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MAGNETIC AND MAGNETORESISTIVE PROPERTIES OF Sr₂FeMoO₆₋₈ NANOSIZED FILMS

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Annealing of nanostructured $Sr_2FeMOO_{6-\delta}$ films in an evacuated quartz tube results in variation of concentration of antistructural defects $[Fe_{Mo}]$, $[Mo_{Fe}]$ and spin polarization degree. The films possess low magnetization and large magnetic inhomogeneity.

1. Introduction

Structurally perfect nanosized Sr₂FeMoO_{6- $\delta}$ (SFMO) films are mostly interesting for applications as electrodes for spin valves and magnetic tunnel devices and for spintronic elements functioning at room temperature [1]. At the temperature higher than the Curie point (T_c), SFMO films are found to be in a paramagnetic state and at $T < T_c$ the magnetic ordering takes place, leading to formation of ferrimagnetic structure, were long chains Fe³⁺-O²⁻-Mo⁵⁺ promote superstructural ordering of cations, in condition of the absence of point defects (antisite defects) [Fe_{Mo}] and [Mo_{Fe}] [2,3]. At the presence of antisite defects, cations of iron and molybdenum can be in different spin states (Fe²⁺ and Mo⁶⁺), affecting magnetic structure of SFMO [4]. In this way, ferrimagnetic (FRM) state, antiferromagnetic (AFM) state or mixed magnetic (FRM-AFM) state can be realized depending on the point defect concentration. Thus, a deeper investigation of magnetic disorder in SFMO films permit to control a spin state of cations.}

2. Experimental

Deposition of $Sr_2FeMoO_{6-\delta}$ films has been carried in a vacuum chamber Z-400 ("Leybold-Heraeus" company) equipped with the oil-free pumping system.

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Target sputtering was performed by 600-800 eV. Phase composition of the films were analyzed by the X-ray diffraction method using a DRON-3 system with CuK_{α} radiation and the ICSD-PDF2 database (Release 2000). Magnetic and magnetoresistive characteristics were investigated with an universal technique "Cryogenic Limited" in the temperature range of 4.2-300 K at a constant magnetic field up to 8 T. Temperature dependence of specific magnetization of the SFMO films was measured in the following modes: preliminary cooling down to 4.2 K in the magnetic field (FC-field cooling) or without field (ZFC-zerofield cooling) and subsequent heating in the magnetic field of 0.01 T.

3. Results and discussion

Two types of SFMO films have been investigated: the film No. 1 deposited with the rate $v_n = 8$ nm/min and substrate temperature of 923 K in gaseous Ar medium, and the film No. 2 obtained at the same conditions but with additional annealing at 1173 K for 1 h in an evacuated quartz tube at $pO_2=10^{-8}$ Pa. Both SFMO films are characterized by the tetragonal symmetry of the unit cell (I4/m). They have different concentration of antisite defects $n = [Fe_{Mo}] + [Mo_{Fe}]$ (n = 0.4for the film No. 1 with a spin polarization degree P(1) = 20 % and n = 0.23 for the film No. 2 with P(2) = 54 %). According to the temperature dependence of magnetization M = f(T) (Fig. 1), the lower M(1) are connected with larger n, which stimulate the cluster formation of $Fe^{3+}-O^{2-}-Fe^{3+}$ and $Mo^{5+}-O^{2-}-Mo^{5+}$ types. Superexchange interaction in this case is realized according to the Kramers-Anderson mechanism, and occurs due to the transition of a part of Fe cations from the high spin state $\operatorname{Fe}^{3+}(t_{2g}^{3}e_{g}^{2})$ to the medium spin state $\operatorname{Fe}^{2+}(t_{2g}^{4}e_{g}^{2})$. A small downward leap of magnetization in the temperature range of 120-130 K (Fig. 1) indicates the larger magnetic inhomogeneity of SFMO film No. 1 as compared with SFMO film No. 2. Such behaviour of magnetic characteristics can be explained by local inclusions of $SrFeO_{3-\delta}$ phase in the film No. 1.

