DIELECTRIC PROPERTIES OF HIGH-T_c SUBSTRATES UP TO 40 GHz

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ABSTRACT

Dielectric properties of $CaNdAlO_4$, $LaAlO_3$, $SrLaAlO_4$, $SrLaGaO_7$ and $NdGaO_3$ monocrystals, prospective substrate materials for the deposition of high-Tc superconductors, were measured with high accuracy at frequencies up to 40 GHz in the temperature range 10 to 300 K. Most materials exhibit anisotropy while in $CaNdAlO_4$ and $NdGaO_3$ pronounced loss increase was found at temperatures below 100 K, probably of magnetic origin.

I. INTRODUCTION

A remarkable effort has been made to characterize the dielectric properties of prospective monocrystalline substrates for growing thin film of high-T_c superconductors ([1] [2] and references therein). However, the design of microwave superconducting devices in both coplanar and microstrip configurations requires much more accurate data than those as yet published. In this paper we present the results of a systematic study of the dielectric permittivities of $CaNdAlO_4$, $LaAlO_3$, $SrLaAlO_4$, $SrLaGaO_7$ and $NdGaO_3$ monocrystals in the frequency range from 4 GHz to 40 GHz based on the measurement of self-resonances of a rectangular cavity completely filled with the investigated material in the temperature range from 10 K to 300 K. In most cases our measurements improve the existing data by at least one order of magnitude and considerably extend the frequency range in which the a.m. materials are characterized.

II. EXPERIMENTAL

The samples of $CaNdAl0_4$, $SrLaAlO_4$, and $SrLaGaO_7$ were prepared from single crystals, exactly X-ray oriented and cut along the crystallographic axes to form a set of 4 rectangular parallelepipeds (with dimensions of about 6.04 x 5.05 x 2.56 mm^3), which were polished to get an optical quality surface on all 6 faces. For anisotropic crystals, two of the parallelepipeds were c-axis oriented along the narrowest side (referred further to as samples A), while the other two had c-axis directed along the wider side (samples B). The samples of $LaAlO_3$ and $NdGaO_3$ had the form of 10 x 10 x .5 mm^3 rectangular parallelepipeds. Small deviations from perfect geometry were found on the level of 0.05% and averaged final dimensions were used in the calculations. The samples were covered on all 6 faces by a thin (~30 nm), sputtered gold film on which a thicker (~30μm), continuous layer of the same metalwas electrolytically deposited. In the final step of sample preparation the gold layer was cut to expose small, 0.4 x 0.4 mm^2 coupling holes in opposing cavity walls. The samples were placed with coupling holes facing very small (~0.3 mm^2) coupling loops terminating two microstrip input and output lines. The transmission losses of the were measured using either a network analyzer or the computer controlled GPIB driven set of separate instruments. Depending on the sample, resonant frequencies of 20 to 32 different cavity modes (from 4 to 40 GHz) were identified and the effective Q-factors measured, though some of them were too weakly coupled to assure good accuracy of the measurements in the full temperature range.

III. MEASUREMENTS AND DISCUSSION

Microwave measurements of anisotropic materials are difficult and require special care in setting-up the experiment, and interpretation of the experimental data. Most of the investigated crystals exhibit uniaxial anisotropy.

The cavity size along optical axis is denoted by c and other two sizes as a and b. The relative permittivity tensor components are $\varepsilon_a, \varepsilon_b, \varepsilon_c$, and the relative permeability tensor components μ_a, μ_b, μ_c. If uniaxial anisotropy is assumed then,

$$\varepsilon_a = \varepsilon_b = \varepsilon, \quad \mu_a = \mu_b = \mu.$$

For this case the resonant frequencies of particular modes become true TM_m and TE_m modes with respect to c-axis and are determined by the set of equations [3], [5]

$$f^2_{TM_m} = \frac{1}{4\mu_o\varepsilon_o\mu}\left(\frac{k^2_{m_c}}{\varepsilon} + \frac{k^2_{m_a}}{\varepsilon_c} + \frac{k^2_{m_b}}{\varepsilon_c}\right), \quad k^2_{m_c} \neq 0, \tag{1a}$$

$$f^2_{TE_m} = \frac{1}{4\mu_o\varepsilon_o\varepsilon}\left(\frac{k^2_{m_c}}{\mu} + \frac{k^2_{m_a}}{\mu_c} + \frac{k^2_{m_b}}{\mu_c}\right), \quad k^2_{m_{a,b}} \neq 0, \tag{1b}$$

