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DIELECTRIC PROPERTIES OF SINGLE CRYSTAL FLUORIDES AT MICROWAVE FREQUENCIES AND CRYOGENIC TEMPERATURES

J. G. Hartnett[1], A. Fowler[1], M. E. Tobar[1], J. Krupka[2]
[1]School of Physics, University of Western Australia, 35 Stirling Hwy, Crawley, WA 6009, Australia
[2]Instytut Mikroelektroniki i Optoelektroniki Politechniki Warszawskiej, Koszykowa 75, 00-662 Warszawa, Poland

*Abstrac*t - **Whispering gallery mode method was used for very accurate permittivity, and the dielectric loss measurements of LiF, and CaF_2 over the temperature range of 4 K – 300 K. This method is the most accurate for determining the loss tangent of very low loss materials. The absolute uncertainty in the real part of permittivity was estimated to be less than 0.1% and it was limited principally by uncertainty in dimensions of the samples. Dielectric loss tangents were measured with uncertainties of about 10% limited by accuracy of Q-factor measurements of whispering gallery modes. For the measured materials dielectric losses varied as a function temperature by a few orders of magnitude, exhibiting dielectric losses of 3×10^{-6} at 4 K. As expected, at temperatures below 10 K turning points were observed in the frequency-temperature dependence of modes probably due to paramagnetic impurity ions, leftover from the manufacturing process. CaF_2 exhibited a turning point at 7.4 K in a whispering gallery mode at 17.5 GHz and a Q-factor of 3.45×10^6. LiF exhibited a turning point at 5.3 K in a whispering gallery mode at 13.5 GHz and a Q-factor of 2.55×10^6.**

Keywords **- dielectric, permittivity, loss tangent, flourides**

I. INTRODUCTION

The Whispering Gallery Mode (WGM) method has proven to be one of the most accurate for measurements of complex permittivity of extremely low loss dielectrics. In earlier papers only the dielectric loss tangent was measured with this technique [1, 2]. However, in recent years the WGM method has been employed for very precise measurements of the real permittivity and the dielectric losses of both isotropic and uniaxial anisotropic materials [3, 4]. The method has also been applied to the measurement of ultra-low loss single crystal materials including sapphire, YAG, quartz, rutile, $SrLaAlO_4$ [5], Titanium doped sapphire [6], and Chromium doped YAG [7] .

II. METHODOLOGY

The use of WGM modes, with high order azimuthal mode numbers, excited in cylindrical dielectric materials, is the most effective way to eliminate conductor losses and hence make accurate dielectric loss tangent measurements. In order to find permittivity tensor components of uniaxial anisotropic materials, the cylindrical specimen is obtained with the cylinder axis oriented along the crystal axis. In the case of isotropic materials, as are Calcium and Lithium Flouride, only one mode of resonance is chosen that exhibits WG-mode electromagnetic field structure. Usually this is from the WGH-mode family as they have the highest Q-factors for a given frequency and are less limited by wall losses. Finally a non-linear determinant equation is solved to evaluate the permittivity,

$$F(f, \varepsilon_r) = 0 \tag{1}$$

where f is the measured resonance frequency for the WG mode, and ε_r is the real part of the permittivity. The eigenvalue equation represented by F results from application of a rigorous method of analysis of the resonant structure (which in our case was a mode matching technique [8]). In the case of Calcium Flouride (CaF_2) and Lithium Flouride (LiF), reported here, only isotropic permittivity and loss tangent need be considered.

Once the permittivity is calculated from (1), the dielectric loss tangent is evaluated from (2),

$$\mathrm{Q}^{-1} = p_e \tan\delta + R_S / G \tag{2}$$

where $tan\delta$ is the dielectric loss tangent; p_e is the electric-energy filling factor of the resonant structure and G is the geometric factor for the WG mode. The electric energy filling factor was determined from the incremental frequency rule [9]:

$$p_e = 2\left|\frac{\partial f}{\partial \varepsilon_r}\right|\frac{\varepsilon_r}{f} \tag{3}$$

and geometric factors were found from

$$G = \frac{\omega \iiint_V \mu_0 H \cdot H^* \mathrm{dv}}{\oiint_S H_t \cdot H_t^* \mathrm{ds}} \tag{4}$$

where ω is the angular frequency of the resonance, μ_0 the permeability of free space, H the magnetic field strength, and H_t the magnetic field strength transverse to the metal walls. In practice for properly chosen azimuthal mode numbers and sufficiently large metal shield radius, any term involving the geometric factor can be neglected.

