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Thin films of dielectric strontium iridates — materials for superconducting cryoelectronics and spintronics

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The structural and electrophysical properties of epitaxial thin films of strontium iridate Sr_2IrO_4, which were obtained by laser ablation, have been studied. Data on the effect of deposition modes on the electrophysical properties of films are presented. Electronic transport models are discussed.

Keywords: strontium iris, band conductivity, hopping conductivity.

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1. Introduction

Interest in strontium iridate Sr_2IrO_4 (SIO4) is associated with its unusual properties. Thus, for example, the possibility of the appearance of superconductivity in Sr_2IrO_4 upon doping is predicted [1]. Most of the properties are associated with the increased strength of the spin-orbit coupling in 5*d* iridates, which distinguishes them from well-studied superconducting 3*d* cuprates [2], for example, La_2CuO_4, with which SIO4 has structural similarity. In electron-doped SIO4, a *d*-wave gap in the electron density of states [1] and an anisotropic magnetoresistance were found. Thin films of SIO4 sometimes exhibit the activation dependence of resistance on temperature: the dependence of specific resistivity is characterized by an exponential function of the reciprocal temperature [see work 3]. At temperatures below 240 K, the SIO4 substance transits into the antiferromagnetic state.

The crystal structure of SIO4 single crystals is layered, of the La_2CuO_4 type, has tetragonal symmetry with the space group $I4/mmm$, in the absence of stresses, the lattice parameters are: $a = 3.888$ Å, $c = 12.90$ Å. The dielectric SIO4 phase requires a synthesis mode that is not standard for oxides, in particular, a low oxygen pressure [3–5]. The purpose of this work is to develop a technique for growing SIO4 films on different substrates, as well as to measure the electrophysical parameters of the obtained films, as well as to compare the electrophysical parameters depending on the type of substrates and the deposition mode.

2. Experimental

Thin SIO4 films were deposited on various substrates by laser ablation. The targets were made from mixture of $SrCO_3$ and IrO_2 powders using standard ceramic processing [6]. SIO4 films were deposited by KrF excimer laser. In the works [3–5], deposition was carried out in an oxygen atmosphere at pressure of 0.0013−0.05 mBar and temperatures from 700 to 850°C. The first SIO4 film was grown in a similar regime at oxygen pressure of 0.05 mbar and temperature of 770−730°C on a (100)$SrTiO_3$ (STO) substrate, which further will be called mode 1. The pulse frequency was 2 Hz, the energy was 1.6 J/cm^2, the number of pulses were 2000. It can be seen from the X-ray diffraction pattern in Fig. 1 that the diffraction peak from the film corresponds to the *c*-parameter of lattice 12.23 Å, which is less than the tabular value for the SIO4 single crystal, equal to 12.891 Å. Probably, the film contains an impurity phase with smaller lattice parameter, approaching in composition to $Sr_3Ir_2O_7$. To exclude the growth of the impurity phase, it was necessary to further reduce the oxygen pressure, which is difficult from a technical point of view. Therefore, an alternative solution was taken: the oxygen atmosphere was changed to argon atmosphere.

It can be seen from the data presented in Fig. 1 that, in an Argon atmosphere the *c*-parameter in the major phase is 12.83 Å (the film thickness is 17 nm). This value is even better than the *c*-parameter obtained in [3] and corresponds to the tabular *c*-parameter for the SIO4 single crystal. First, the pressure of 0.5 mbar Ar and temperature of 750°C were applied (second mode of deposition). Debugging of the deposition mode

