PHYSICAL PROPERTIES OF LANTHANUM GALLIUM TANTALATE CRYSTALS FOR HIGH-TEMPERATURE APPLICATION

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Abstract - A major trend in the sensor industry is developing a solution to control physical processes at high temperatures (500…1000^{0}C). Currently used piezoelectric materials such as piezoelectric ceramics and quartz have a number of physical properties limiting their use in such environment. Thus sensor designers and developers are in search of new piezoelectric materials that can function properly under harsh conditions. Many world companies have been developing high-temperature piezoceramics. However they face a number of hurdles: operating temperature is limited by 500^{0}C, pyroelectric effect and temperature hysteresis of piezoelectric constant.
This work represents Lanthanum Gallium Tantalate (LGT), a piezoelectric crystal that could be a promising solution for a high-temperature application.

I. INTRODUCTION

Both Langatate (LGT) and Langasite (LGS) crystals belong to gallo-germanate family. Unlike LGS that is used for SAW and BAW applications LGT has properties that make it an attractive material for devices on the direct piezoelectric effect.

LGT is more sensitive to mechanical stresses than LGS. A critical parameter for a sensor on the direct piezoelectric effect is the dielectric properties of a sensing element, in particular electrical resistance and its change in a temperature range.

The basic characteristics of LGT, quartz and piezoceramics are listed in Table 1. As you can see LGT has no phase transitions, piezoelectric constant (d11) is three times higher than that of quartz, dielectric constant is much lower than the same value of piezoceramics.

LGT crystals display no pyroelectric, ferroelectric properties and hysteresis

II. ELECTRICAL RESISTANCE LGT CRYSTAL

Electrical resistance is a key characteristic of a material converting mechanical deformations into electric charge. Usually electrical resistivity of piezoelectric materials is rather high (about $10^{13}…10^{15}$ Ohm/cm) at room temperature but it decreases with temperature rising.

The mechanism of electrical conductivity in dielectrics is very complicated. As a rule it increases with temperature and relates to a structural perfection of a crystal. Any imperfections result in free carriers contributing to the crystal conductivity that increases at high temperatures.

A sensor element is a round or rectangular blank with electrodes deposited on working surfaces. The most common coating is gold with chromium or titanium adhesion layer. Due to high temperatures (550°C) and the passage of electric current through the blank the plated material diffuses into the crystal increasing its conductivity. This results in a noise increase vs the desired signal reducing a sensor performance.

The authors have carried out work to increase electrical resistance and preserve electrical characteristics of LGT crystals over time by improving crystal growth technique and after-growth processing.

Experimental results are summarized in the graphs below.

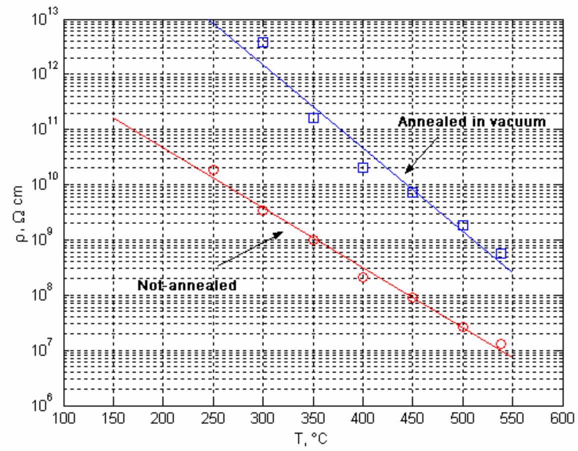

Fig.1 LGT crystals grown in oxygen atmosphere

Figure 1 shows the temperature dependence of electrical resistivity of LGT crystal grown in oxygen atmosphere and annealed in vacuum. A blue curve demonstrates a significant growth of electrical resistivity after vacuum annealing

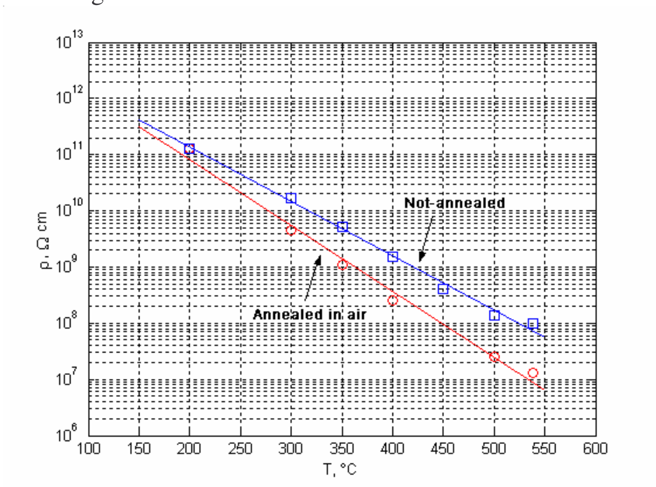

Fig.2 LGT crystals grown in inert-gas atmosphere.

