies ranging from 2 to 10 Hz and fluences of 0.5–5 J/cm^2. The deposition processes developed in vacuum (about 10^{5} Pa) to avoid oxidation of the films and targets. The temperature of the substrates during the depositions was kept below 500 K to minimise interface interdiffusion. From X-ray diffraction, we found that the films are crystalline and slightly oriented. The stoichiometric compositions of the films were further checked by EDS, while the size and density of droplets were determined by scanning electron microscopy (SEM). The magnetic properties of the films are consistent with those of the bulk material used as target.

2. EXPERIMENTAL.

2.1. TARGET ELABORATION.

Near-single crystalline Heusler samples have been grown by the vertical gradient freeze (VGF) method, in argon [9]. High purity powders of the constituting elements Co, Mn, Si, Sn, Ga, Sb (Alfa Aesar) were weight in stoichiometric 2:1:1 amounts, mixed together by manual grinding, then pressed into pellets under 6 t and annealed 60 min at 1000 8C. The pellets thus obtained make the batch for VGF in a RF furnace (MRS 2, Cambridge Ltd.). In each case, the melting temperature has been chosen according to the phase diagrams of the corresponding binary compounds due to the lack of ternary phase diagrams for Co_2MnX (X = Si, Ga, Ge, Sn, $Sb_{0.8}Sn_{0.2}$). A plateau of about 180 min at the melting point has been kept in each run, then cooling down to room temperature followed at a rate of 4 8C/min. The samples are cut into several slices (about 5 mm thick) perpendicularly to the axis of the ingot. Previous to be use for PLD, the targets are prepared according to the following protocol: polishing with abrasive paper (grain size 800), polishing using silicon carbide on a glass plate, rinsing for 10 min in distilled water, polishing with Al_2O_3 (200A) on felt, boiling in acetone for 20 min, rinse in methanol in a US bath, final rinse in cold acetone.

2.2. PLD SET UP AND THIN FILMS GROWTH.

Ablation is performed in a stainless steel vacuum chamber [10] (working pressure 10^{5} Pa). The substrates were polished (1 0 0) silicon wafers and III–V narrow gap semiconductors as we already mentioned. Ablation/deposition processes were run using a KrF excimer laser (Lambda Physik LPX 220 I, 248 nm, t = 20 ns) delivering up to 400 mJ/pulse at a repetition frequency of 1–10 Hz. Our experiments were carried out at 7 Hz for 20,000–50,000 pulses. To select the most homogeneous part of the beam, we use a mask allowing 90% of the laser energy to be employed. After having been reflected on a 99% high-energy laser mirror, the laser beam enters the stainless steel vacuum chamber through a quartz window. It is focused through a 150 mm focal lens on a rotating target under an incidence angle of 458. The laser impact is a 1 mm^2 rectangular surface (4 mm 0.25 mm). The substrate is positioned parallel to the target at a distance of 40 mm [11] and heated up to 473 K during the depositions. In order to improve the morphology of the films we performed a study to optimise the laser fluence. The density of incident energy onto the surface has been varied between 3 and 5 J/cm^2. As it is seen in Fig. 1, the best results were obtained with 3 J/cm^2, which has been employed for all the PLD experiments carried out with Co_2MnX (X = Si, Ge, Ga, $Sb_{0.8}Sn_{0.2}$, Sn). The process conditions for the series of PLD Heusler films we have produced are summarized in Table 1. Except the compounds with gallium and tin– antimony, whose characterisation is ongoing, the other three ones have been fully investigated for structure, morphology and magnetic properties.