The cylindrical dielectric sample is loaded into a cylindrical copper cavity (Fig. 1). The families of WG modes are identified with the method described above and a suitable mode is chosen whose Q-factor is not limited by wall losses. This is usually for modes with azimuthal mode numbers m ≥ 8. Once the mode is clearly identified at room temperature the cavity is inserted into a vacuum chamber and cooled to 77 K with liquid nitrogen and then to 4 K with liquid helium.

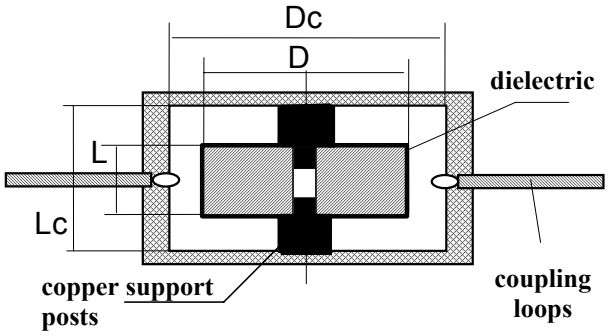

Fig. 1: Copper cavity used to make measurements.

The dimensions of the cavity and dielectric samples were as follows. (Note: reference is made to parameters in fig. 1.)

A) The inside diameter of the cavity used to measure the CaF_2 was Dc = 34.80 ± 0.01 mm and the height was Lc = 25.66 ± 0.01 mm. The diameter of the CaF_2 sample was D = 24.90 ± 0.01 mm and height L = 9.88 ± 0.01 mm.

B) The inside diameter of the cavity used to measure the LiF was Dc = 50.00 ± 0.01mm and the height was Lc = 27.80 ± 0.01 mm. The diameter of the LiF sample was D = 33.965 ± 0.005 mm and height L = 11.975 ± 0.005 mm.

C) Both samples had a 3 mm hole through their center along the cylinder axis. They were polished on all surfaces except this hole.

The resonance modes were excited with a Marconi 2030 synthesizer, which was mixed with a HP 8673G synthesizer. The microwave power was coupled into the cavity via coaxial transmission lines into the cryogenic environment and coupled to the resonance modes via loop probes (shown in fig. 1). A computer-controlled program [10] stepped through the resonance and fitted a Lorenztian curve to the data of the microwave detector power as a function of frequency. The temperature was monitored with a carbon glass sensor attached to the copper cavity while the cavity itself was allowed to slowly warm. This was a very slow process (taking the order of 4 days), which meant that the data was taken in a quasi-equilibrium state. Measurements indicate a temperature error of order 0.1 degrees near 77 K but about 0.5 K near 4 K where the thermal gradients are larger.

The modes chosen for this analysis were the $WGH_{8,0,0}$ mode in the CaF_2 sample with a nominal frequency of 17.5 GHz and the $WGH_{10,0,0}$ mode at 13.5 GHz in the LiF sample. The loaded Q-factors were recorded as a function of temperature as described above. Also the program calculated the coupling coefficients and with which we calculated the unloaded Q-factors. See fig. 2. A 10% error is assigned to this measurement mostly due to Lorentzian line fit errors. This is mostly caused by interactions with background spurious modes in the crystal, unmatched VSWR in the transmission lines and loop probe reactance. Within this level of error both dielectrics exhibited the same unloaded Q-factor at the respective frequencies. This was about 3×10^6. At higher temperatures, however this was not the case. At 300 K, the Q-factor of the $WGH_{8,0,0}$ mode (17.5 GHz) in the CaF_2 was about 5×10^4, while for the $WGH_{10,0,0}$ mode (13.5 GHz) in the LiF sample it measured over 1×10^5.

The measured frequencies for the $WGH_{10,0,0}$ mode (13.5 GHz) in the LiF dielectric and the $WGH_{8,0,0}$ mode (17.5 GHz) in the CaF_2 are shown in figs 3 and 4 respectively. In fig. 3 a single data point is shown for the LiF resonance just below 200 K. In fig. 4 a few points are shown just above 200 K. These were obtained using dry-ice (solid CO_2) with the resonator sitting in the dry-ice. As a result the error in the quoted temperature may be larger than with other data points. In both cases, the data appear not to fall on the natural curve (broken line) drawn through the data. These points are consistent with temperature scale errors of the order of a few degrees.