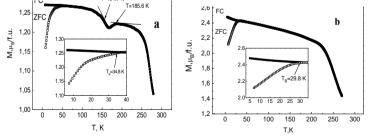
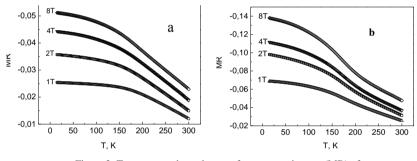
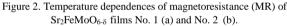


Figure 1. Temperature dependences of magnetization (M) of Sr₂FeMoO_{6- δ} films No. 1 (a) and No. 2 (b).

Relying on the fact that only negative exchange interactions are possible between $Fe^{2+}(3d^6)$ ions, the formation of $Fe^{2+}-O^{2-}-Fe^{2+}$ clusters promotes the negative exchange interaction with antiferromagentic ordering of magnetic moments. In its turn, the presence of magnetic clusters, which was observed by the authors using the small-angle neutron scattering method in the samples with 20-30 nm grains, is indirectly confirmed by magnetoresistance measurements [5].

As a result of resistivity measurements in the magnetic field ranged from 0 to 8 T, the highest magnetoresistance (MR) was determined at 15 K and B = 8 T for the sample No. 2. (Fig. 2). Tunneling magnetoresistance (TMR) effect associated with the charge carrier tunneling through grain boundaries is expected to be responsible for that.





The SFMO films under study are considered as a domain system consisting of alternating magnetic and non-magnetic domains with different orientations of magnetization. The magnitude of the TMR effect, TMR = $2P_1P_2/[1 - P_1P_2]$, where P_1 , P_2 are spin polarizations of magnetic grains, depends both on the spin polarization degree in the grains and on the relative direction of the grains magnetization due to various probabilities of tunneling of charge carriers with different spin orientations. The spin polarization P, defined as $P = \{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)\}/\{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)\}$, where $N_{\uparrow,\downarrow}(E_F)$ is the density of electronic states at the Fermi level with a spin orientation " \uparrow " or " \downarrow ", is determined by the presence of antisite defects in Sr₂FeMoO_{6-δ}. With an increase of the antisite defect concentration in SFMO films, the interaction of magnetizations in different cation sublattices takes place, leading to an antiferromagnetic state. In that case the growth of antiferromagnetic clusters Fe³⁺-O²⁻-Fe³⁺ and Mo⁵⁺-O²⁻-Mo⁵⁺ can occur resulting in a decrease of magnetization of the SFMO films. A change of

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the spin orientation in such clusters does not change the total energy of the system, which can lead to the fluctuating character of the magnetization and the sharp drop of P.

4. Conclusion

 $Sr_2FeMoO_{6-\delta}$ films, deposited with the rate of 8 nm/min and substrate temperature of 923 K in Ar medium and $Sr_2FeMoO_{6-\delta}$ films prepared with additional annealing in an evacuated quartz tube have different concentrations of antisite defects and different degrees of spin polarization. Fresh $Sr_2FeMoO_{6-\delta}$ films possess lower magnetization and are more magnetically inhomogeneous. They include phase separation with formation of local inclusions of $SrFeO_{3-\delta}$.

References

- 1. J. L. MacManus-Driscoll, A. Sharma, Adv. Mater. 54, 67 (2005).
- 2. R. Boucher, J. Phys. Chem. Solids 66, 1020 (2005).
- 3. A. Di Trolio, R. Larciprete, V. Marotta *et al.*, *Phys. Stat. Sol.* (*c*) **3**, 3229 (2006).
- 4. M. Kalanda, G. Suchaneck, A. Saad *et al.*, *Mater. Sci. Forum* **636**, 338 (2010).
- M. Watahiki, J. Suzuki, Y. Tomioka, Y. Tokura, J. Phys. Soc. Jpn. Suppl. A 67 (2001).