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High-temperature ferromagnetism of $Si_{1-x}Mn_x$ ($x \approx 0.52-0.55$) alloys

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ABSTRACT

The paper reports on the comprehensive study of properties of nonstoichiometric $Si_{1-x}Mn_x$ alloys slightly enriched in Mn ($x \approx 0.51$–0.55) as compared to the stoichiometric monosilicide MnSi. Mosaic type $Si_{1-x}Mn_x$ films 55–70 nm in thickness were produced by the pulsed laser deposition (PLD) method onto the single crystalline Al_2O_3 substrates at 340 °C. The Curie temperature T_C in nonstoichiometric $Si_{1-x}Mn_x$ ($x \approx 0.52$–0.55) films exceeds room temperature, while in their stoichiometric counterpart, MnSi, the T_C value does not exceed ≈ 30 K. The consistent data on anomalous Hall effect and transverse Kerr effect prove the global character of ferromagnetic (FM) order caused by magnetic defect formation rather than the presence of FM clusters. At Mn content $x \le 0.55$, the magnetization data testify to a good homogeneity in the distribution of magnetic defects without their segregation: variations of the saturation magnetization M_s do not exceed 6% in the temperature range $T = 10$–100 K and are well described by the Bloch law. It is also revealed that textured high-quality $Si_{1-x}Mn_x$ films with $x \approx 0.52$ and $T_C \sim 300$ K could be formed by PLD method in the "shadow" geometry (at lower energy of deposited atoms).

1. Introduction

Si–Mn alloys attract an increasing interest for application in spintronic as integrated-circuit elements easily incorporated into the existing microelectron technology [1] and also exhibit unusual magnetic and transport behavior [2–5].

The fabrication of Si–Mn alloyed thin films with a uniform distribution of magneto-active component presents a challenge. Indeed, most of the studies of $Si_{1-x}Mn_x$ system have been performed on the samples with relatively low content of manganese ions ($x < 0.05$) incorporated by ion-implantation (see [1–4] and references therein) and FM ordering above room temperature (RT) has been revealed. However, these dilute alloys turn out to be strongly inhomogeneous materials due to their phase segregation, with isolated magnetic $MnSi_{1.7}$ nanoparticles ($x \sim 0.35$) precipitated in the Si matrix [1,2].

We believe that concentrated nonstoichiometric $Si_{1-x}Mn_x$ alloys with composition close to the MnSi stoichiometry are more promising for spintronic applications than dilute $Si_{1-x}Mn_x$ alloys. Further still, in Refs. [6,7] we have found that in $Si_{1-x}Mn_x$ alloys with small Mn excess ($x = 0.52$–0.55), the Curie temperature increases more than an order of magnitude ($T_C \ge 300$ K) as compared to MnSi ($x = 0.5$, $T_C \approx 30$ K). The studied alloys also exhibit anomalous Hall effect (AHE) at RT indicating the spin polarization of carriers. These peculiarities have been qualitatively interpreted in terms of a model of defect-induced FM order in metallic alloys with local magnetic moments (LMMs) on the defects coupled following the spin-fluctuation mechanism [4].

Below we present a comparative study of the properties of (1) mosaic (polycrystalline) $Si_{1-x}Mn_x$ ($x \approx 0.52$–0.55) films and (2)

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$Si_{1-x}Mn_x$ films ($0.5 < x \le 0.52$) with variable stoichiometry (gradually changing across the sample) and with much better structural quality films. The first series of films was fabricated by traditional pulsed laser deposition (PLD) technique in the ordinary ("direct") geometry when the working surface of the substrate is subjected to the Si–Mn plume produced by laser. While the second series of films was fabricated recently by an alternative PLD technique employing the "shadow" geometry and using Kr as a buffer gas.

2. Sample preparation and experimental details

$Si_{1-x}Mn_x$ films 55–70 nm in thickness (with Mn content $x=0.44$–0.6) were produced by the PLD technique employing time-of-flight separation of the ejected particles [8]. Si–Mn films were grown on Al_2O_3 (0001) substrates under vacuum conditions ($< 10^{-6}$ mbar) at $T=340$ °C at the rate of 1.5 nm/min. The investigation of magnetic, transport and magneto-optical properties was performed on the same samples fabricated in the shape of the Hall bar having the conduction channel with the width $W=1.2$ mm, the length $L=4$ mm with the distance between potential probes $l=1.4$ mm.

The structural features of the films were studied by X-ray diffraction analysis without a monochromator and a diaphragm before the detector. The chemical composition of the films was determined by X-ray photoelectron spectroscopy (XPS). The measurements were performed after cleaning of the film surface for 60 s by argon ions with energy of 2 kV. The thickness of the removed layer was about 5–10 nm.

The magnetization of $Si_{1-x}Mn_x$ samples was measured by VSM Lake Shore 7407 vibrating-sample magnetometer at applied magnetic field up to 15 kOe within the 90–400 K temperature range. Study of the magnetization in the low temperature region, at $T < 100$ K, was performed using the MPMS-3 (SQUID-VSM) magnetometer in fields up to 40 kOe. Uncertainty of measurements did not exceed 0.2%.