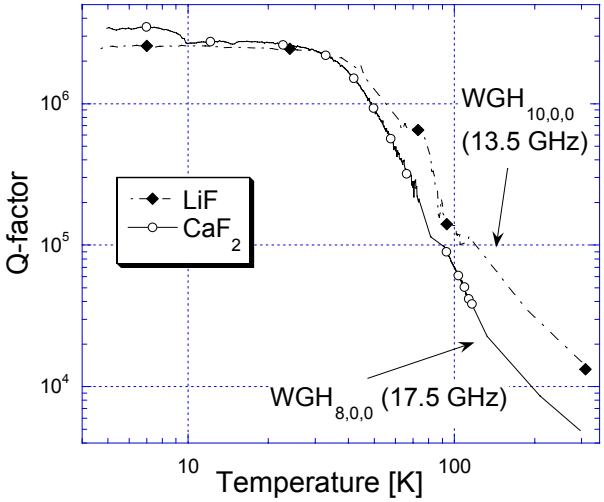

Fig 2. Q-factors of the WGH modes measured in the two dielectric samples. For clarity only 3% of the measured data points are shown.

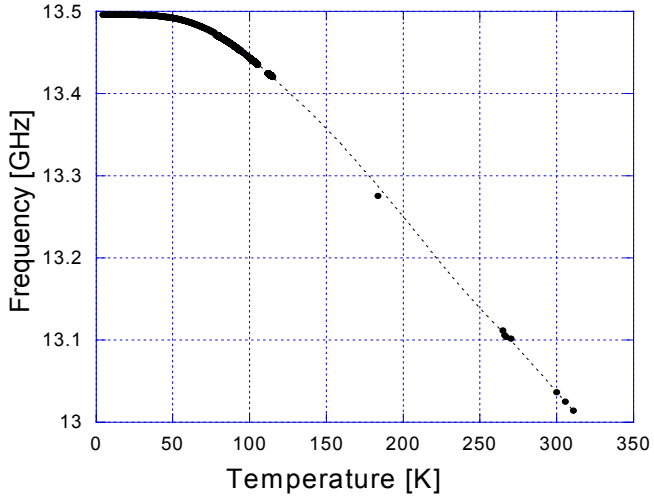

Fig 3. The frequency of the $WGH_{10,0,0}$ (13.5 GHz) mode in LiF as a function of temperature.

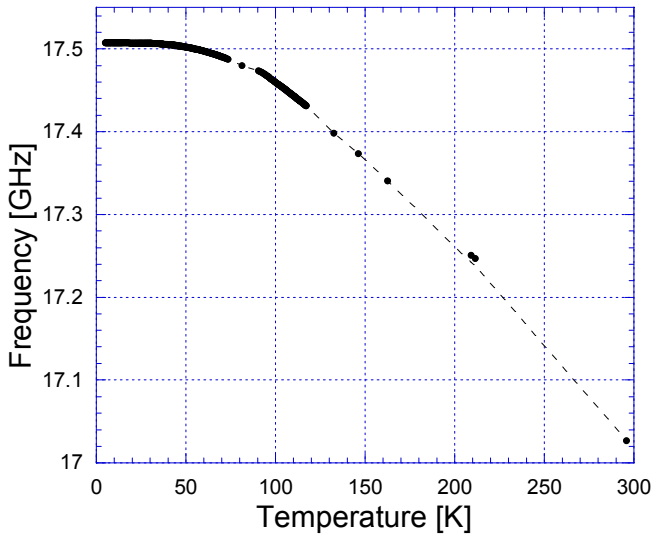

Fig 4. The frequency of the $WGH_{8,0,0}$ (17.5 GHz) mode in CaF_2 as a function of temperature.