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where $m(m_1, m_2, m_3)$ is the mode index and k_a, k_b and k_c are the wave numbers.

Observe that if the relative permeabilities $\mu \neq \mu_c \neq 1$, than the resonant frequency measurements allow to determine only three quantities e.g. $\mu\varepsilon_c$, μ_c/μ and $\mu\varepsilon$ instead of the four unknowns μ, μ_c, ε and ε_c.

Equations (1a) and (1b) are valid for a cavity completely enclosed by perfectly conducting walls and filled with lossless medium. Deviation from these conditions introduces a coupling between the cavity modes and changes their resonant frequencies in a manner for which no detailed theory is available.

Assuming the case of small losses, we used Eqs. (1a) and (1b) to determine the real parts of the permittivity tensor elements, inserting successively an appropriate pair of measured resonant frequencies of the identified modes, carefully corrected for detuning effects caused by the finite conductivity of the walls and the experimentally determined susceptances of the coupling apertures.

Dielectric losses for each mode were calculated from the measured half-power linewidth $2\Delta F$ and the relation

$$\tan\delta = \frac{1}{Q_{eff}} - \frac{1}{Q_{walls}} - \frac{2}{Q_{coup}}, \qquad (2)$$

where $Q_{eff} = F_{res}/2\Delta F$. Calculation of wall losses was done using the conductivity of gold $\sigma = 4.17 \times 10^7$ $[1/\Omega m]$ multiplied by a coefficient associated with the deposition technology, which from our experience with similar gold films on GaAs was taken to be equal to 0.25. The influence of couplings on the effective Q-factor was experimentally determined by measuring the Q's of cavities with subsequently increased surface of the coupling holes.

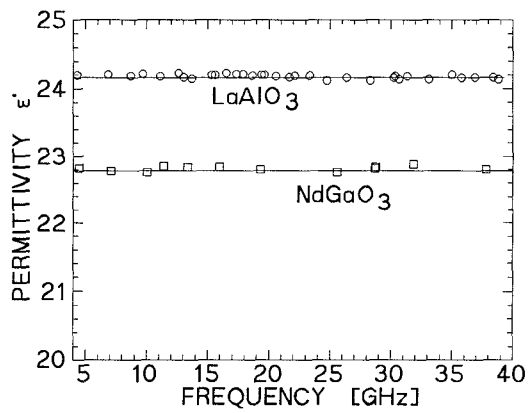

FIG. 1. Real part of the permittivities of $LaAlO_3$ and $NdGaO_4$ versus frequency at 298 K.

In Fig.1 we present dielectric permittivities ε' of $LaAlO_3$ and $NdGaO_3$ versus frequency, demonstrating the accuracy of our measurements. Up to the highest frequencies at which measurements could be performed, the permittivities were found to be constant within the experimental error.

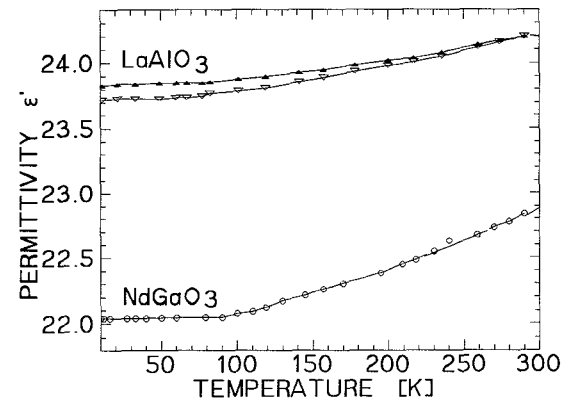

FIG. 2. Temperature variation of the permittivities of $LaAlO_3$ and $NdGaO_3$. Upper curve for $LaAlO_3$ was corrected for thermal contraction of the sample (thermal expansion coeff. $\alpha = 10^-5$). The α for $NdGaO_3$ was not known to us.

