Technical Physics Letters, Vol. 28, No. 3, 2002, pp. 221–223. Translated from Pis'ma v Zhurnal Tekhnicheskoĭ Fiziki, Vol. 28, No. 6, 2002, pp. 12–17.
Original Russian Text Copyright © 2002 by Makeev, Motornenko, Cherpak, Babiĭchuk, Kosmyna.

On the Anisotropy of Dielectric Permittivity in Single Crystal Lanthanum Aluminate Substrates

Yu. G. Makeev, A. P. Motornenko, N. T. Cherpak,
I. P. Babiĭchuk, and M. B. Kosmyna
Usikov Institute of Radiophysics and Electronics, National Academy of Sciences of Ukraine, Kharkov, Ukraine
e-mail: briz@ire.kharkov.ua
Institute of Single Crystals, National Academy of Sciences of Ukraine, Kharkov, Ukraine

Received April 13, 2001; in final form, October 22, 2001

Abstract—The dielectric properties of $LaAlO_3$ single crystal plates were studied by microwave techniques in a frequency range from 20 to 28 GHz. The measurements were performed using a resonator representing crossed round (cylindrical) and radial evanescent waveguides with the polarization-degenerate fundamental oscillation mode. Various single crystal samples, including those obtained from different manufacturers, exhibit anisotropy in the (100) crystal plane, whereby the permittivity varies within $\Delta\varepsilon = 0.1–0.9$. © *2002 MAIK "Nauka/Interperiodica".*

The epitaxial growth of high-temperature superconductor film structures determines the need in single crystal dielectric substrates with the surface possessing fourfold crystallographic symmetry [1]. The requirements on such substrates are satisfied, in particular, by perovskites with the general formula ABO_3, among which special attention was devoted to lanthanum aluminate ($LaAlO_3$). The results of investigations of the dielectric properties of this compound were reported in [2–6]. The published data on the lattice parameters of $LaAlO_3$ single crystals show evidence of a certain deviation of the crystal structure from cubic. By these data, the structure of $LaAlO_3$ can be considered as pseudocubic, composed of unit cells with a side edge length a and an angle α [2, 3]. The perovskite crystal may feature the so-called twinning. According to [2], this can lead an uncertainty in the dielectric permittivity of $LaAlO_3$ crystals and, hence, to difficulties in developing microwave devices in which the resonance frequency has to be specified with a high precision.

It was pointed out [1] that "$LaAlO_3$ substrates possess rather high dielectric permittivity ($\varepsilon = 24$ at $T = 77$ K) slightly varying with orientation due to the twinning effect." Therefore, questions arise as to what is the possible anisotropy in the microwave properties of $LaAlO_3$ and the possible factors determining this effect. These factors may include a deviation of the crystal structure from cubic and the influence of various defects, primarily those caused by the crystal twinning. In connection with this, we have studied the dielectric properties of single crystal $LaAlO_3$ substrates from the standpoint of the possible influence of the anisotropy on the frequency splitting of the polarization-degenerate fundamental oscillations in the microwave range.

A method used for the measurement of the dielectric permittivity of anisotropic single crystal samples is based on using a polarization-degenerate fundamental mode $HE^{c,s}_{11\delta}$ in a cylindrical waveguide resonator [7, 8]. When an anisotropic dielectric of a finite length is placed into a waveguide, so that the sample occupies the entire waveguide cross section, the degeneracy of oscillations is removed and two self-oscillation modes ($HE^{c}_{11\delta}$ and $HE^{s}_{11\delta}$) appear instead of the single degenerate mode. The polarization planes of these modes contain the directions of maximum and minimum permittivity for the crystals of orthorhombic, hexagonal, and cubic systems. The permittivity of a crystal sample can be calculated using the measured fundamental frequencies of a resonator formed by an evanescent cylindrical waveguide of a finite length and a disk made of the material studied. The directions of axes are determined by the polarization of oscillations relative to the crystal.

The computational formulas were presented and the measuring unit described elsewhere [7]. However, that system was designed to measure the parameters of disk-shaped substrates and could not be used to study the samples of different shapes.