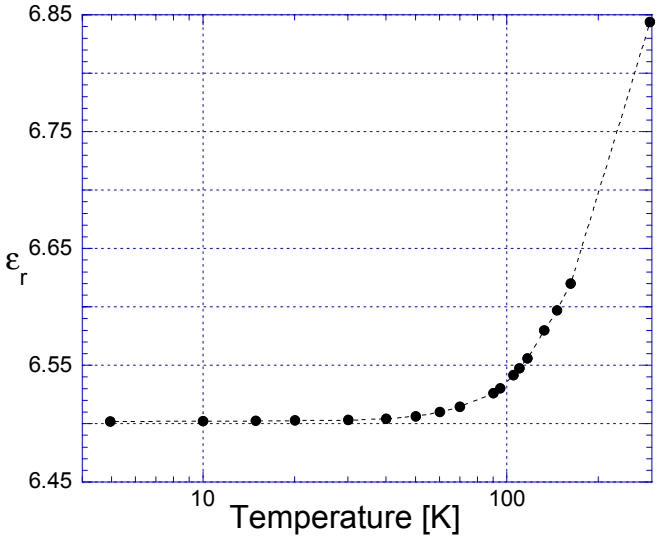

Fig 5. The real part of the relative permittivity of CaF_2 as a function of temperature.

For measurements above 250 K, a Peltier controlled vacuum chamber was used with a 1000 Ω thermistor as the sensor. In that temperature region the temperature errors are much less than 0.1 K.

III. Results

A. Permittivity

From the measurement data of figs 3 and 4 we calculated the real part of the relative permittivity as a function of temperature using (1). The thermal expansion of the crystals and the copper cavities were taken into account. That data was derived from [11]. The results are shown in fig. 5 for the CaF_2 crystal and in fig 6 for the LiF crystal. The absolute accuracy of measurements for the real permittivity was limited principally by the uncertainty in the dimensions of the samples and estimated to be better then 0.1%. The relative accuracy versus temperature depends on inaccuracies in the thermal expansion coefficients data. Influence of thermal expansion is especially important at low temperatures when permittivity changes are small. At temperatures in the range of 4 -10 K presence of paramagnetic impurities often affect the measurement uncertainty of the real permittivity. Though they are not apparent in figs 5 and 6, closer examination of the frequency data indicates otherwise. Resonance frequency changes below 10 K mostly depend on the kind and amount of paramagnetic impurities. This is discussed later.

Analysis of the permittivity data show that the derivatives of the permittivity data for the two dielectric fluorides measured here are identical between 4 K and 120 K within limits of the measurement uncertainty. A polynomial fit yields

$$\frac{d\varepsilon}{dT} = 6.961\times10^{-5} - 1.367\times10^{-5}T + 5.785\times10^{-7}T^2 - 2.541\times10^{-9}T^3 \quad (5)$$

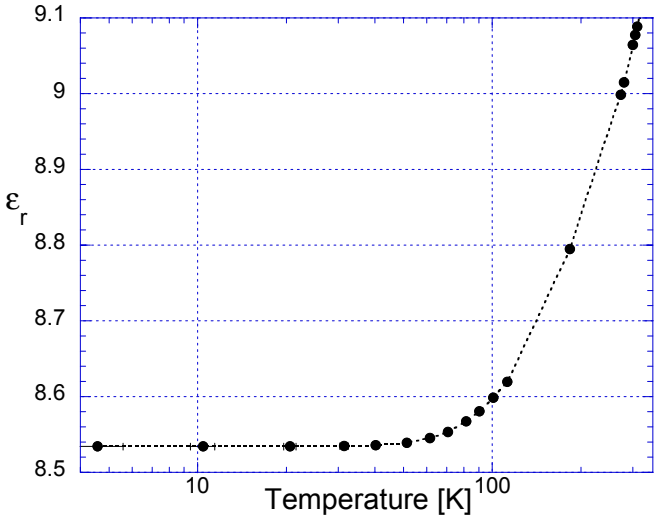

Fig 6. The real part of the relative permittivity of LiF as a function of temperature

