

# DIELECTRIC PROPERTIES OF LANGASITE MEASURED WITH THE WHISPERING GALLERY MODE TECHNIQUE

V.Giordano<sup>a)</sup>, J. G. Hartnett<sup>b)</sup>, J. Krupka<sup>c)</sup>, Y. Kersalé<sup>a)</sup>, P.Y. Bourgeois<sup>a)</sup> and M.E. Tobar<sup>b)</sup>.

<sup>a)</sup> LPMO CNRS, 32 av. de l'Observatoire, F-25044 Besançon, France  
fx +33.381.666998, e-mail giordano@lpmo.edu

<sup>b)</sup> Department of Physics, University of Western Australia  
35 Stirling Hwy, Crawley WA 6009 Australia

<sup>c)</sup> Institute of Microelectronics and Optoelectronics, Dep. of Electronics  
Warsaw University of Technology - Koszykowa 75, 00-662 Warsaw Poland

*In a search of efficient dielectric materials for microwave applications, single crystal materials have to be considered. They generally present low dielectric loss and as a result can be used to design high-Q resonators or sharp band-pass filters. Sapphire, Quartz and Rutile have been extensively studied and have demonstrated impressive performance as high-Q resonator media. Nevertheless, these materials present high temperature sensitivity of permittivity leading to poor long term stability performance. Thus, there is a great interest in the measurement of other dielectric crystals to extend the range of available materials for microwave resonator design. In a preceeding paper<sup>1</sup>, preliminary measurements on Lanthanum Gallium Silicate (LGS) were presented. Although, the available sample that was used in the first attempt was of poor crystal quality, interesting properties for the design of a self-compensated resonator was demonstrated. In this paper, we present the results of a new investigation conducted on a higher quality crystal and based on the whispering gallery mode technique. This more accurate method leads to better permittivity and dielectric loss evaluations. We report measurements of dielectric permittivity and dielectric loss at microwave frequencies of single crystal Lanthanum Gallium Silicate between 4.2K and 300K.*

## 1 Introduction

Langasite (LGS) and other compounds of the same family were first discovered at the end of the seventies. Developed for laser applications, they have since been studied for their piezoelectric properties leading to the achievement of efficient acoustic wave filters and resonators<sup>2</sup>. Although LGS dielectric properties at low and radio frequencies have been already measured by several authors<sup>3;4</sup>, only recently, its use as a microwave dielectric medium has been considered<sup>5</sup>.

LGS is an anisotropic crystal with two relative permittivity components  $\epsilon_{\perp}$  and  $\epsilon_{\parallel}$  in the directions perpendicular and parallel to the crystal C-axis respectively. One of the main interests of LGS is that  $\epsilon_{\perp}$  and  $\epsilon_{\parallel}$  present temperature coefficients of opposite sign. It has been then demonstrated that this property can be exploited to design a self-temperature-compensated microwave resonator<sup>1</sup>. Other dielectric monocrystals, such as sapphire, quartz or rutile, don't exhibit such behavior. For these materials, the lack of a turnover temperature induces a high frequency-temperature sensitivity, which alters the long term frequency stability performance. Unfortunately, our first attempt to evaluate the dielectric properties of LGS in the microwave domain were conducted with a small sample machined from a low quality crystal. Interesting results were produced, nevertheless we were unable to determine LGS dielectric losses with any great accuracy.

In this paper, we present dielectric measurements conducted on a larger, better quality crystal in order to evaluate more accurately the potential of LGS as dielectric material for microwave applications. The whispering gallery mode resonator method<sup>6</sup> has been used, leading to a more accurate determination of LGS dielectric properties.

## 2 Methodology

To evaluate the dielectric parameters, i.e. relative permittivity  $\epsilon_r$  and the loss tangent  $tg\delta$ , of any dielectric material in the microwave frequency range, the resonant methods yield the more accurate results. A dielectric sample is inserted in a structure to form a microwave resonator of which an accurate theoretical description is available. Then a well identified mode is excited and its frequency ( $\nu$ ) and its Q-factor (Q) are measured. The dispersion equation for the considered mode is numerically solved to evaluate  $\epsilon_r$ . Once the permittivity is known, the electric filling factor inside the dielectric sample can be calculated and then  $tg\delta$  is evaluated from the measured Q-factor.

