OBSERVATION OF AN ANOMALOUS TUNING RANGE OF A DOPED BST FERROELECTRIC MATERIAL DEVELOPED FOR ACCELERATOR APPLICATIONS*

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Abstract

The BST based ferroelectric-oxide compounds have been found as suitable materials for a fast electrically-controlled RF switches and phase shifters that are under development for accelerator applications in X, Ka and L-frequency bands. The BST(M) material (BST ferroelectric with Mg-based additives) allows fast switching and tuning in vacuum and in air both; switching time of material samples < 10 ns has been demonstrated [1-2]. One of the problems related to accelerator application of BST ferroelectric is its high dielectric constant. Decreasing the permittivity however is usually strongly correlated with a decrease in the tunability (k(E)=ε(0)/ε(E)) of ferroelectrics. The use of linear dielectric inclusions in BST ceramics could result in significant suppression of the mentioned k(E) dependence, with the best case being that the tunability vs. ε decrease could be unchanged. On the basis of our measurements we report here two unusual phenomena observed [3]: (i) the increase both the dc and the dynamic tunability with a decrease of the dielectric constant; (ii) the dynamic tunability was observed to exceed the static tunability at specific magnitudes of the applied field.

INTRODUCTION

With this paper, we consider to demonstrate our BST (BaTiO$_3$ - SrTiO$_3$ solid solution) (BST) composite material operating in the L-band frequency range as a ferroelectric active phase shifting and control elements for accelerator applications. Relatively low loss factor, high dielectric breakdown strength and easy mechanical treatment make ferroelectrics promising active elements in tuning and switching accelerator devices. In many aspects semiconductor, ferrite and plasma based devices cannot compete with ferroelectric loaded systems. Ferrite compounds are lossy and require high current pulsed solenoids in contrast to ferroelectric based tuning devices.

Euclid developed in 2005-2007 the BST ferroelectric with Mg-based additives that allows fast switching and tuning in vacuum at a high biasing voltage of 50 kV/cm [1-4]. This material was developed for the X-band frequency range (11.424 GHz) and demonstrated loss tangents as low as tan δ = 5×10-3 at 10 GHz. Tunability and loss factor measurements for large bulk ferroelectric samples have been done by Omega-P, Inc./Yale University [5-7]. With the current project, the goal was to develop a new BST-based material with a tunability of 6-8% at 15 kV/cm biasing field to be applied in air and to demonstrate this BST composition material operating in the L-band in ferroelectric active phase shifting and control elements of accelerator devices. Development of this type of material is a challenge; there are no other materials available with a tuning range and loss factor close to those listed above. The previously developed compositions cannot work in air in this biasing field ranges because it exhibits a tunability of only 2% at 15 kV/cm. The tuning behavior of the dielectric constant is extremely nonlinear vs. bias voltage in this field magnitude range. At the same time, optimized Ba/Sr ratios could in principle enhance the tunability of the entire composition but the loss factor increases as well prohibiting the use of this ferroelectric at high frequencies due to the high power absorption. Consequently, the new tunable material has to be developed to be able to work in air and at a relatively low biasing voltage of 15 kV/cm.

Figure 1: Comparative data on the electrical parameters: the dielectric constant ε, its maximum operating temperature ($T_m$), tunability (n) and loss tangent at 10 GHz for the BST mixture vs. the additive percentage of two types of Mg-based additives [3,4].

The overall goal of the current program was to design an L-band externally-controlled fast ferroelectric tuner for controlling the coupling of superconducting RF cavities for design and development of a fast electrically-controlled 704 MHz, 50 kW tuner based on a ferroelectric...
We also considered the 1.3GHz frequency range for the ILC and Project X use. The tuner prototype has been built and experimentally bench tested by Omega-P, Inc. using ferroelectric bars developed by Euclid Techlabs; a time response of <30 ns has been demonstrated [7].

