Temperature Dependent Material Parameters of $Sr_3NbGa_3Si_2O_{14}$ (SNGS) Single Crystal

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Abstract— **We have successfully grown high quality piezoelectric single crystal of $Sr_3NbGa_3Si_2O_{14}$ (SNGS) by the Czochralski technique. Transparent and pale yellow boules of approximately 60 mm in length and 16-18 mm in diameter were obtained. A full set of material parameters including dielectric, elastic and piezoelectric constants of SNGS crystal were measured in a temperature range from 220 K to 310 K. Components of the thermal expansion tensor were also determined.**

I. INTRODUCTION

Oxide crystals of $Ca_3Ga_2Ge_4O_{14}$ (CGG) structure with the general formula $A_3BC_3D_2O_{14}$ are of current interest as very promising materials for both bulk (BAW) and surface (SAW) acoustic wave devices [1-10]. The crystals belong to the same trigonal crystal class 32 as SiO_2 but compared with quartz they have two-three times higher piezoelectric coefficients (and hence, higher electromechanical coupling coefficient) and principal possibility to operate at very high temperatures due to the absence of structural phase transition from room temperature up to the melting point. The later opens a promising way for high-temperature sensors (utilizing both BAW and SAW) of various physical values like temperature, pressure, gases concentration, etc.[11-13] Obviously, the design and manufacture of new BAW and SAW devices require a full set of material parameters in the temperature range of device operation. A set of the room temperature material parameters for SNGS has been published in [9]. In this paper, we report on the measurements of the temperature dependent material parameters of the $Sr_3NbGa_3Si_2O_{14}$ (SNGS) single crystals.

II. CRYSTAL GROWTH AND SAMPLES PREPARATION

SNGS single crystals were grown by the Czochralski technique. The SNGS single crystal growing had to start with a metal seed, because Y-oriented LNG seeds yielded polycristalline zones. As a reason for this the differences of the lattice constants could be considered. Even with native Y-oriented SNGS seeds the single crystal growing is rather complicated. With a 40 mm diameter iridium crucible it was possible to pull transparent and pale yellow boules free of cracks and macroscopic defects with a final length up to approximately 60 mm and cross-section of 18 mm x 16 mm.

For the ultrasonic wave velocity measurements cubes of approximately 8 x 8 x 8 mm^3 size were cut from the as grown boule in two different orientations: (i) with the edges parallel to the main X, Y, Z crystallographic axes, and (ii) rotated by -45^0 around the X axis. The (i) sample was used also for the thermal expansion coefficients measurements. For the dielectric constants studies thin plates of X- and Z-cuts with the averaged dimensions of 8 x 5 x 1 mm^3 were used. All the samples were carefully ground followed by fine lapping or polishing to achieve parallelism and flatness of the opposite faces.

III. SOUND VELOCITIES

Measurements of the bulk acoustic wave velocities propagating along certain crystallographic directions were carried out by a RITEC Advanced Ultrasonic Measurement System RAM-5000. The system realizes pulse-echo method of time propagation measurements using phase detectors with an accuracy of about 10^{-4}. To generate longitudinal and shear ultrasonic waves, Y+36^0- and X-cut $LiNbO_3$ transducers of 10 and/or 30 MHz central frequency were used. A special attention was paid to the couplant materials to bond transducer to the sample, especially in the case of shear modes. All temperature measurements were done using a continuous flow Oxford cryostat in the 220 K to 310 K range with an accuracy and stability of 0.1 K. Temperature dependences of the elastic wave velocities are shown in Figs. 1-3. Unfortunately elastic waves with positive temperature coefficient were not found in the measured temperature

This work was supported by the DFG under the grants WE 2065/8-1 and GO 677/8-1

range. These data were used for the calculations of the elastic constants (see below).

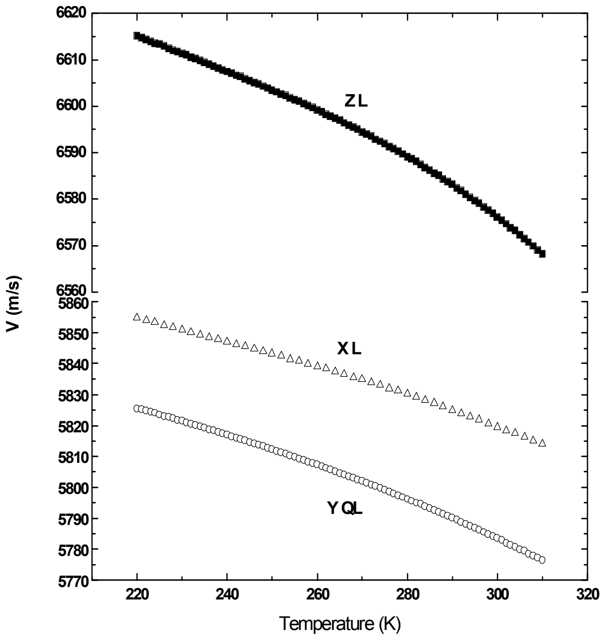

Figure 1. Velocities of longitudinal and quasi- longitudinal acoustic waves versus temperature along X-, Y- and Z-directions