Fig.2 shows the variation of ε' with temperature for these materials. Upper curve for $LaAlO_3$, marked with full triangles, has been corrected for the thermal contraction of the resonator. The measurements were performed around X-band frequencies but no appreciable deviations from the $\varepsilon'(T)$ curves were found at higher frequencies.

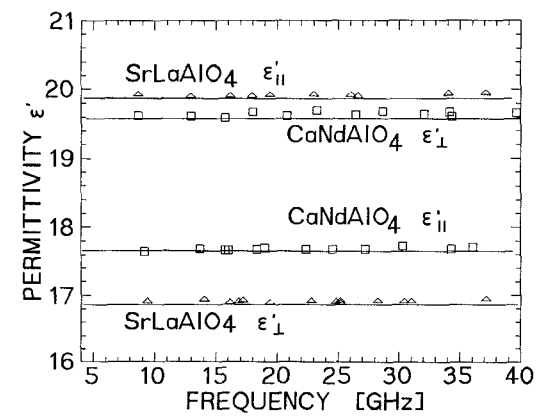

FIG. 3. Dielectric permittivities of $CaNdAlO_4$ and $SrLaAlO_4$ versus frequency. Note much higher anisotropy of the $SrLaAlO_4$ and the reversal of magnitudes of ε'_c and ε' in both materials.

In Fig.3 the dielectric permittivities of ε'_c and ε' (ε'_{ab}) for $CdNdAlO_4$ and $SrLaAlO_4$ versus frequency are presented. Here also ε'_c and ε' are within experimental errors constant over the microwave region from 8 to 40 GHz. Our results and in particular strong anisotropy of ε' are first reported for these materials at microwave frequencies. It is interesting to note that in $SrLaAlO_4$ $\varepsilon'_c > \varepsilon'$, while in $CaNdAlO_4$ $\varepsilon' > \varepsilon'_c$.

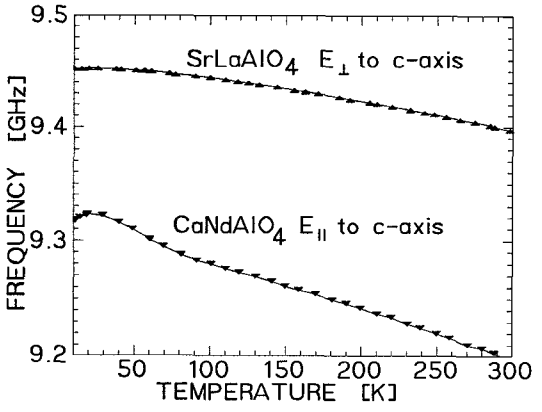

FIG. 4. Resonant frequency of the fundamental mode of the cavity filled with $SrLaAlO_4$ (geometry B) and with $CaNdAlO_4$ (geometry A)

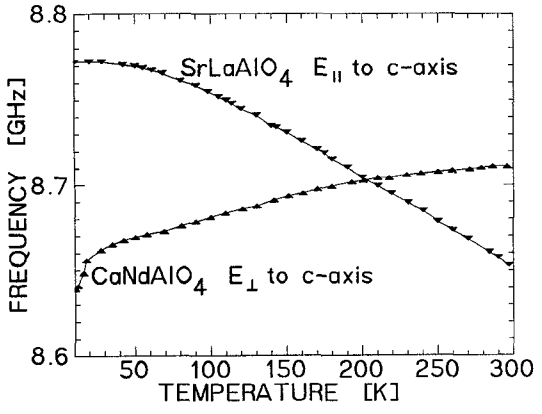

FIG. 5. Resonant frequency of the fundamental mode of the cavity filled with $SrLaAlO_4$ (geometry A) and with $CaNdAlO_4$ (geometry B). Note opposite slope of the latter curve.