Figure 2 shows the temperature dependence of electrical resistivity of LGT crystals grown in the ambient inert-gas atmosphere and annealed in air. A blue curve demonstrates electrical resistivity reduction after annealing in air.
Both sets of curves illustrate the influence of oxygen on electric characteristics of LGT crystal

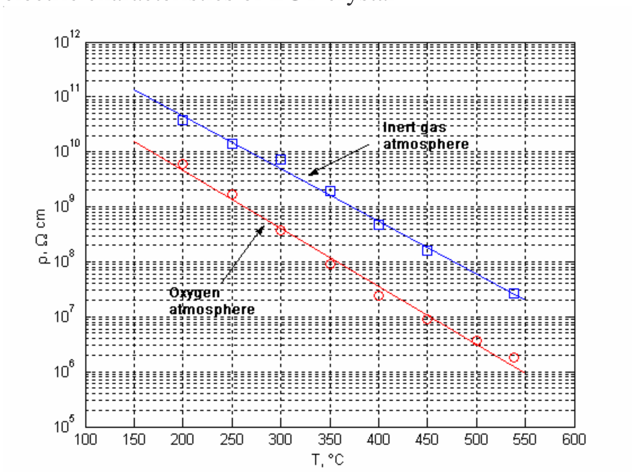

Fig.3 LGT crystals grown in oxygen and inert-gas atmosphere. Electrical resistivity vs. temperature.

Figure 3 illustrates how the electrical resistivity of LGT crystals grown in different conditions varies with temperature.

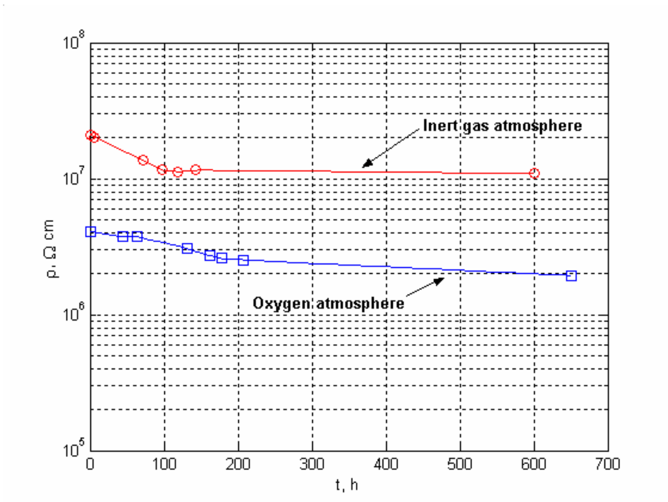

Fig.4 LGT crystals grown in oxygen and inert-gas atmosphere. Electrical resistivity vs. Temperature.

Figure 4 illustrates how the electrical resistivity of the crystals grown in different conditions varies in time. The crystals were exposed to a temperature of 5500C within 650 hours.

III. PIEZOELECTRIC CONCTANT (d11).

Piezoelectric constant (*d11)* is a critical parameter of a material employing in sensors working on the direct piezoelectric effect.
d11 of the test samples was measured at room temperature by a quasistatic method. However the use of the method is limited by high temperatures. So the temperature dependence of the piezoelectric constant (*d11)* was determined by a "resonance –antiresonance" method. It should be noted that the measurement results were greatly affected by the temperature drift of spurious resonances. The problem was caused by the difference of temperature-frequency coefficients of the fundamental and spurious modes of the test samples.
The temperature dependence of the piezoelectric constant (*d11)* is represented in Figure 5. The obtained results show that in the range of 100 … 550 °C the piezoelectric constant (*d11)* is not highly dependent on temperature up to 450^0C.

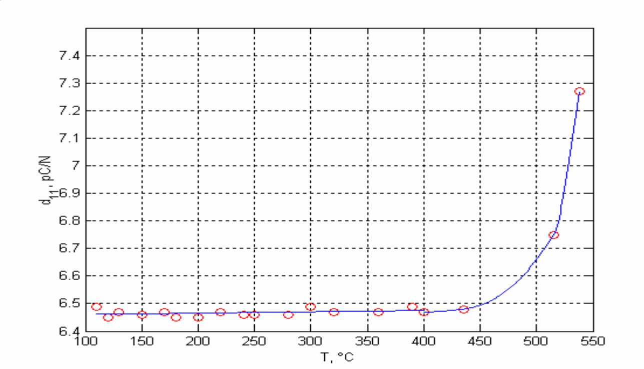

Fig.5 Temperature dependence of piezoelectric constant (d11)

IV.INVESTIGATION OF STRUCTURAL PERFECCTION OF LGT CRYSTAL X-RAY TOPOGRAPHY METHOD.