**BST FERROELECTRICS AS TUNING ELEMENTS FOR L-BAND SC CAVITIES**

We used a heterogeneous mixture of ferroelectric and magnesia containing additives. The ferroelectric is a (Ba,Sr)TiO$_3$, or (BST) solid solution with the optimum ratio of barium and strontium. The barium/strontium correlation in the compound with a perovskite structure determines the Curie temperature of the ferroelectric. The value of the Curie temperature shifts towards lower temperatures with the increase of strontium concentration in the solid solution. Meanwhile, the increase of the strontium titanate content leads to the sharp decrease of both the dielectric constant of the BST solid solution (~several thousand close to the ferroelectric phase transition) and the dc electric field tunability. It is well known that the BST based compositions with high electric field tunabilities display very high dielectric losses, which seriously limit their practical applications.

The required level of the dielectric constant in the system of BST solid solutions with perovskite structure (Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ (x = 0.55) can be achieved by increasing the content of the bulk concentration of non-ferroelectric additive with low $\varepsilon$ in heterogeneous mixture of 30% and above. It is possible to use BST-MgO/MgTiO$_3$ composites to produce ferroelectrics with different permittivities, including $\varepsilon$ in the range of 200-300 and lower. However, in this case the tunability of $\varepsilon$ by DC field is sharply reduced and becomes negligibly small.

In our investigations, a new additive was found recently[3,4]. This compound also belongs to the group of linear dielectrics with small $\varepsilon$ and very small dielectric losses in the microwave range. The influence of this additive on the dielectric properties of a composite based on BTO/STO differs essentially from the influence of known earlier additives. On the one hand, the increase of the its concentration in the BST composition leads to the decrease of the dielectric constant of the ferroelectric composite, analogous to the influence of traditional magnesium-containing additives on the value of $\varepsilon$. However, an increase in the content of this linear dielectric in the volume of the composite results in the increase of the tunability coefficient $K_{dc}$, which is quite unusual. $K_{dc}$ increases by almost a factor of two with the increase of the additive to 60 wt.%., Fig.1.

It is especially important that such compositions demonstrate high tunability when applying short pulses (dynamic tunability $K_{dyn}$) at relatively low DC field magnitudes $\sim$15 kV/cm. Our measurements showed that in this case $K_{dyn}$ may even exceed $K_{dc}$ [3]. These differences, as the measurement data showed, are especially significant for samples of BST compositions with increased concentration of the additive (more than 40 mass%) operating in the high biasing field range exceeding 30 kV/cm. The nature of this anomalous behavior of the dielectric constant is not completely understood yet.

Figure 2: Static (dc) and dynamic tunability as a function of permittivity for ceramics with differing Mg-based additives content at $E_{dc}=E_{pulse}=4 \text{ V/}\mu\text{m}$. The lower curve shows the losses of the varactor as a function of permittivity at ~10 GHz and $E_{dc}=0$ [3].

Ferroelectric capacitors were fabricated as (1×1×0.5) mm$^3$ ceramic with (1×1) mm$^2$ contact gold metallization deposited on the sides. A specially developed microwave measurement technique was used to obtain static and dynamic CV characteristics. Dynamical CV characteristics were obtained for unipolar 1 μsec long voltage pulses with leading front duration of 5 ns. Each value of capacitance on the CV curve corresponds to a voltage pulse amplitude, which could be varied in the range U$_p$=(0.0-2.5) kV. The procedure allows direct comparison between dc and pulse CV characteristics with a relative error that does not exceed 0.1% of the capacitance scale [9].

Comparison of the dynamic (pulse) and static (dc) tunability is illustrated in Fig. 2. One can see that the nonlinear response to both the dc and pulse voltage application increases with the decrease of the ceramic permittivity. Furthermore for ceramic with $\varepsilon$=260 at elevated values of E-field strength (E ≥ 3 V/μm) the dynamic tunability exceeds the static tunability. Note that in spite of the small microwave losses in Mg-based additives in comparison with BST, the losses of the BST ceramic increase with the increase of the volume content of the linear additive, Fig.2.

In this section we present our progress on the development of the measurement setup to characterize the brazed ferroelectric material at 600-1300 MHz (i.e. measurements of the dielectric constant and loss tangent of the brazed ferroelectric ceramic bars). Bar sizes are cross section 6×4 mm$^2$ and length 66.5 mm. The width of the bars were metalized (Cr/Cu/Au) by magnetron sputtering methods (Cr/Cu layers) and the
required gold layer thickness was produced using the galvanic deposition method (Au layer). Thicknesses of the Cr/Cu/Au layers were 0.050/1.2/2 μm respectively.