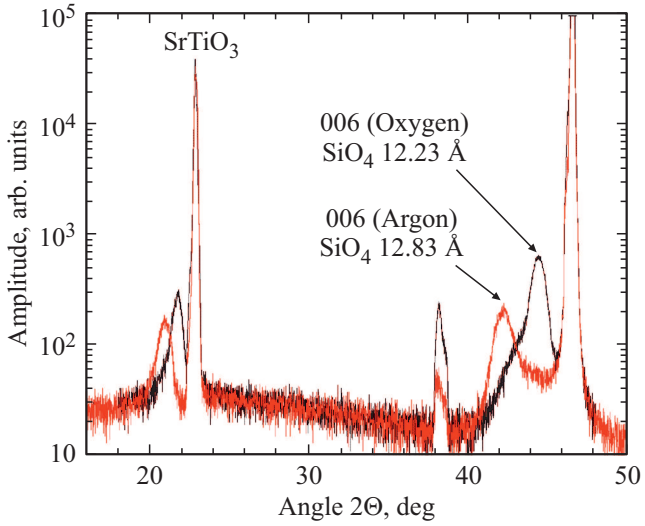

Figure 1. X-ray diffraction pattern, $\theta/2\theta$ scan of two SIO4 films on STO substrates.

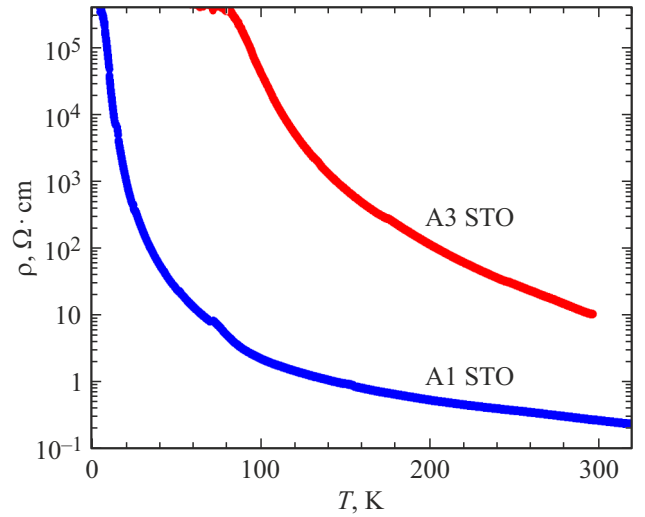

Figure 2. Specific resistivity dependencies on the temperature of SIO4 films on STO substrates: (A1) $d = 17$ nm is deposited in oxygen, (A3) $d = 34$ nm is deposited in argon.

showed that the best structure of the films grows at temperature of $T = 800-760°C$, which we will call as „mode 3“İn this mode, films were deposited on 4 types of substrates: (100)$SrTiO_3$ (STO), (110)$NdGaO_3$ (NGO), (100)$Ls_{0.3}Sr_{0.7}Al_{0.65}Ta_{0.35}O_3$ (LSAT) and (100)$LaAlO_3$ (LAO). Films on (100) $SrTiO_3$ substrates had the best structure, the c-parameter of such films was 12.87 Å.

Several films were chosen for the experiment: deposited one in mode 1 on $SrTiO_3$ A1 STO, fabricated one in mode 3 on $SrTiO_3$ A3 STO, as well as films deposited on substrates $NdGaO_3$ at oxygen pressures of 0.05 mbar (sample A NGO) and 0.3 mBar (sample B NGO). For two films on STO, the dependence of specific resistivity on temperature $\rho(T)$ is shown in Fig. 2.

The SIO4 film deposited in mode 1 has the lowest resistance. The specific resistivity of the samples is characterized by the activation energy ΔE_A, which corresponds to the band gap of the dielectric. For band dielectrics, the dependence of resistance on temperature has the form [3,7]:

$$\rho(T) = \rho_0 \exp\left(\frac{\Delta E_A}{2kT}\right), \quad (1)$$

where k is Boltzmann's constant. For dielectrics, the activation energy depends only slightly on temperature. Activation energies were calculated using the same formula as in [7]:

$$\Delta E_A = \frac{d(\ln\rho)}{d(T^{-1})}. \quad (2)$$

Temperature dependence of the activation energy is shown in Fig. 3. For SIO4 films on strontium titanate, the activation energies changed slightly in the range of 100−250 K. The ΔE_A value depends on the film deposition mode. Energies ΔE_A were 50−70 meV in oxygen deposition mode, and it were 200−260 meV in argon mode. For film A1, the $\Delta E_A = c + bT^{3/4}$ was approximated (shown by the dash-dotted line in Fig. 3). The parameter c, estimating ΔE_A as the temperature tends to zero, was 10−15 meV. This indicates the activation mechanism for the transport of charge carriers in films on strontium titanate substrates.

