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X-RAY DIFFRACTION INVESTIGATIONS OF $NdGaO_3$ SINGLE CRYSTALS

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Neodymium gallium perovskite single crystals grown with the Czochralski method were examined with several complementary X-ray methods. By means of X-ray diffraction topography and reciprocal space diagram the structural perfection and crystal homogeneity of the studied wafers were determined. Additionally, the results of the X-ray reflectometry investigations of the surface perfection after the mechanochemical treatment are presented.

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1. Introduction

Neodymium gallium perovskite single crystal is one of the most promising substrate materials for epitaxial growth of high temperature superconductors. It has an orthorhombic structure isostructural with the $GdFeO_3$. Lattice constants in this system reported so far were $a = 5.428$ Å, $b = 5.498$ Å and $c = 7.710$ Å [1].

One of the most important factors from the viewpoint of epitaxy is the lattice matching of the substrate and film materials. The lattice mismatch between $NdGaO_3$ ($\sqrt{a+b/2}$) and the tetragonal a axis of YBaCuO was estimated to be only 0.27% around 700°C [2]. Therefore, $NdGaO_3$ is quite appropriate for practical use as a substrate.

It is well known that the structural defects in the substrate may imply the limitation of the homogeneous effective area of the monocrystalline epitaxy layer and are detrimental to high temperature superconductivity. But it depends on their nature. In our paper the structural perfection in the substrate $NdGaO_3$ was determined by means of X-ray diffraction topography and reciprocal space mapping. Additionally, the results of the X-ray reflectometry investigations of the surface perfection after mechanochemical treatment are presented.

2. Experimental procedure

2.1. Crystal growth and sample preparation

One of the most important problems during growth of $NdGaO_3$ by the Czochralski method is their strong tendency towards twinning.

The first of authors [3] reported that twinning is correlated with second-order phase transition. Many other authors regard unsuitable thermal gradients as reason for the formation of twins [1, 4].

Investigations which were made in IKZ in Berlin [6] show one more mechanism of twinning in $NdGaO_3$. Due to unfavorable thermal condition in the growth assembly and to the good thermal conductivity of $NdGaO_3$ crystal there arise instabilities at the interface after seeding. According to the convex melting isotherm most of molten materials is transported to the central region (where melt is undercooled) and it crystallizes rapidly. This region is strongly distorted and it becames a source of twins. The investigations were made for crucible diameter about 50 mm. Together with enlargement of crucible diameter, unprofitable thermal gradients increase.

In this study crystals 2″ ($\approx$ 52 mm) in diameter were grown and crucible 100 mm in diameter was used. To suppress twinning in $NdGaO_3$ crystals we introduced modification of the growth assembly and technique.

Firstly, to achieve the best condition for 2″ $NdGaO_3$ growth and to obtain flattening of melt isotherm and for reason of decreasing the supercooled region, except an upper active afterheater, a bottom afterheater was used. Additionally, in order to remove residual rapidly crystallized material in seeding phase, the dipping had to proceed in two steps until about several millimeters of the seed were touched into the melt.

Mixture of Nd_2O_3 and Ga_2O_3 (both 4N purity) in stoichiometric ratio (Nd:Ga = 1:1) was used as raw materials. The pulling rate of $1 \div 2$ mm/h and crystal rotation rate of $6 \div 12$ rpm were used. The growth chamber was filled with N_2 gas. Seed crystals [110] and [101] oriented in the orthorhombic symmetry were used in this study. The cooling rate was < 50 K/h.

In the above conditions twin-free crystals were obtained without crack, $50 \div 56$ mm in diameter and $40 \div 60$ mm in length of cylindrical part. The color of crystal was deep purple.

The samples were cut parallel to (101) plane from the one crystal and parallel to (100) planes from the second crystal with [100] growth axis. All samples were mechanochemically polished from both sides and the thickness of the samples was reduced to 200 μm.

