PHYSICS, CHEMISTRY AND APPLICATION OF NANOSTRUCTURES, 2009

STRUCTURE AND MAGNETIC PROPERTIES OF $Sr_2FeMoO_{6\pm\delta}$ NANOSIZED FILMS

M. A. KALANDA, L. S. LOBANOVSKY
Scientific-Practical Materials Research Center NASB
P. Brovka 19, 220072 Minsk, Belarus
Kalanda@physics.by

L. I. GURSKII, E. V. TELESH, D. A. KOTOV
Belarusian State University of Informatics and Radioelectronics
P. Brovka 6, 220013 Minsk, Belarus

A. SAAD
Al-Balqua Applied University, 11953 Salt, Jordan

B. ADOLPHI, M. PLÖTNER
Dresden University of Technology, D-01062 Dresden, Germany

A technology of $Sr_2FeMoO_{6\pm\delta}$ (SFMO) nanosized films deposition by ion-beam sputtering is described. Optimization of deposition conditions on formation of structurally-perfect SFMO double perovskite films is presented. Several problems arise with the use of the ion-beam sputtering method concerning the films inhomogeneity, the presence of multiple phases and Fe_{Mo} and Mo_{Fe} antistructural defects. It is shown that they are solved by means of complex selection of parameters: substrate temperature, deposition rate and subsequent thermal processing.

1. Introduction

Layered magnetic semiconductors $Sr_2FeMoO_{6\pm\delta}$ (SFMO) having high Curie temperature ($T_c \sim 420$ K), high level of spin polarization (~ 100%), low operating magnetic field (up to 1 T) and conducting ferromagnetic character could be considered among the most promising materials for spintronics [1,2]. These properties of a double perovskite are most interesting for their use in the form of structurally perfect nanosized SFMO films for electrodes, spin valves and magnetic tunneling devices, as well as for spintronic elements in actuators, spin-diodes and spin-transistors operating at room temperature [3].

Still the elaboration of the SFMO films with $T_c \sim 410$-450 K and desirable magnetoresistive and magnetic characteristics is connected with a number of technological problems. It is known that such deposition conditions as substrate temperature (T_s), film deposition rate (v_d), *etc*. are determining factors for the growth of structurally perfect nanostructured SMFO films [4,5]. In this paper, the influence of conditions of ion-beam sputtering deposition of SMFO films on their morphology and magnetic characteristics are reported.

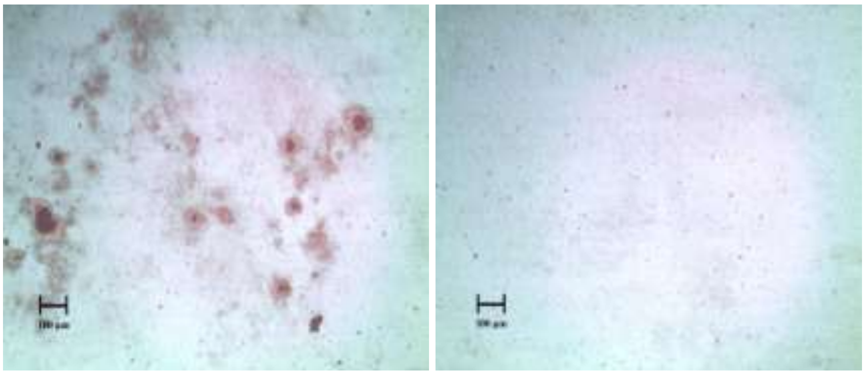
2. Experimental

The deposition of the SFMO films was carried out using the vacuum setup Z-400 ("Leybold-Heraeus" company) equipped with the oil-free pumping system. The vacuum chamber was supplied with a two-beam ion source with a close Hall current on the base of an accelerator with an anodic layer. A specific feature of this source is a possibility of the generation of two ion beams, one of which provides sputtering of the target material, and the other one is used for cleaning of the substrate. Polycore substrates were placed on the holder of the carousel-type at a distance of 100 mm in front of the target surface. After that the chamber was pumped out down to the residual pressure of 10^{-3} Pa. The substrates cleaning was carried out by Ar ions with energies of 300-400 eV. The ion cleaning current was 10 mA and the cleaning time was 10-15 min.

The SFMO targets with diameter of 50 mm and thickness of 4 mm were used as a spray material. The sputtering was carried out by Ar ions of 1800-2400 eV at the current of 30 mA. Phase composition of the materials was analyzed by the X-ray diffraction method using a "DRON-3" setup with a Cu K_{α}-irradiation and ICSD-PDF2 database (Release 2000). Magnetic properties of the samples were studied by using a magnetometer in magnetic fields up to 14 T.