Films deposited on $NdGaO_3$ substrates at oxygen pressures of 0.05 mbar (sample A NGO) and 0.3 mBar (sample B NGO) had lower specific resistivities than films deposited on strontium titanate A1 STO and A3 STO. The dependences $\rho(T)$ for samples A NGO and B NGO are shown in Fig. 4.

The activation energies of these films calculated by formula (2) are shown in Fig. 5.

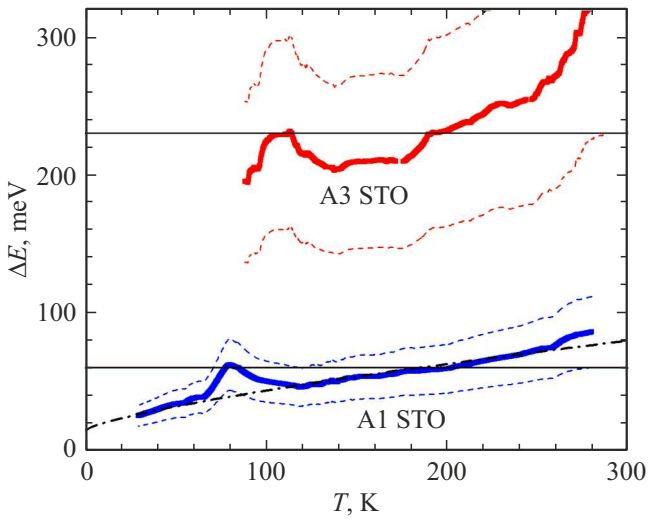

Figure 3. Temperature dependences of activation energies for SIO4 films on STO substrates in modes (1) and (3) are thick lines. Dotted lines are errors in energy calculation equal to 30 percent. Straight lines are estimates for ΔE_A.

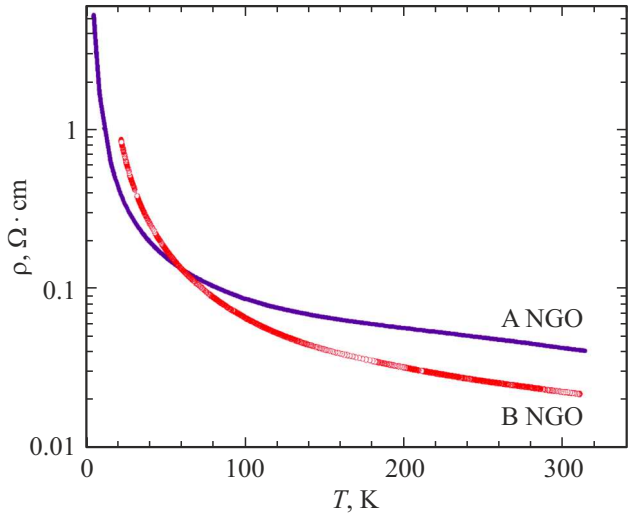

Figure 4. Specific resistivity of films deposited on NGO substrates at pressures O_2: 0.05 mBar is A NGO, 0.3 mBar is B NGO, depending on temperature.

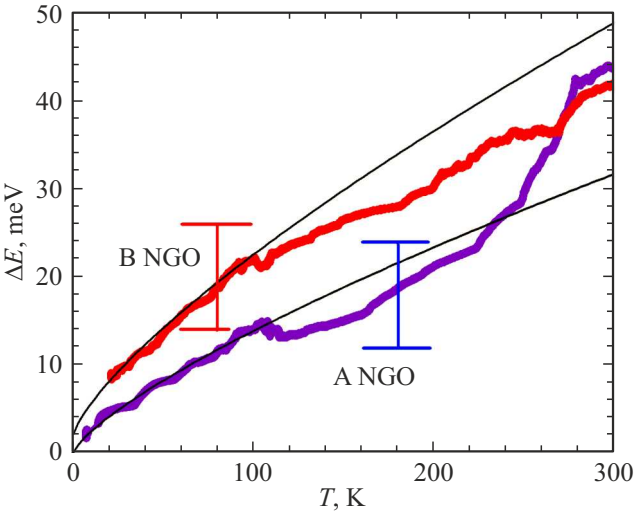

Figure 5. Activation energies of SIO4 films deposited on neodymium gallate at 0.05 mBar (A NGO) and 0.3 mBar (B NGO). Approximations for ΔE_A are implemented in the model of three-dimensional hopping conductivity according to the formula (4) by solid lines.