Although the resonant structure can be arbitrary chosen, the configuration using Whispering Gallery Modes (WGM) of a cylindrical dielectric sample appears the most suitable. Indeed WGM are characterized by an efficient confinement of the electromagnetic fields inside the dielectric sample.

Provided that the mode of interest is a sufficiently high order mode, the frequency and Q-factor are neither affected by resonator walls nor by its mounting structure. It then results in an accurate determination of both  $\epsilon$  and  $tg\delta$ .

In the case of an anisotropic dielectric sample, the symmetry axis of the cylindrical resonator is choosen parallel to the crystal C-axis. Two WG modes are then required to evaluate the complete set of dielectric parameters:  $\epsilon_{\perp}$ ,  $\epsilon_{\parallel}$ ,  $tg\delta_{\perp}$  and  $tg\delta_{\parallel}$ .

### 3 Resonator Mounting and Mode Identification

Our preceeding measurements were conducted on a sample cut from a crystal grown about ten years ago. It was not transparent but a pale yellowish color. Moreover, we only had at our disposal a small cylindrical sample about 10 mm in diameter and 12 mm high. We tried to excite WG modes in this resonator without success. Low-Q resonances were numerous and it was not possible to identify clearly any WG modes of resonance.

We recently got a higher quality crystal from Fomos Technology (Russia). This new sample is transparent with a bright orange colouring. It has a diameter of 25 mm and a thickness of 20 mm with all surfaces polished. The C-axis is aligned with the cylinder axis within  $1^{\circ}$ . A 4 mm hole has been machined along this axis to enable a rigid mechanical mounting in the experimental set-up (schematic shown in figure 1). The LGS sample was supported by two copper

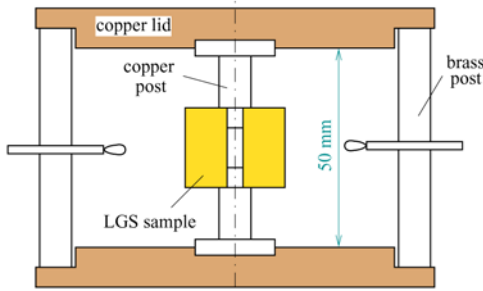


Figure 1: Resonator used to excite Whispering Gallery Modes in the LGS resonator

posts and placed between two cylindrical copper lids connected by side brass posts. These posts also support the coupling probes. There was no cylindrical metallic wall around the dielectric sample. Providing that the interior wall of the cryogenic can has been covered with an efficient microwave absorber, this open resonator configuration attenuates most of the spurious modes of resonance. As the value of LGS permittivity is relatively high, the electromagnetic field is well confined in the dielectric and the radiation losses are expected to be negligible.

The radial positioning of the probes has been set to obtain small coupling coefficients, less than 0.05 in the whole temperature range. As a result, the measured Q-factor is assumed to be the unloaded Q-factor.

We identified a few WG modes that are given in the following table.

Table 1: mode identification in LGS sample. The modes, noted in bold, were later used for the dielectric permittivity and loss tangent measurements

frequency (GHz)	m		
3.347	3	WGH <sub>3,0,0</sub>	N <sub>13</sub>
3.916	4	WGH <sub>4,0,0</sub>	N <sub>14</sub>
4.494	5	WGH <sub>5,0,0</sub>	N <sub>15</sub>
5.061	4	WGH <sub>3,?,1</sub>	?
5.077	6	WGH <sub>6,0,0</sub>	N <sub>16</sub>
5.530	5	WGH <sub>5,?,1</sub>	?
<b>5.659</b>	7	<b>WGH</b> <sub>7,0,0</sub>	N <sub>17</sub>
6.244	8	WGH <sub>8,0,0</sub>	N <sub>18</sub>
5.880	3	WGE <sub>3,0,0</sub>	S <sub>33</sub>
5.887	?	?	?
<b>6.892</b>	4	<b>WGE</b> <sub>4,0,0</sub>	S <sub>34</sub>
<b>7.917</b>	5	<b>WGE</b> <sub>5,0,0</sub>	S <sub>35</sub>
8.935	6	WGE <sub>6,0,0</sub>	S <sub>46</sub>

In this table two common mode nomenclatures are given:

- $WGH_{m,n,l}$  for quasi-TM mode or  $WGE_{m,n,l}$  for quasi-TE. The subscripts  $m, n$  and  $l$  refer respectively to the azimuthal, radial and axial variation of the electromagnetic field components.