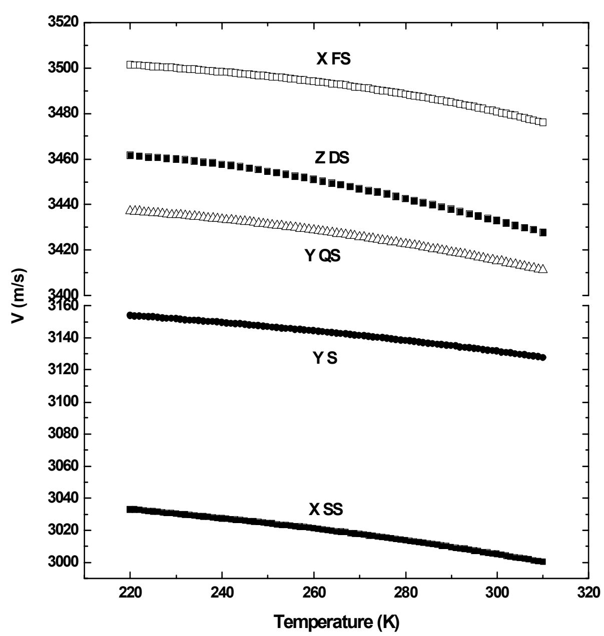

Figure 2. Velocities of shear (fast and slow) and quasi-shear acoustic waves versus temperature along X-, Y- and Z-directions

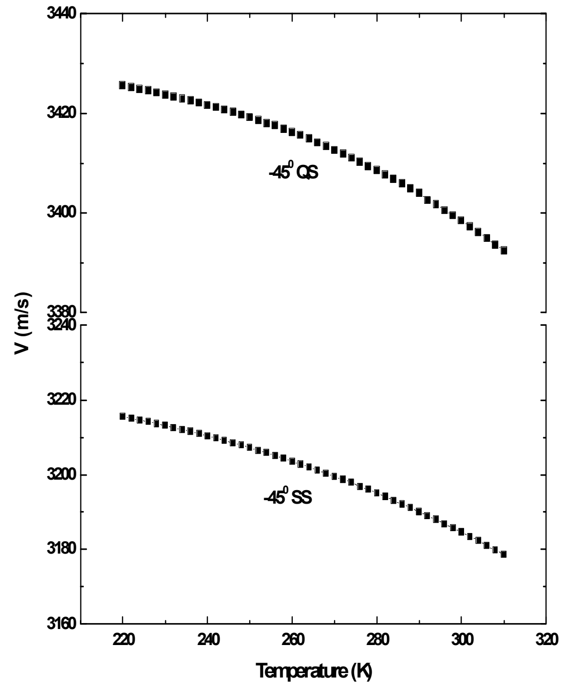

Figure 3. Velocities of shear and quasi-shear acoustic waves versus temperature along -45^0 rotated around X-axis direction

IV. DIELECTRIC CONSTANTS

The relative dielectric constants ε_{11} and ε_{33} were obtained by measuring the capacitance of X- and Z-cut plates with gold electrodes on the big opposite faces at a frequency of 1 kHz which is far below any frequency corresponding to the electromechanical (piezoelectric) resonance. The low frequency (constant stress) $\varepsilon_{ij}^{\sigma}$ and clamped (constant strain) ε_{ij}^{η} for the crystals of 32 symmetry class obey the following relations

$$\varepsilon_{11}^{\eta} = \varepsilon_{11}^{\sigma} - \frac{e_{11}^{2}}{\varepsilon_0 \cdot C_{11}}, \qquad \varepsilon_{33}^{\eta} = \varepsilon_{33}^{\sigma},$$

where e_{11} is the piezoelectric stress constant, C_{11} the elastic constant and $\varepsilon_0 = 8.85 \cdot 10^{-12}$ F/m. Temperature dependences of the dielectric constants are shown in Figs. 4,5 As is seen from Figs. 4,5, both constants decrease with temperature decreasing in contrast to LGS and LGN crystals where ε_{33} increases with temperature decreasing. Obviously, such a behavior is typical of the ordinary linear dielectrics.