Fig.4 and Fig.5 show and compare the temperature dependence of the resonant frequencies of the fundamental mode of $CaNdAlO_4$ and $SrLaAlO_4$ for samples A (TE_{110} with respect to c-axis) and B (TM_{011} also with respect of c-axis). For these modes the electric field is directed respectively along and perpendicular to c-axis of the crystal. Surprisingly the frequency shift for the principal mode of $CaNdAlO_4$ with E fields perpendicular to the c-axis has the opposite slope as compared to $CaNdAlO_4$ with E parallel to c-axis, as well as compared to both orientations of E in $SrLaAlO_4$. This behaviour was verified on all $CaNdAlO_4$ samples for all resonant modes with E fields perpendicular to the optical (c) axis. The $\varepsilon'(T)$ dependencies shown in Fig.4 and in Fig.5, when corrected for all experimental errors (forgetting for a moment about the irregularities of the experimental curves of $CaNdAlO_4$ below 50 K), can be explained by a small decrease of the dielectric constant. Indeed, a similar behaviour was found in $LaAlO_3$, $NdGaO_3$ and $SrLaGaO_7$.

However, no reasonable mechanism related to electric polarization, which could cause the decrease of the resonant frequency (i.e. an increase of ε') of the $CaNdAlO_4$ resonator with falling temperature shown in Fig.5 can be suggested. Thermal contraction of the cavity as a possible cause of such an effect can be easily ruled out. The thermal expansion coefficients for both materials has been recently reported by P.Byszewski et.al[4]. These coefficients show an appreciable anisotropy but they are all positive and comparable, so that they influence the resonance frequencies in a similar way.

One explanation which was considered, is a small increase of the permeability μ_c which can be due to the orientation of the spins of the Neodymium Nd^{3+} ions. The spin orientation in insulating crystals containing rare earth ions (with partially filled f-shells) grows as 1/T and even within the model of noninteracting Nd ions can largely account for the observed effect. Another explanation which also was in view, is a crystallographic phase transition in $CaNdAlO_4$ below 100 K. The physics of these effects is, however, beyond the scope of this paper and will be published elsewhere [5].

We would like to emphasize, that the described above behaviour of the dielectric permittivities of $CaNdAlO_4$ will cause different frequency shift with temperature in resonators designed respectively in coplanar and microstrip geometries. In particular the increase of the permittivity with temperature fall, may be advantageous and should lead to partial compensation of the troublesome temperature frequency shift of the superconducting filters, which is due to the inherent and inevitable temperature change of the kinetic inductance of the superconducting strips.

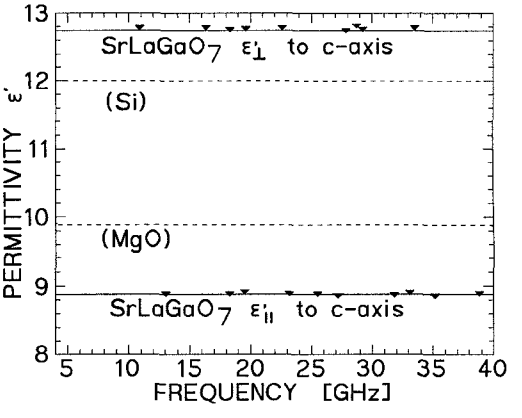

FIG. 6. Frequency dependence of the permittivities of $SrLaGaO_7$ (geometry A and B). Permittivities of Si and MgO, marked with dotted lines, are shown for comparison.

The dielectric permittivities of $SrLaGaO_7$ (geometries A and B) are shown in Fig.6. This material exhibits the highest anisotropy, offering at the same time for the designers of superconducting microwave components the lowest dielectric permittivity (for E field parallel to c-axis) even as compared to MgO. The temperature behaviour of the dielectric constant is relatively similar to that of $SrLaAlO_4$.