- $Nk_m$  or  $Sk_m$ . This notation due to Krupka<sup>7</sup> appears more suitable in the case of material presenting an high anisotropy ratio, as is LGS where  $\frac{\epsilon_{\perp}}{\epsilon_{\parallel}} \approx 3$ . Most of the WG modes are hybrid in nature and the common designation based on the concept of quasi-TM or quasi-TE modes has no real physical meaning.  $N$  ( $S$ ) refers to a nonsymmetric (symmetric) magnetic field distribution along the axial direction. The number  $k$  designates the frequency order of the mode and the subscript  $m$  is again the azimuthal mode number.

The identification of  $WGE$  modes turned out to be difficult. The resonance frequencies were higher than  $WGH$  modes and spurious resonances were numerous. These spurious resonances correspond to hybrid  $WGH$ -like modes or to high order modes presenting multiple field variations in the axial or

radial directions, i.e.  $n \neq 0$  and/or  $l \neq 0$ .

#### 4 Permittivity measurement results

The resonator were placed successively in a liquid helium bath cryostat (at UWA) and in a cryocooler (at LPMO). Cooling was first achieved before increasing the temperature slowly in order to follow the test mode and to measure its frequency and Q-factor. The  $N1-7(WGH_{7,0,0})$  and  $S3-4(WGE_{4,0,0})$  modes were selected due to the absence of nearby spurious modes. The following figures show the experimental results. The anisotropy of LGS is clearly shown in figure 2.

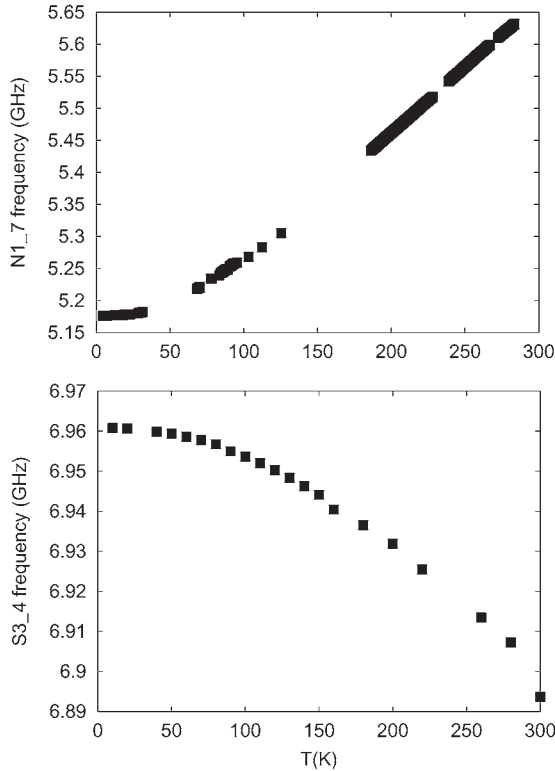


Figure 2:  $N1-7$  and  $S3-4$  resonance frequency vs temperature

The frequency temperature coefficients of the two considered modes are opposite in sign. Over the whole temperature range, the relative frequency change for the  $N1_7$  mode is about 10 percent, whereas, it is about 1 percent for  $S3_4$ . We can expect that  $\epsilon_{\perp}$  has a positive temperature coefficient about 10 times smaller than that for  $\epsilon_{\parallel}$ . The two anisotropic permittivity components were calculated using this experimental data and published thermal expansion coefficients for LGS [1]. The results are shown in the figure 3.

As expected,  $\epsilon_{\perp}$  and  $\epsilon_{\parallel}$  present opposite temperature coefficients. This fact allows for some mode to have self-compensated behavior. Indeed neglecting thermal expansion as a second order effect, the frequency temperature dependance of any resonance