Details of AHE and transverse Kerr effect (TKE) measurements were described in [6,7].

Alternatively, $Si_{1-x}Mn_x$ film with slightly variable composition was grown on Al_2O_3 (0001) substrate at elevated temperature $T=340$ °C by PLD in the shadow geometry in Kr buffer gas under pressure $P \sim 10^{-2}$ mbar (see Fig. 1). The $Si_{1-x}Mn_x$ film on the rectangular substrate 10×15 mm^2 in size yielded the varying composition $x=0.506$–0.52 at respective thickness $d=270$–70 nm depending on the distance from the stoichiometric MnSi target revealed from Rutherford Backscattering Spectroscopy (RBS) analysis (He^{++}, $E_0=1.7$ MeV). The structural quality of thus grown $Si_{1-x}Mn_x$ film was additionally attested by comparing RBS spectra in random and channeling orientations.

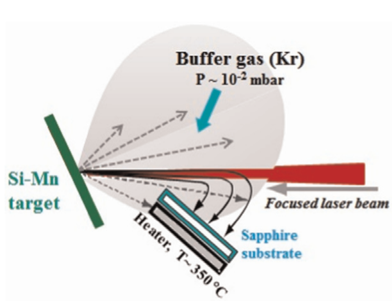

Fig. 1. The scheme of PLD method in the "shadow" geometry.

3. Experimental results and discussion

3.1. Structural characterization

The XPS data were used mainly for the analysis of a relative content of Si and Mn in the $Si_{1-x}Mn_x$ films: $y=(1-x)/x$ [6]. To calculate y values, we used the data reported in [9]. The XPS analysis of chemical composition of the films allowed us to select for our studies $Si_{1-x}Mn_x$ the samples with different Mn content, $x=1/(1+y)=0.44$–0.63. The AHE study in these samples shows that only films with a small Mn excess ($x \approx 0.52$–0.55) exhibit FM order with the Curie temperature about RT [6].

XRD data for $Si_{1-x}Mn_x/Al_2O_3(0001)$ sample with $x \approx 0.52$ are shown in Fig. 2. The XRD pattern contains intense reflection peaks from Al_2O_3(0006) for the Cu$K_{\alpha1}$, Cu$K_{\alpha2}$, and Cu$K_{\beta1}$ lines. In addition, there is a broad peak at $2\theta=44.43°$ related to MnSi (100) plane. The structural perfection of the film is described by the FWHM ($\Delta\omega \approx 0.4°$) of the peak $2\theta=44.43°$. Such a broad peak observed for the film is a signature of a pronounced mosaicity of this structure (see inset to the left of Fig. 2).

The XRD pattern taken from $Si_{1-x}Mn_x/Al_2O_3(0001)$ sample prepared by PLD in "shadow" geometry is shown Fig. 3a, while the RBS spectra measured in the center of the sample is given in Fig. 3b. XRD results clearly demonstrate the presence of ε-MnSi phase with B20 structure. The FWHMω parameter is $\Delta\omega \approx 0.2°$, i.e. is twice less than for the films prepared by "direct" PLD approach. The differences in XRD patterns are confirmed by scanning electronic microscopy (SEM) inspection revealing that there is no film mosaicity in the "shadow" PLD fabricated $Si_{1-x}Mn_x$ film (cf. insets of Fig. 2) apparently due to the lower energy of deposited atoms in the latter case. Random vs. channeling RBS spectra also indicate some ordering normal to the substrate surface in the "shadow" PLD fabricated $Si_{1-x}Mn_x$ film (Fig. 3b).

3.2. Magnetization

The temperature dependence of the saturation magnetization $M_s(T)$ for sample with the low excess of Mn ($x \approx 0.52$) is shown in Fig. 4a. A single magnetic phase is observed for this sample. In such case, an accurate fit is assured through the use of the function

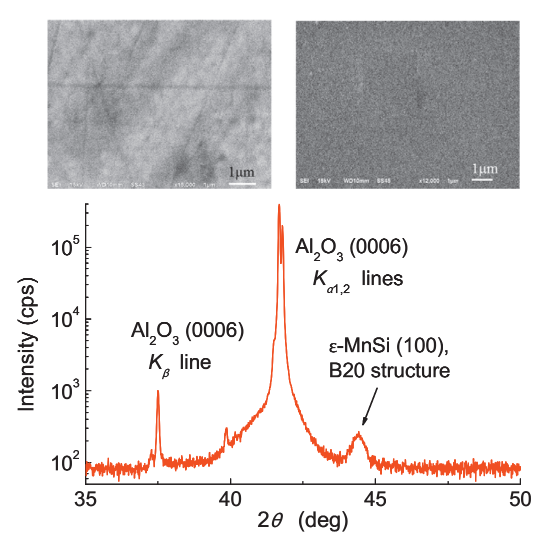

Fig. 2. The X-ray diffraction pattern of $Si_{1-x}Mn_x/Al_2O_3$ structure with $x \approx 0.52$ prepared by "direct" PLD method. The insets show SEM images of the surface $Si_{1-x}Mn_x$ films fabricated in "direct" (left) and "shadow" geometries (right).

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