V. THERMAL EXPANSION

The thermal expansion coefficients were measured using an inductive dilatometer in the 200 K to 380 K temperature range. The accuracy for the thermal coefficients of expansion determination was about 5 %. The measurements were carried out on the same cube sample with X-, Y- and Z-cut faces as

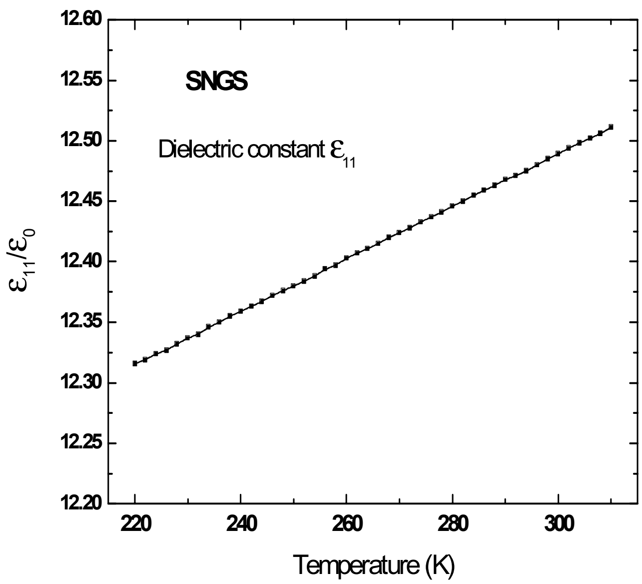

Figure 4. Temperature dependence of the dielectric constant ε_{11}

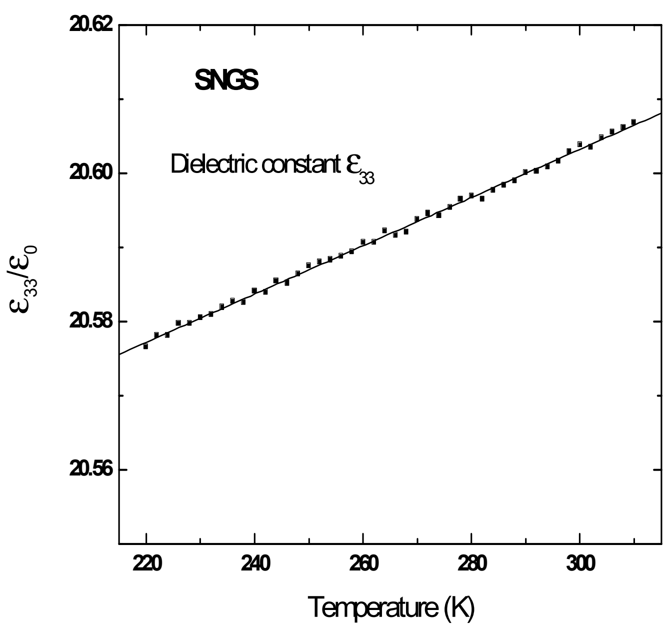

Figure 5. Temperature dependence of the dielectric constant ε_{33}

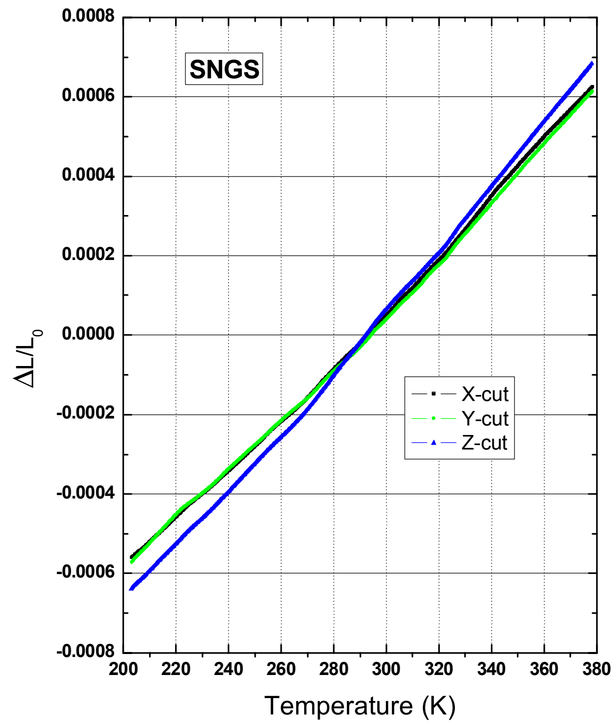

Figure 6. Relative length change versus temperature for the X-, Y- and Z-cut faces

was used for the sound velocities studies. Because of the symmetry for the 32 class crystals, the X- and Y directions have the same thermal expansion coefficient. However, all three directions have been measured. The results obtained are depicted in Fig. 6. After averaging the thermal expansion coefficients were determined to be $\alpha_{11} = 6.5\cdot10^{-6}$ K^{-1} and $\alpha_{33} = 7.5\cdot10^{-6}$ K^{-1}. Note that α_{33} is comparable with published results and α_{11} is about two times larger as compared with α_{11} from [14].