B. Loss Tangent

For the chosen modes, the $WGH_{8,0,0}$ mode in CaF_2 and the $WGH_{10,0,0}$ mode in LiF, the effect of the copper conducting shield was neglected in (2). Using (3), (2) was then solved for the loss tangent ($tan\delta$) as a function of temperature from the data of fig. 2. The results are shown in fig. 7. The errors associated with curve fitting to the resonance line limit the error here to about 10%. Curve fitting to the $tan\delta$ data in fig. 7 for temperatures above 40 K yields very similar power law dependence. For the CaF_2 (curve 1) loss tangent we get

$$\tan\delta = 1.056\times10^{-11}T^{2.88} \quad (6)$$

and for LiF (curve 2) we get

$$\tan\delta = 0.895\times10^{-11}T^{2.92}. \quad (7)$$

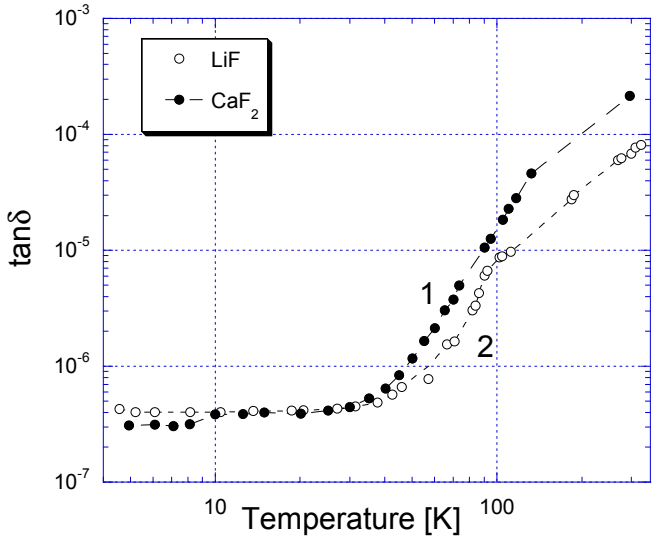

Fig 7. The loss tangent for CaF_2 (curve 1) and LiF (curve 2) as a function of temperature

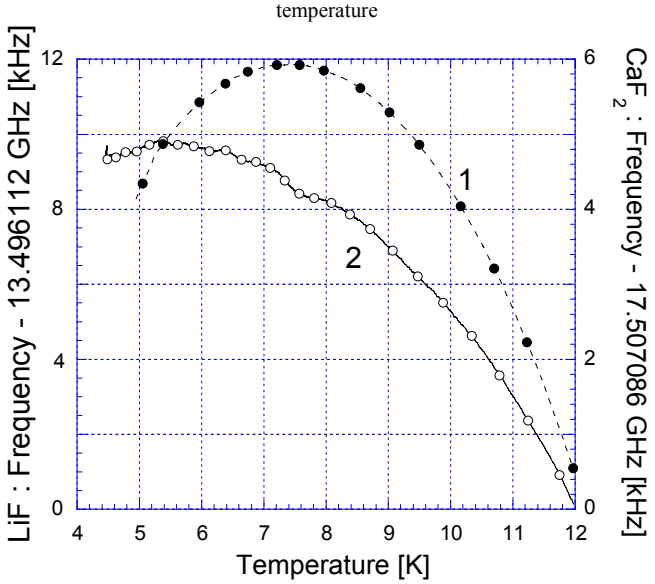

Fig 7. The frequency-temperature dependence for CaF_2 (curve 1) and LiF (curve 2) below 12 K.

IV. Discussion

The measured frequency-temperature data for both modes exhibited turning points below 8 K and are shown in fig. 8. The WG mode in CaF_2 turns over at 7.4 K (curve 1) while the WG mode in the LiF turns over at 5.3 K (curve 2). These turning points can be attributed to incidental paramagnetic ions, leftover from the manufacturing process. This work has been part of a program to find high-Q dielectric materials that have suitable turnover temperatures for the design of temperature compensated resonator oscillators. Previously we have examined various materials [5-7] where these were shown to exist. In those cases all could be attributed to paramagnetic ions. Therefore the relative permittivity values shown in figs 5 and 6 include small errors as a result of not allowing for these turning points.

V. Conclusion

Two single crystal dielectric materials, Calcium and Lithium Flouride, have been characterized at microwave frequencies and as a function of temperature between 4 K and 300 K. Their relative permittivity (ε_r) and loss tangent ($\tan\delta$) have been calculated and presented here. They were found to be very low loss with permittivity variations of 6.50 (4 K) to 6.84 (300 K) for CaF_2 and 8.53 (4 K) to 9.06 (300 K) for LiF. Loss tangents varied from about 4×10^{-6} (4 K) to about 10^{-4} (300 K).

Acknowledgment

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