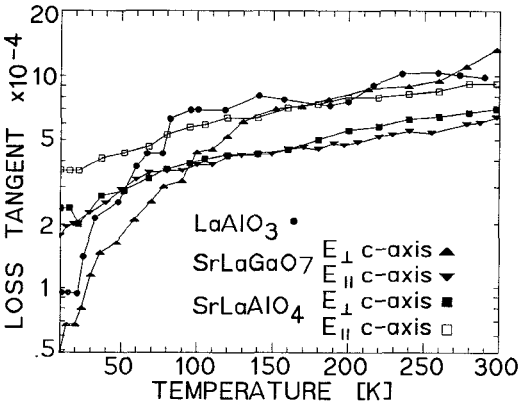

FIG. 7. Loss tangent ($\varepsilon''/\varepsilon'$) of $LaAlO_3$, $SrLaAlO_4$ and $SrLaGaO_7$.

Dielectric losses versus temperature for $LaAlO_3$, $SrLaALO_4$ and $SrLaGaO_7$ are jointly presented in Fig.7. All these materials exhibit quite similar $\varepsilon''(T)$ dependence, however the lowest losses have been measured for ε_c'' in $SrLaGaO_7$.

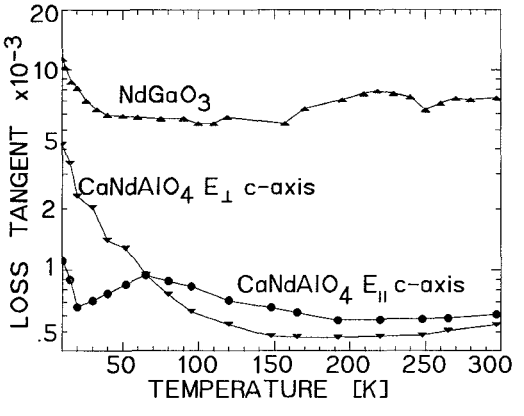

FIG. 8. Loss tangent of $CaNdAlO_4$ and $NdGaO_3$. Note the increase of dielectric losses at low temperatures and the local maximum of losses for E field parallel to c-axis in $CaNdAlO_4$.

Fig.8 shows the loss tangent versus temperature for $CaNdAlO_4$ and $NdGaO_3$. These materials were placed together, because they exhibit a peculiar behaviour of the losses. In both cases the loss tangent increases at low temperatures. The increase of microwave losses in $NdGaO_3$ below 170 K at 6.5 GHz was recently observed by Konaka et al.[1] and also at 16 GHz by N.Klein[6]. Both materials contain Nd^3+ ions in $4f^3$ configuration. Each ion has the magnetic moment equal 3.62 Bohr magnetons. We suggest that the collective interaction of these magnetic moments with microwave fields can be responsible for the increase of losses at low temperatures [5].

IV. CONCLUSIONS

In conclusion, dielectric properties of $CaNdAlO_4$, $LaAlO_3$, $NdGaO_3$, $SrLaAlO_4$, and $SrLaGaO_7$ monocrystals have been measured at microwave frequencies from 4 to 40 GHz and at temperatures from 10 to 300 K with the accuracy sufficient for precise designing of microwave superconducting components. Most materials were found to be uniaxially anisotropic, exhibiting at 300 K permittivities ε_c' ranging from 8.88 for $SrLaGaO_7$ (for E field parallel to the optical c-axis) to 24.18 for $LaAlO_3$. In all materials ε' decreases with temperature with the exception of $CaNdAlO_4$ where the ε_c' (along optical c-axis) increases with decreasing temperature. In $NdGaO_3$ and in $CaNdAlO_4$ microwave losses unexpectedly increase below 100 K. It is suggested that the magnetic interactions within the sublattice of neodymium ions with microwave fields can be responsible for the observed effects.

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