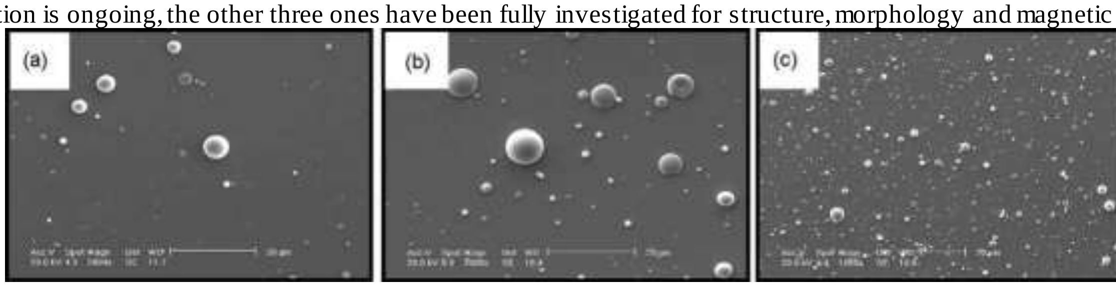

Fig. 1. SEM study of the surface morphology of Co_2MnSi films deposited on Si substrate at different fluences: (a) 3 J/cm^2, (b) 4 J/cm^2 and (c) 5 J/cm^2.

Table 1
Co-based Heusler PLD thin films series and process conditions

Samples	Target	Substrate	Conditions of deposition		
			Fluence (J/cm^2)	Temperature (K)	Thickness (nm)
FHSi 1	Co_2MnSi	Si(1 0 0)	3	473	300
FHSi 2	Co_2MnSi	Si(1 0 0)	4	473	350
FHSi 3	Co_2MnSi	Si(1 0 0)	5	473	400
FHSi 4	Co_2MnSi	GaAs(1 0 0)	3	473	160
FHGa	Co_2MnGa	Si(1 0 0)	3	473	160
FHGe	Co_2MnGe	GaAs(1 0 0)	3	473	160
FHSbSn	$Co_2MnSb_{0.8}Sn_{0.2}$	Si(1 0 0)	3	473	160
FHSn 1	Co_2MnSn	InAs(1 0 0)	3	473	160

The films have been deposited on both lattice- and not lattice-matched substrates, like for instance Co_2MnSi/GaAs (FHSi 4) 0.012% or Co_2MnSi/Si (FHSi 1–3) 3.96% [12].

2.3. CHARACTERISATION.

Structural characterization of the samples has been performed by X-ray diffraction measurements and complementary by energy dispersive X-ray spectroscopy (EDS). The X-ray diffraction patterns were recorded in a classical u–2u coupled mode, with a 2u step of 0.038 at 5 s per step, using a Siemens-Brucker D5000 equipment (copper X-ray source, l = 1.5406 A). The diffractometer holds a secondary monochromator and a rotating sample holder.

SEM images were taken with a Philips XL 30 SEM equipped with an EDS microanalysis system. The back-scattered electron (BSE) configuration was employed to enable a better observation of chemical in homogeneities [13].

Magnetization measurements have been carried out on an Oxford Instruments vibrating sample magnetometer. Magnetization (M) was measured versus applied field (H) up to 2 T at 50 K intervals from 300 to 10 K. The slope of M versus H well above the saturation field was used to correct for the diamagnetic contribution from the substrate and sample holder. The saturation

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magnetization in units of Bohr magnetons per formula unit was calculated by assuming a smooth surface and using a uniform film thickness determined by cross-sectional SEM or profilometry. The thickness of the films depends, in principal, on the pressure in the chamber, the ablation rate of the target material and the number of laser shots employed. In our study, the pressure was 10 ^{5} Pa in all runs. Having in mind that we have used various CO_2MnX compositions, we accounted for the ablation rates accordingly and varied the number of laser shots between 20,000 and 50,000 such a way to obtain, in the given deposition conditions, thick-ness values ranging from 160 to 400 nm (see Table 1).

3. RESULTS AND DISCUSSION.

The structural investigations show a good agreement between the Co-based Heusler targets and the corresponding PLD films. We give, for example, the XRD patterns of Co_2MnSi bulk and thin films compared with the theoretical spectrum calculated from structure data (Fig. 2). The influence of the Si and GaAs substrates on the respective films' quality is obvious. The lattice match between Co_2MnSi and GaAs made the corresponding film to grow smoother and better ordered, although texturing is also observed on the film grown onto silicon [14]. The compositions of the films were analysed by EDS and compared with the values obtained from both the fresh and the ablated surfaces of the targets.