2.2. Experimental methods

In the paper the structural perfection of $NdGaO_3$ was investigated with three different X-ray methods: the Lang method (i), reciprocal space mapping (ii) and X-ray reflectometry (iii).

The Lang method was realized in transmissions mode using Mo K_{α_1} radiation. Topographs were taken with 112 type reflections. The generator RIGAKU RU-200PL with a rotating anode and a Lang camera were used. The topographs

of the whole crystal sample are formed by traversing the crystal and film together across the beam. They are sensitive to both extinction and orientation contrast, the orientation sensitivity being about 5×10^{-4} rad.

The reciprocal space mapping was performed on the X-ray high resolution diffractometer using Cu K_{α_1} with a line focus and a Ge (400) monochromator in the primary beam with a very narrow slit (about 0.02 mm). The monochromator optics provides the spectral width $\Delta\lambda = 1 \times 10^{-3}$ Å and the divergence of the primary beam is $\Delta\Theta_{pr} = 4.6 \times 10^{-4}$ rad. The minimal size of the detector acceptance angle is 3.6 arcsec (when a Si channel cut analyzer crystal with 400 reflection is used). Finally, the size of the reciprocal space probe — the region in reciprocal space within which the scattered intensity is collected and integrated — is 1.1×10^{-5} Å$^{-1} \times 4.2 \times 10^{-4}$ Å^{-1} [5].

The X-ray reflectometry measurements were performed with SIEMENS X-ray reflectometer at the Institut für Ionenstrahlphysik und Materialforschung of the Forschungszentrum Rossendorf. This reflectometer equipped with conventional X-ray tube provides the reproducibility of the absolute value of the incident angle with an accuracy of about 0.001° and enables the measurement of reflectivity curves displaying dynamical range of almost 6 orders of magnitude. Collimation of the incident beam is achieved by an adjustable knife edge over the sample surface. The evaluation of results is performed by means of a simulation procedure (Refsim program).

3. Results

It was stated that both investigated crystals were generally twin-free. The topographs representative to the greatest part of volume of the investigated crystals are shown in Fig. 1. The defects revealed in the topographs were the growth striations and dislocations. The distribution of dislocations was not homogeneous. It changed from more than 10^2 cm^{-2} to 10^3 cm^{-2}. In some cases one could see that the arrays of dislocations began to be close to the natural crystal wall. The space between some of them was nearly dislocation-free. These arrays of dislocations cannot be easily interpreted as glide bands. The reciprocal space mapping and X-ray reflectometry measurements were taken from the center of the sample, where such arrays are not present.

The reciprocal space mapping shown in Fig. 2 enabled the evaluation of FWHM of the 400 reciprocal lattice points (RELP) in both [001] and [010] directions. The experimental values are equal to 9.5 and 13 arcsec, respectively. The corresponding theoretical FWHM of (400) reflections [7] in $NdGaO_3$ is 9.5 arcsec. The collected data demonstrate that the crystal perfection in the [100] direction is close to the theoretical value.

The X-ray reflectivity measurements in $NdGaO_3$ were performed in the sample presented in Fig. 1a. The results from two perpendicular positions of the same sample are shown in Fig. 3. The surface roughness was evaluated to be less than 2 Å. The surface density of this bulk sample can be determined from the critical angle of total reflection. For the fitting process the theoretical density $r = 7.564$ g/cm^3 was taken, which agrees perfectly with the measured critical angle taking absorption into account. This means that the surface and the bulk