In this study, we employed a resonator representing crossed perpendicular round (cylindrical) and radial evanescent waveguides [8]. An advantage of the measuring unit of this type as compared to the aforementioned design is the possibility of studying plane-parallel single crystal dielectric samples of a given thickness and arbitrary shape. The only requirement on the samples studied was that the spot of the electromagnetic field in the radial waveguide would be smaller than the sample area. The permittivity of a crystal sample stud-

1063-7850/02/2803-0221$22.00 © 2002 MAIK "Nauka/Interperiodica"

Table 1. Data on the geometry and permittivity of $LaAlO_3$ substrates cut from single crystals grown in the Institute of Single Crystals

Sample no.	1	2	3	4	5
Sample shape	Square	Circle	Circle	Circle	Rectangle
Dimensions, mm	10 × 10 × 0.32	∅7	∅7	∅7	22 × 20 × 0.50
ε_1	24.2	23.7	23.9	23.8	23.7
ε_2	23.3	23.1	23.4	23.6	23.6
$\Delta\varepsilon$	0.9	0.6	0.5	0.2	0.1

ied can be calculated by relationships presented in [9], using the measured resonance frequencies and the known sample thickness and the resonator geometry. In order to establish the orientation of crystal axes in the sample, it is necessary to determine the directions of the projections of an excitation element onto the substrate plane at the moment of suppression of one of the self-oscillations. These directions coincide with the directions of projections of the axes of the refractivity ellipsoid [10].

Preliminarily, this method was used in the control measurements performed on dielectric samples with known values of the dielectric permittivity. For this purpose, we employed the samples of fused and single crystal quartz. The single crystal quartz samples were cut in the planes for which the $\varepsilon_\perp$ and $\varepsilon_\parallel$ values could be determined. The samples had the shape of either disk or parallelepiped with lateral dimensions 5–10 mm and a thickness from 0.25 to 1.0 mm. The ε values were 3.80 ± 0.04 for fused quartz samples and $\varepsilon_\perp = 4.40 \pm 0.04$ and $\varepsilon_\parallel = 4.60 \pm 0.04$ for single crystals. In the course of measurements, the samples were moved relative to the center of the radial waveguide and the resonance frequencies f_r were measured in each position. It was found that the f_r values were constant for $z \geq 0.60$ mm, where z is a minimum distance from the sample edge to the inner cylindrical waveguide edge. This implied that, in this range (for $z \geq 0.60$ mm), neither size nor shape of the samples affected the results of measurements.

The dielectric properties of $LaAlO_3$ samples were measured in the millimeter wavelength range. For this purpose, the internal diameter of the cylindrical waveguide of the measuring resonator was taken equal to 3.01 mm. The experiments were performed with $LaAlO_3$ samples obtained from two manufacturers: (i) the Institute of Single Crystals (National Academy of Sciences of Ukraine, Kharkov) and (ii) the Coating & Crystal Technology company (USA). In the former case, the crystals were grown by the Czochralski technique on a "Kristall" setup with iridium crucibles and an ingot cross-section control system. The ingots, grown from a charge prepared by mixing the initial La_2O_3 and Al_2O_3 oxides, were 30 mm in diameter; the crystallization front changed from convex toward the melt to virtually flat.

The structure of $LaAlO_3$ single crystals was studied by X-ray diffraction. The results of optical measurements showed that the single crystal was homogeneous with respect to elastic stresses. The samples for the dielectric measurements were prepared as follows. First, the (100) single crystal plane was determined and the single crystal ingot was cut into plates oriented in this direction with the aid of a Microslice cutting device. Then the plates were ground with a diamond powder (10–14 μm mesh size) and polished from both sides. Finally, the plates were cut to square, rectangular, or disk shape with a thickness of 0.30–0.60 mm. The deviation from plane-parallel configuration (thickness variation) did not exceed 0.01 mm. The same precision was observed with respect to the inner dimensions of the measuring resonators.

The samples of single crystals from Coating & Crystal Technology company with a thickness of 0.52 mm were also oriented in the (100) direction. These samples were ground and polished on one side.