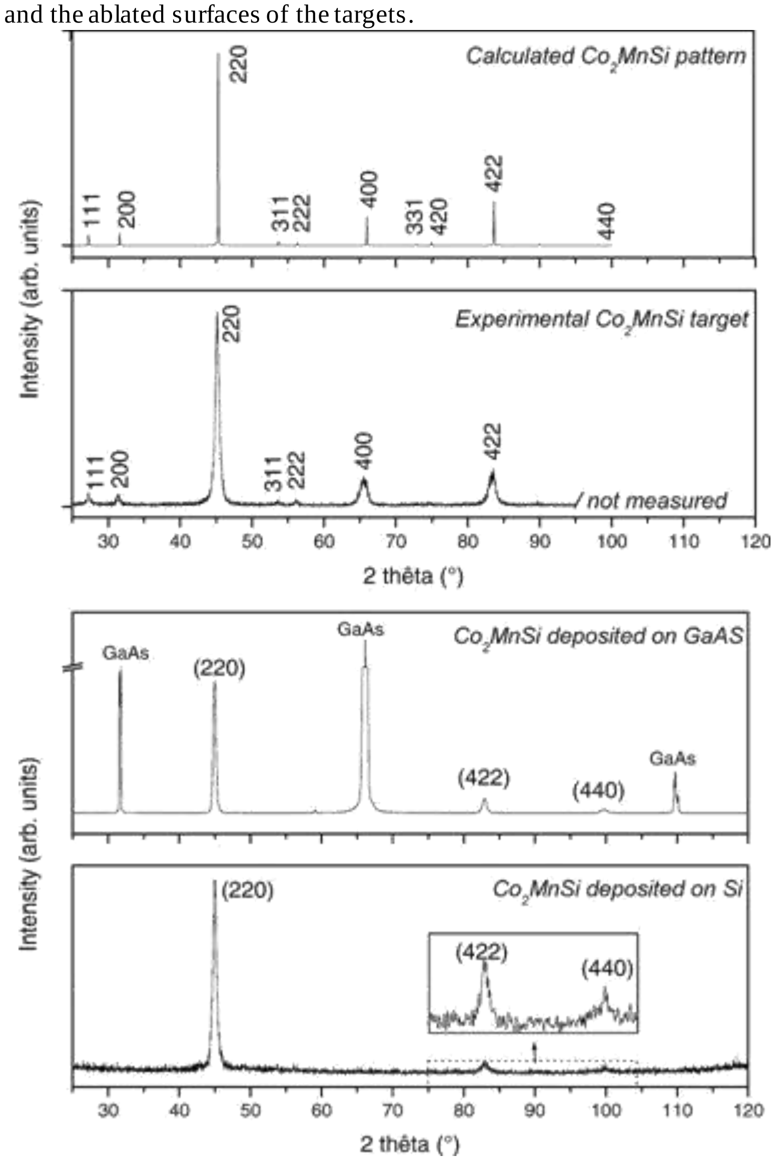
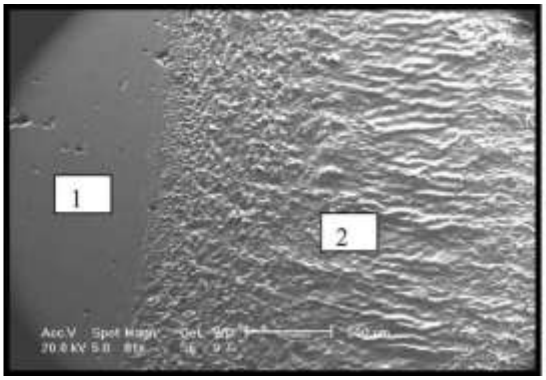


Fig. 2. Co_2MnSi XRD patterns: comparison between the experimental spectrum of the target and the one calculated from structural data, the spectra of the films FHSi 4 and 3.

The results show that the stoichiometry's of the targets are very well reproduced in the ones of the films (see, for example Fig. 3). This affirmation is also supported with the results obtained from the magnetic measurements shown in Fig. 4a and b. One can notice the similarity between the evolution of the saturation magnetization of the thin films and bulk up to 150 K for all films except the one grown on lattice-matched substrate (Fig. 4a).



Target	%at. Co	%at. Mn	%at. Si
1 (fresh)	48	25	27
2(ablated)	48	25	27
Film FHSi4	53	22	25

Fig. 3. SEM image of the Co_2MnSi target showing the (1) fresh and (2) ablated surfaces and their respective microanalyses compared with that of the corresponding film.