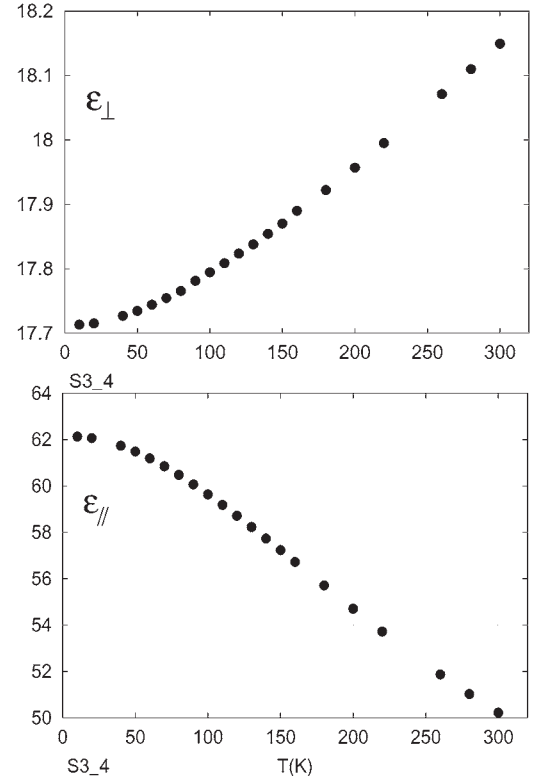


Figure 3: LGS relative permittivity components as a function of temperature

can be written as:

$$\frac{1}{f} \frac{\partial f}{\partial T} \approx -\frac{1}{2} p_{\perp} \left[ \frac{1}{\epsilon_{\perp}} \frac{\partial \epsilon_{\perp}}{\partial T} \right] - \frac{1}{2} p_{\parallel} \left[ \frac{1}{\epsilon_{\parallel}} \frac{\partial \epsilon_{\parallel}}{\partial T} \right] \quad (1)$$

$p_{\perp}$  and  $p_{\parallel}$  are the electric energy filling factors in the dielectric sample perpendicular and parallel to the z-axis respectively. Temperature compensation occurs when the two right hand side terms in equation (1) are equal in magnitude but opposite in sign. As  $\left[ \frac{1}{\epsilon_{\parallel}} \frac{\partial \epsilon_{\parallel}}{\partial T} \right]$  is larger than  $\left[ \frac{1}{\epsilon_{\perp}} \frac{\partial \epsilon_{\perp}}{\partial T} \right]$  self compensation occurs only in WGE-like modes for which  $p_{\perp} \gg p_{\parallel}$ . This is demonstrated by the  $S3_5$  mode behavior depicted in figure 4.

In this figure the experimental data are represented by the solid line whereas squares correspond to some calculated frequencies. The latter have been obtained from the Mode Matching model of the resonator and the preceding measured permittivities. These results demonstrate that the  $S3_5$  mode exhibits a turnover temperature at about 190 K. This comparison strenghtens our confidence in the measured permittivity values.

#### 5 Dielectric losses evaluation

Figure 5 represents the Q-factor values as a function of temperature for the 3 resonance modes:  $N1_7$ ;  $S3_4$  and  $S3_5$ .

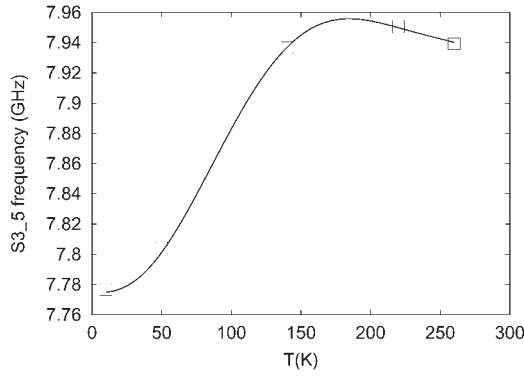


Figure 4:  $S3_5$  mode frequency vs temperature; *solid line* experimental data;  $\square$  calculated frequencies

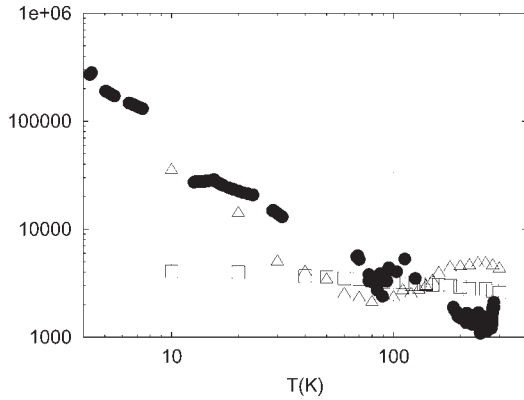


Figure 5: Q-factor vs temperature for different modes:  $\square$   $S3_4$ ;  $\triangle$   $S3_5$ ;  $\bullet$   $N1_7$