The values of dielectric permittivity measured in the central region of the $LaAlO_3$ crystals grown at the Institute of Single Crystals are presented in Table 1. The data in Table 1 are arranged in the order of decreasing $\Delta\varepsilon$. The values of ε_1 and ε_2 were obtained by averaging over a series of not less than ten measurement cycles. In each series of measurements, the central part of the sample was placed in the region of maximum electromagnetic field strength of the resonator. The scatter of experimental values of the resonance frequency did not exceed ±50 MHz, which corresponds to an uncertainty in $\Delta\varepsilon$ not exceeding ±0.01. The measurements were performed in a frequency range from 20 to 28 GHz.

Table 2 shows the ε_1 and ε_2 values obtained in a similar manner for the $LaAlO_3$ single crystal plates from Coating & Crystal Technology company. As can be seen from a comparison of data for sample 5 in Table 1 and sample 2 in Table 2, both crystals possess $\Delta\varepsilon = 0.10$ despite different preparation technologies and sample geometries.

In order to check for the possibility that the results obtained for sample 5 in Table 1 are due to this plate having maximum dimensions among all samples in this

Table 2. Data on the geometry and permittivity of $LaAlO_3$ substrates cut from single crystals obtained from Coating & Crystal Technology company

Sample no.	1	2
Sample shape	Rectangle	Rectangle
Dimensions, mm	9.4 × 6.2 × 0.52	12 × 6 × 0.52
ε_1	23.7	23.5
ε_2	23.4	23.4
$\Delta\varepsilon$	0.3	0.1

series, we shifted the plate relative to the center of the radial waveguide and measured the f_r value (the experiments were similar to those with quartz samples). These measurements gave $\Delta\varepsilon = 0.1$ for $z \geq 0.30$ mm. A decrease in the minimum z value for the $LaAlO_3$ sample as compared to that for quartz is quite reasonable, since the field spot in the radial waveguide with $LaAlO_3$ sample was smaller as compared to that for quartz because of large values of the permittivity.

The above data indicate that lanthanum aluminate single crystals are anisotropic in the (100) crystal plane, but the degree of anisotropy is small. An analysis of the results obtained for a series of samples (see tables) shows that there are differences both in the degree of anisotropy and in the values of permittivity, which depend both on the crystal growth technology and on the particular sample (for a series grown by the same technology). This scatter is probably related to the crystallographic anisotropy and to the presence of defects (in particular, twins) in the $LaAlO_3$ crystal structure. Determining the relative contributions of these factors would require further complex investigations.

The results of our experiments show that the development and fabrication of microwave devices based on high-temperature superconductor films deposited onto $LaAlO_3$ plates require microwave monitoring of the dielectric properties of these substrates prior to the subsequent film growth.

REFERENCES

1. Z. Y. Shen, *High-Temperature Superconducting Microwave Circuits* (Artech House, Boston, 1994).
2. K. H. Young, G. V. Negrete, M. M. Eddy, *et al.*, in *Proceedings of the ICMCTE-91, San Diego, 1991.*
3. R. Ramesh, A. Snam, W. A. Bonner, *et al.*, Appl. Phys. Lett. **55** (11), 1138 (1989).
4. T. Konaka, M. Sato, H. Osano, and S. Kubo, J. Supercond. **4** (4), 283 (1991).
5. J. Konopka and I. Wolff, IEEE Trans. Microwave Theory Tech. **40** (12), 2416 (1992).
6. C. Zuccaro, M. Winter, N. Klein, and K. Urban, J. Appl. Phys. **82** (11), 5695 (1997).
7. Yu. G. Makeev, V. A. Korobkin, N. I. Pyatak, *et al.*, Prib. Tekh. Éksp., No. 6, 104 (1978).
8. V. A. Korobkin and Yu. G. Makeev, USSR Inventor's Certificate No. 1117538, Byull. Izobret, No. 37 (1984).
9. Yu. G. Makeev and A. P. Motornenko, Zh. Tekh. Fiz. **64** (9), 117 (1994) [Tech. Phys. **39**, 924 (1994)].
10. V. A. Korobkin, Yu. G. Makeev, N. P. Katrich, *et al.*, Prib. Tekh. Éksp., No. 4, 156 (1981).

Translated by P. Pozdeev