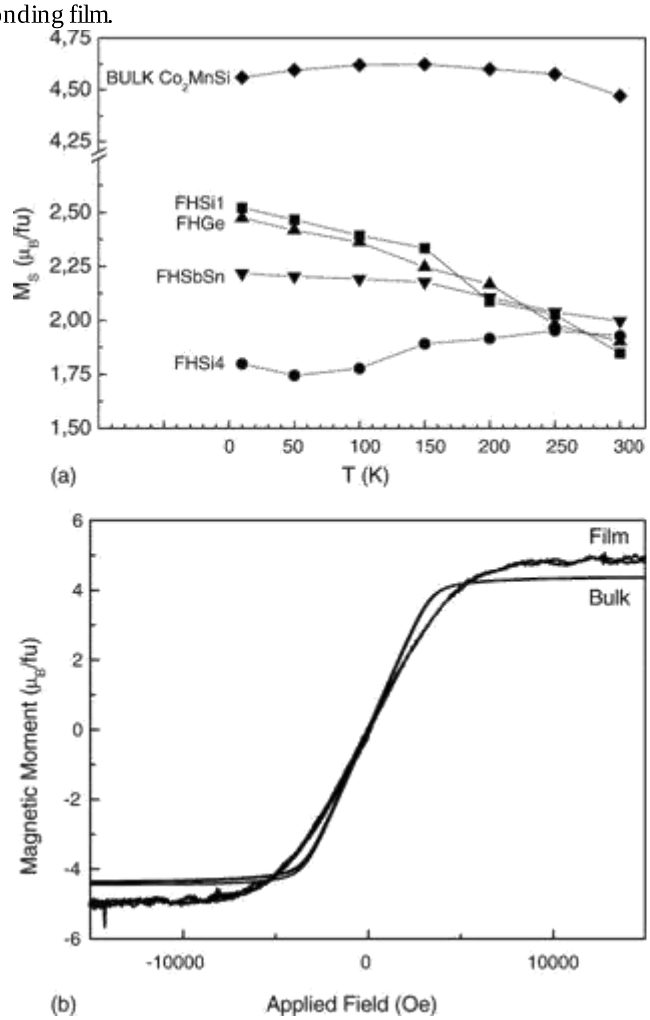


Fig. 4. (a) Magnetic properties of Co_2MnX thin films (the labels correspond to those in Table 1) and (b) magnetic moment vs. the applied field— comparison between the values of the Co_2MnSn target and the PLD thin film onto a lattice-matched substrate (InAs).

The curve corresponding to that film (FHSi 4) keeps the correspondence with the bulk at higher temperatures too. The lower M_S value of the film FHSi 4 in comparison with the bulk could possibly arise from some magnetic moments disorder in the film. The measured moment of the target Co_2MnSn (Fig. 4b) corresponds to a zero temperature moment of 4.3– 4.6mB/fu. Within the accuracy of the measurement, the saturation magnetization of target and film FHSn 1 on InAs (also lattice-matched) is close to those of the single crystals [15]. The influence of the film/substrate lattice mismatch on the magnetic properties of the film is thus obvious: the values for H_c correlate with the structural properties, so that values closer to the ones measured for the bulk are expected for better film/ substrate lattice-mismatch.

4. CONCLUSIONS.

We have prepared a series of PLD Heusler films in the system Co_2MnX (X = Si, Ga, Ge, Sn, SbSn) aiming at being employed as magnetic contacts for spintronic applications. The structural properties of the films show high crystalline quality, especially for the samples lattice-matched to the substrates. The morphology study in connection with the laser fluences employed in the PLD runs show that lower fluence values are more in favour of less droplets on the film's surface in the case of the Heusler compounds we have studied. The initial stoichiome-tries of the targets are preserved during the PLD process and transferred in the deposited thin films for all the samples in the investigated series, which is shown by the EDS analyses of fresh and respectively ablated surfaces of the targets and also reflected by the magnetic measurements performed on both targets and corresponding films. The films lattice-matched to the substrates show magnetic moment values close to the ones of the corresponding targets and comparable with those for single crystalline material. We believe that this is witness of bulk-like ordering in our PLD films.

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