The Q-factor of the  $S3_4$  mode appears limited by some extra losses, which may be due to radiation. As a result, this low order mode ( $m = 4$ ) is not suitable for the evaluation of LGS dielectric losses. To determine a preliminary value of the loss tangent, we assumed the electric filling factors of the  $N1_7$  and the  $S3_4$  modes to be constant as a function of temperature and calculated values at 300K to be:

The loss tangent values were then extracted from:

	mode 1	mode 2
	$N1_7$	$S3_5$
$p_{\perp}$	0.1071	0.9502
$p_{\parallel}$	0.8873	0.0205

$$Q_1^{-1} = p_{\perp 1} \text{tg} \delta_{\perp} + p_{\parallel 1} \text{tg} \delta_{\parallel}$$

$$Q_2^{-1} = p_{\perp 2} \text{tg} \delta_{\perp} + p_{\parallel 2} \text{tg} \delta_{\parallel}$$

where the subscripts 1 and 2 refer to the  $N1_7$  and the  $S3_5$  modes, respectively. By solving these linear equations at a number of temperature points, we get for the anisotropic loss tangent the values presented

in figure 6. It is also assumed the mode frequencies are sufficiently close to not appreciably affect the calculation.

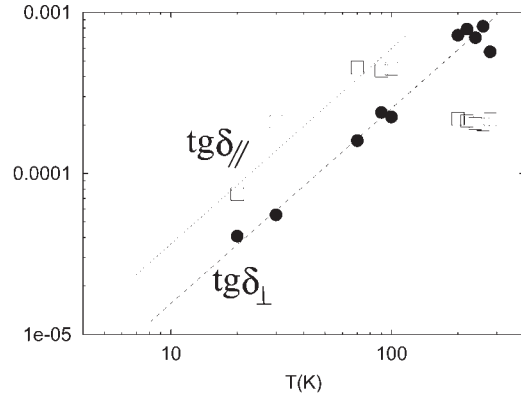


Figure 6: Loss tangents vs temperature:  $\square$   $\text{tg} \delta_{\parallel}$ ;  $\bullet$   $\text{tg} \delta_{\perp}$

## 6 Conclusion

The whispering gallery mode method has been used to determine the dielectric properties of Lanthanum Gallium Silicate over the temperature range 4–300K at microwave frequencies. LGS presents two permittivity components having opposite temperature coefficients. Although LGS appears to be a relatively high dielectric loss material compared to sapphire or quartz, it may be exploited in designing self-temperature-compensated microwave filters.

## References

- [1] V. Giordano, Y. Kersalé, and J. Boy, "Measurement at microwave frequencies of dielectric properties of  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  between 10k and 400k," in *Proc. 15th European Frequency and Time Forum*, (Neuchâtel (CH)), pp. 183–186, Mars 6–8th 2001.
- [2] G. D. Mansfeld, "Langasite as a material for piezoelectric devices," in *Proc. 12th European Frequency and Time Forum*, (Warsaw, Poland), pp. 61–65, Tele and Radio Research Institute, Warsaw, Poland, Mar. 10–12, 1998.
- [3] B. Mill and Y. Pisarevsky, "Langasite-type materials: from discovery to present state," in *Proc. 2000 IEEE/EIA Int. Frequency Control Symposium*, pp. 133–144, 2000.
- [4] I. Silvestrova, Y. Pisarevskii, P. Senyushchenkov, and A. Krupnyl, "Temperature dependences of the elastic properties of  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  single crystals," *Sov. Phys. Solid State*, vol. 28, pp. 1613–1614, Sept. 1986.
- [5] V. Giordano, Y. Kersalé, and J. Boy., "Dielectric properties of  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  at microwave frequencies between 10k and 400k," *Applied Physics Letters*, vol. 78, Apr. 2001.
- [6] J. Krupka, K. Derzakowski, A. Abramowicz, M. Tobar, and R. Geyer, "Complex permittivity measurements of extremely low loss dielectric materials using whispering gallery modes," in *IEEE MTT-S Int. Microwave Symp. Dig.*, 1997.
- [7] J. Krupka, D. Cros, A. Luiten, and M. Tobar *Electronics Letters*, vol. 32, pp. 670–671, 1996.