VI. MATERIAL PARAMETERS

From the above experimental data, the elastic stiffness coefficients C_{ij} and piezoelectric stress constants e_{ij} can be derived using a system of relations between sound velocities measured at different directions for different modes for 32 symmetry class crystals [10]. Here the following relations were used (Table I).

Table 2 shows a full set of material parameters at room temperature (T = 298 K). The results obtained are generally in a good agreement with published data [9] except elastic constant C_{11} which is somewhat less as compared to the published value. The results obtained give the opportunity to calculate first- and second order temperature coefficients of material parameters which are also included in Table II.

TABLE I. RELATIONS BETWEEN ACOUSTIC BULK VELOCITIES AND MATERIAL CONSTANTS FOR THE CRYSTALS OF 32 SYMMETRY CLASS

Orientation	Mode (polarization)	Effective stiffness
X-cut	Longitudinal (L)	$C_{11}+\frac{e_{11}^2}{\varepsilon_{11}}$
X-cut	Fast shear (FS)	$\frac{C_{66}+C_{44}}{2}+\frac{\sqrt{(C_{44}-C_{66})^2+4C_{14}^{\;2}}}{2}$
X-cut	Slow shear (SS)	$\frac{C_{66}+C_{44}}{2}-\frac{\sqrt{(C_{44}-C_{66})^2+4C_{14}^{\;2}}}{2}$
Y-cut	Quasi-longitudinal (QL)	$\frac{C_{44}+C_{11}}{2}+\frac{\sqrt{(C_{11}-C_{44})^2+4C_{14}^{\;2}}}{2}$
Y-cut	Fast quasi-shear (QS)	$\frac{C_{44}+C_{11}}{2}-\frac{\sqrt{(C_{11}-C_{44})^2+4C_{14}^{\;2}}}{2}$
Y-cut	Slow shear (SS)	$C_{66}+\frac{e_{11}^2}{\varepsilon_{11}}$
Z-cut	Longitudinal (L)	C_{33}
Z-cut	Degenerate shear (DS)	C_{44}
-45^0 rotated around X	Slow shear (SS)	$\frac{1}{4}(C_{44}+C_{66}+2C_{14})+\frac{1}{2}\frac{(e_{11}-e_{14})^2}{\varepsilon_{11}+\varepsilon_{33}}$
-45^0 rotated around X	Quasi-longitudinal (QL)	$\frac{1}{4}(C_{11}+2C_{14}+2C_{44}+C_{33})+\frac{1}{4}\sqrt{(2C_{14}+C_{11}-C_{33})^2+4(C_{13}+C_{44}+C_{14})^2}$
-45^0 rotated around X	Fast quasi-shear (QS)	$\frac{1}{4}(C_{11}+2C_{14}+2C_{44}+C_{33})-\frac{1}{4}\sqrt{(2C_{14}+C_{11}-C_{33})^2+4(C_{13}+C_{44}+C_{14})^2}$

TABLE II. MATERIAL PARAMETERS OF SNGS SINGLE CRYSTAL AND THEIR TEMPERATURE COEFFICIENTS

Material constant	Value at T = 298 K	First order TC, in 10^{-6} / K	Second order TC, in 10^{-9} / K^2
C_{11}	155.2 GPa	-157.6	-553
C_{12}	68.2 GPa	-192	394
C_{13}	82.6 GPa	0.7	350
C_{14}	4.4 GPa	843	-5380
C_{33}	201.5 GPa	-135	-515
C_{44}	54.9 GPa	-238	-1021
e_{11}	0.478 C/m^2	-520	
e_{14}	0.15 C/m^2		
$\varepsilon_{11}/\varepsilon_0$	12.74	174.5	2.09
$\varepsilon_{33}/\varepsilon_0$	20.6	16.5	4.8
α_{11}		6.5	6.1
α_{33}		7.5	4.2
ρ	4650 kg/m^3		

VII. Conclusion

A full set of the dielectric, elastic and piezoelectric constants as well as the thermal coefficients of expansion for the $Sr_3NbGa_3Si_2O_{14}$ (SNGS) single crystal were obtained in a temperature range from 220 K to 310 K. Using the experimental data, temperature coefficients of material parameters were calculated. Although temperature compensated cuts for sound velocities in the measured temperature range were not found, the obtained results show that SNGS is a promising material for sensor applications at elevated temperatures.

Acknowledgment

The authors would like to thank G. Behr from Leibniz Institute for Solid State and Materials Research Dresden for the samples preparation.

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