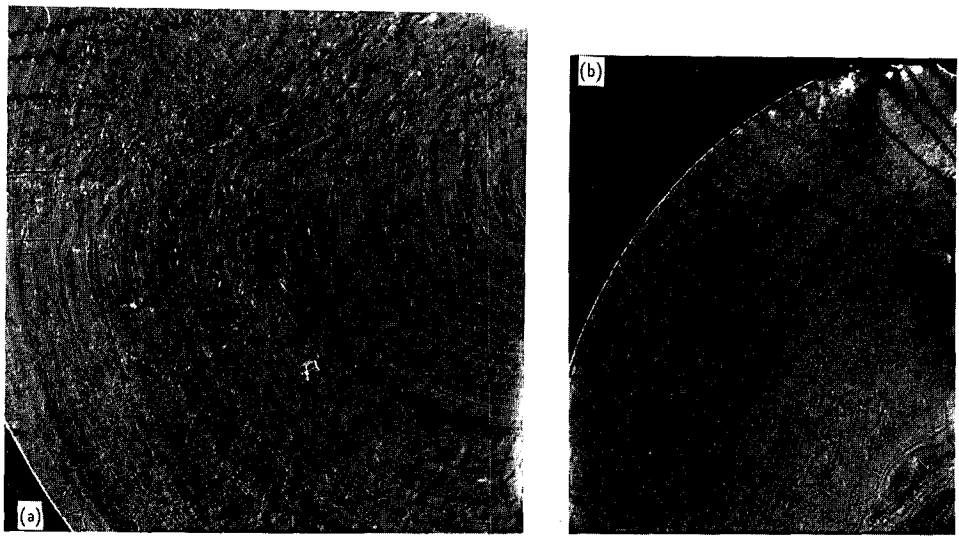

Fig. 1. (a) 112 Lang transmission topography of the $NdGaO_3$ sample cut along a (101) plane. (b) 112 Lang transmission topography of the $NdGaO_3$ sample cut along a (100) plane.

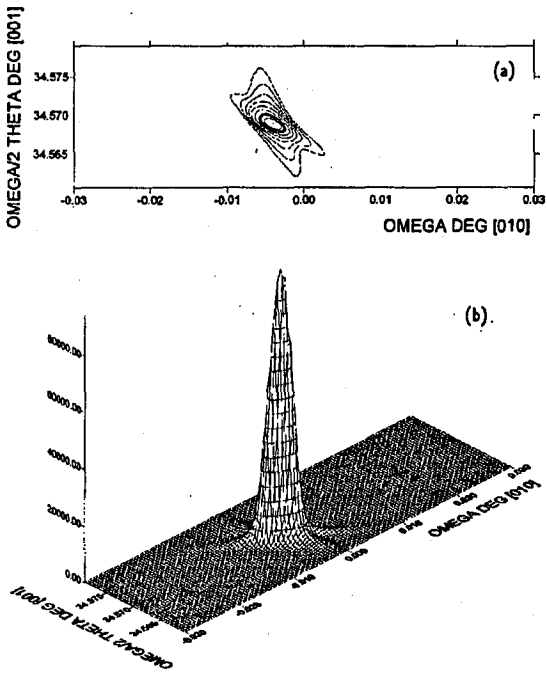

Fig. 2. Reciprocal space map around (400) RELP of the $NdGaO_3$ measured in the sample shown in Fig. 1b (Cu K_{α_1} radiation): (a) contour map, (b) three-dimensional map.

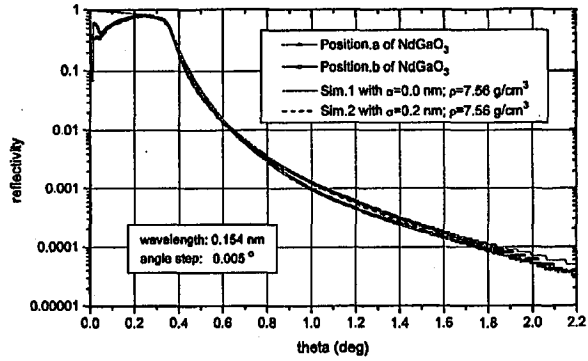

Fig. 3. Reflectivity curves with fitted simulations measured in $NdGaO_3$ wafer shown in Fig. 1a. The two curves (a, b) correspond to the two azimuths of the sample differing by 90°.

density of the wafer are the same. The surface roughness of 2 Å is comparable to Si epi-ready wafers and gives us information about the super polishing treatment of presently examined $NdGaO_3$ wafers.

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