The errors in determining these energies are shown by conventional signs for the point error and are about 30 percent. Errors arise in numerical differentiation of experimental curves $R(T)$. For three-dimensional hopping conductivity, the temperature dependence of specific resistivity has the form

$$\rho(T) = \rho_0 \exp\left[\left(\frac{T_0}{T}\right)^{1/4}\right], \qquad (3)$$

where ρ_0 and T_0 are constants determined from experiment. In this case, the activation energy depends on temperature according to the law [8]:

$$\Delta E_A = \left(g(\mu)\cdot\alpha^3\right)^{-1/4}\cdot(kT)^{3/4}, \qquad (4)$$

where $g(\mu)$ is the density of states at the Fermi level, α is the localization radius of charge carriers. It can be seen from formula (4) that in the model of hopping conductivity, the activation energy vanishes as the temperature tends to zero. The magnitudes of activation energies for films on neodymium gallate also tend to zero with decreasing temperature (Fig. 5). This indicates the absence of a band gap in these films. The probable transport mechanism in SIO4 films on neodymium gallate is three-dimensional hopping conductivity. Differences in the properties of films deposited on different substrates are attributed to stresses that arise due to the mismatch between the parameters of the crystal lattice of the substrate and the film.

3. Results and discussion

In the work [9], laser deposition was carried out from a $Sr_3Ir_2O_7$ target. At an oxygen pressure of 0.001 mBar thin films of the composition Sr_2IrO_4 were obtained, and with an increase in pressure O_2 to 0.07 mBar, the composition $Sr_3Ir_2O_7$ corresponding to the target is obtained. Similar results were obtained in the work [10], where the growth of films $SrIrO_3$, Sr_2IrO_4 and $Sr_3Ir_2O_7$ from the same $SrIrO_3$ target, was researched. The composition of the deposited iridate films in these works varied depending on the oxygen pressure and the substrate material. In the deposition mode with oxygen pressure of 0.05 mBar, we obtained SIO4 films with low activation energy of 50−70 meV and admixture of the impurity phase (sample A1 STO in Fig. 3). To obtain a low partial pressure of oxygen, sputtering was carried out in pure Ar [6]. As a result, high-resistance films with activation energies of 200 meV were obtained. SIO4 crystals have a-parameter of lattice about 3.89 Å. The mismatch in a-parameter with STO substrates, for which $a = 3.90$ Å, for SIO4 films is minimal. Substrates with smaller a-parameters, for example, NGO ($a = 3.86$ Å), introduce compressive stresses into the films, which may lead to the formation of defects.

In the work [3] SIO4 films were deposited on STO substrates at 0.05 mBar of O_2. The hopping conductivity mode prevailed in films with thicknesses up to 150 nm, but at thicknesses of 200−300 nm, the activation conductivity mode was also observed. In the work [5] SIO4 was deposited at 0.001 mBar of oxygen. As a result, films were obtained with activation energies ΔE_A of about 100 meV at temperature of 100 K, and this energy decreased with decreasing temperature. The mechanism of film conductivity (hopping one or activation one) has not been finally elucidated [5]. The hopping conductivity mechanism is tunneling between impurities or between structural defects [8]. Comparing our data with the results of the mentioned works, we point out that our samples had

higher specific resistivities, their crystal lattice c-parameter was closer to the tabular value of SIO4 single crystals.

Samples of SIO4 films on STO substrates had activation conductivity, high resistances, and probably had fewer defects than films with hopping conductivity mechanism obtained by deposition in oxygen in previous works [3,5].

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Conflict of interest

The authors declare that they have no conflict of interest.

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