3. Results and discussion

It is known that structure and density of SFMO films depend on the surface diffusion coefficient of the adsorbed material, which first of all is determined by the substrate temperature (T_s). The surface diffusion coefficient (D_s) of the deposited film components depends on the deposition rate (ν_d) as well. At its critical value ν_d relaxation time constant τ tends to infinity. With that $D_s \to 0$ and conglomerates of the deposited material remain in the same place where they have precipitated, which results in the film inhomogeneity. It was determined that at ν_d = 15-20 nm/min and T_s = 120, 280 and 440ºC the SFMO films deposited in Ar environment got a mirror black surface. They are amorphous with high resistivity. At the further T_s increase up to 650ºC, the films become inhomogeneous containing a mixture of different phases of $SrMoO_4$ and $Sr_2FeMoO_{6\pm\delta}$ with high electrical conductivity (Fig. 1a). At $\nu_d \sim$ 9-15 nm/min and T_s=650ºC one observes an increase of the SFMO films density and homogeneity. At the lower deposition rate (ν_d = 7-9 nm/min), the films are characterized by a clearly observed homogeneous dense structure (Fig. 1b).



a b

Figure 1. Surface view of SFMO films deposited on the polycore substrate at T_s= 650ºC and ν_d =15-20 nm/min (a) and ν_d = 7-9 nm/min (b).

According to the X-ray diffraction analysis, the films have a single-phase composition of $Sr_2FeMoO_{6\pm\delta}$. At the same time, the superstructural ordering of Fe^{3+} and Mo^{5+} cations were not observed (Fig. 2).

The magnetization measurements at T = 6 K for the external magnetic filed applied along the film surface (Fig. 3) showed the magnetization saturation M_{sat} ~ 1.6 μ_B/f.u., which is considerably lower than the theoretical value (M_{theor} = 4 μ_B/f.u.). Low M_{sat} is caused by the presence of antistructural defects of Fe_{Mo} and Mo_{Fe} type which disturb the superstructural ordering of Fe/Mo cations and change the orientation of highly hybridized 4*d* t_{2g}-orbitals of Mo^{5+} (S=1/2) cations and 3*d* t_{2g}-orbitals of Fe^{3+} (S=5/2) cations. These defects influence the transport and magnetic properties of SFMO [2,4].

An additional annealing of SFMO films deposited at ν_d = 7-9 nm/min and T_s=650ºC was carried out in vacuumized quartz ampules in the presence of a gettering agent at 900ºC and $P(O_2)=10^{-8}$ Pa during 1 h with a goal of the decrease of Fe_{Mo} and Mo_{Fe} concentration. A sharp increase of M_{sat} up to 3.4 μ_B/f.u. at 6 K is observed. This is indicative of a decrease of Fe_{Mo} and Mo_{Fe} concentrations and the appearance of the superlattice ordering of Fe/Mo cations (Fig. 3).

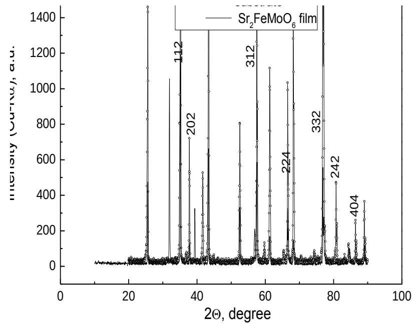


Figure 2. X-ray diffraction spectrum of the SFMO film deposited at $T_s = 650$ºC and ν_d = 7-9 nm/min.

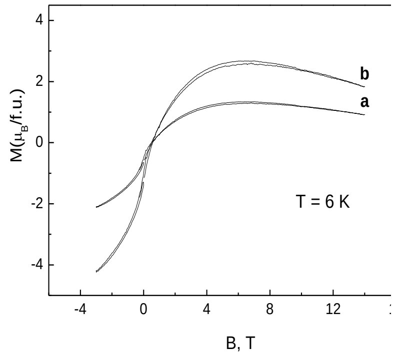


Figure 3. Hysteresis cycles measured for $Sr_2FeMoO_{6\pm\delta}$ films deposited at $T_s = 650$ºC and ν_n = 7-9 nm/min (a) and annealed at 900ºC, $P(O_2)=10^{-8}$ Pa for 1 h (b).

4. Conclusions

It is determined that the structure as well as magnetic properties of the SFMO films strongly depend on their deposition conditions. The SFMO films deposited at ν_d = 7-9 nm/min and $T_s = 650$ºC and then annealed in the presence of a gettering agent at 900ºC and $P(O_2)=10^{-8}$ Pa during 1 h improved structural and magnetic characteristics providing $M_{sat} = 3.4\ \mu_B$/f.u. at 6 K. This is caused by formation of a homogeneous microstructure and single-phase composition of the films and low concentration of Fe_{Mo} and Mo_{Fe} antistructural defects there.

References

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