The conductivity of LGT crystal is determined by crystal structure perfection. LGT crystals are grown by the Czochralski technique. During the crystal growth, several types of growth defects are formed, which deteriorate the crystal properties. The formation of defects in LGT crystal is determined by both the charge makeup and growth-atmosphere composition and the temperature composition during the growth. The following types of growth defects were revealed in LGT crystal crystals by X-ray topography: decorated F-centers, growth banding, twins, and amorphous glassy inclusions. However only decorated F-centers strongly change the crystal properties. Formation of decorated F-centers in LGT crystals is due with presence of surplus of oxygen atoms. Presence of superfluous oxygen results in change of parameters of a crystal lattice in the given area and, as consequence, to change of physical properties. Investigation of structural perfection of LGT crystals was carried out at Kurchatov synchrotron source of X-ray radiation with use of a white bunch. Fig. 6 presents X-ray topograph of the (022)-cut of a LGT crystal received for reflection (022) at X-ray energy of E=17 кэВ. On the topograph the area of the decorated F-centers is allocated, and it is possible to observe the boundaries of the given area. Note, that large number of dislocation is formed on the boundary between two regions which is due with different crystal lattice parameters in two neighboring regions.

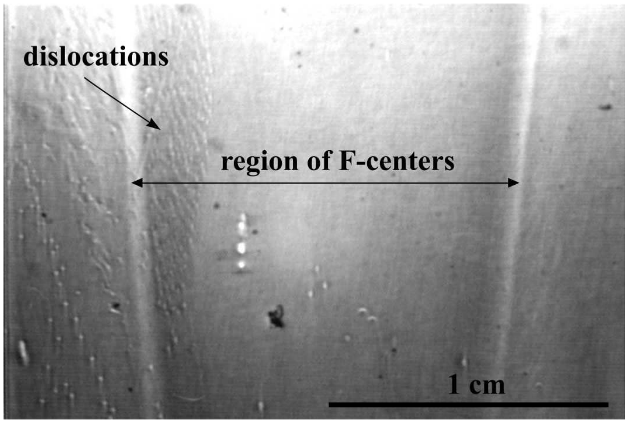

Fig. 6. White beam synchrotron topograph of the (022)-cut of a LGT crystal obtained at X-ray energy of E=17 keV. Reflection (022); $\Theta_{Br} = 10°$.

VI. Discussion

The studies show that the temperature and time dependences of electrical resistivity are related to the crystal growth environment and after-growth processing. The electrical resistivity value is greatly affected by a crystal growth atmosphere.
Electrical resistivity decreases in oxygen. The same effect is observed for the crystals grown in the inert-gas ambient environment and annealed in air
Prolonged exposure of the crystals to 5500C has the same result. Electrical resistivity of crystals grown in oxygen atmosphere eventually falls down reducing their characteristics.

VII. Conclusion

The experiments show that LGT crystals grown in the inert-gas environment feature the best electrical resistivity. They are also more stable over time in high temperatures. Additional vacuum annealing essentially raises electrical conductivity but at high temperatures crystal blanks should be isolated from the atmosphere. It can be achieved by vacuuming the sensor or filling it with inert gas.

IX. References

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Properties	Unit	NAVY Symbol	**LGT**	Quartz	APC-850 N-2	APC-856
Relative Dielectric Constant	1	K = εT33/ε0	**80.3**	4.6	1750	4100
Curie temperature	C0	Tc	**-**	570	360	150
Piezoelectric Charge Constant	10-12 C/N or m/V	-d11	**6.5**	2.3	d33 = 400	d33 = 620
		d14	**4.7**	0.9	d15 = 590	d15 = 710
Piezoelectric Voltage Constant	10-3 Vm/N or m2/C	-g11	**38**	58	g33 = 26	g33 = 18.5
		g14	**27.7**	18	g15 = 36	g15 = 25
Elastic Modulus (Young's Modulus)	1010 N/m2	YE11	**11**	7.8	6.3	5.8
		YE33	**19**	10.4	5.4	4.5
Elastic Compliance	10-12 m2/N	SE11	**9.0**	12.8	15.3	15.0
		SE33	**5.2**	9.6	17.3	17.0
Density	g/cm3	ρ	**6.13**	2.65	7.7	7.5
Electrical Resistivity	Ohm*cm T=3500C	ρ	**1010**	-	-	-