Figure 3: Photo of the high Q resonant cavity and brazed ferroelectric ceramic bar with two side gold metallization.

Table 1: Dielectric Constant, Effective Quality Factor $Q_{eff}$, Effective Loss Tangent $(1/Q_{eff})$ and Figure of Merit $Q_{eff} \times f$ versus Mode Frequency for the Brazed Ferroelectric Bar

<table>
<thead>
<tr>
<th>Frequency, GHz</th>
<th>$\varepsilon$</th>
<th>$\tan\delta_{eff}$</th>
<th>$Q_{eff}$ factor</th>
<th>$Q_{eff} \times f$, GHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.401</td>
<td>258</td>
<td>0.0011</td>
<td>909</td>
<td>364</td>
</tr>
<tr>
<td>0.509</td>
<td>260</td>
<td>0.0012</td>
<td>833</td>
<td>423</td>
</tr>
<tr>
<td>0.643</td>
<td>260</td>
<td>0.0013</td>
<td>769</td>
<td>494</td>
</tr>
<tr>
<td>0.782</td>
<td>261</td>
<td>0.0014</td>
<td>714</td>
<td>558</td>
</tr>
<tr>
<td>0.919</td>
<td>263</td>
<td>0.0015</td>
<td>666</td>
<td>611</td>
</tr>
<tr>
<td>1.047</td>
<td>265</td>
<td>0.0016</td>
<td>625</td>
<td>654</td>
</tr>
<tr>
<td>1.184</td>
<td>265</td>
<td>0.0017</td>
<td>588</td>
<td>696</td>
</tr>
<tr>
<td>1.315</td>
<td>265</td>
<td>0.0018</td>
<td>556</td>
<td>731</td>
</tr>
<tr>
<td>1.445</td>
<td>265</td>
<td>0.0019</td>
<td>526</td>
<td>760</td>
</tr>
<tr>
<td>1.574</td>
<td>266</td>
<td>0.0020</td>
<td>500</td>
<td>787</td>
</tr>
</tbody>
</table>

The special resonant test fixture has been modeled, designed and manufactured for $\varepsilon$ and $\tan\delta$ measurements in the frequency range of 400 – 1600 MHz, Fig.3. Photos of the high Q resonant cavity (with no cover) and brazed ferroelectric ceramic bar with two side gold metallization are shown in Fig.3. The inner surface finish of the cavity was improved with the gold deposited by the galvanic deposition method. Table 1 shows the dielectric permittivity, effective quality factor $Q_{eff}$ consistent with dielectric losses and metal losses on the surface gold metallization and the effective loss tangent $(1/Q_{eff})$ versus mode frequency for the brazed ferroelectric bar. Measurements have been carried out in the 400-1600 MHz frequency range. The last column to the right presents the effective figure of merit $Q_{eff} \times f$ of the brazed ferroelectric bars. The permittivity of our typical brazed ferroelectric ceramic bars is practically constant ($\varepsilon\approx$600) in the frequency range 400—1600 MHz. The effective loss tangent (including metal losses) of ferroelectric bar with gold metalized surface was found to increase linearly from 0.0011 ($Q=910$) up to 0.0019 ($Q=510$) with the frequency swept from 400 MHz to 1600 MHz.

SUMMARY

A BST(M) composition material operating in air in the L-band frequency range has been developed for use as a ferroelectric active phase shifting and control elements for accelerator devices. Development of production techniques for new materials with $\varepsilon\approx$ 150-300 continued with the expectation of enabling operations in air at 15 kV/cm bias field and of lowering the loss tangent to values of (5-6)×10$^{-4}$ at 300-1300 MHz. It was found that by increasing by a specific ratio the overall content of linear dielectric additives in the ferroelectric composite based on a solid solution with the ratio BaTiO$_3$/SrTiO$_3$ = 55/45, one can reduced the dielectric permittivity from 600 to 150 and below, respectively. A high Q test cavity intended for dielectric constant and loss tangent measurements of the brazed bulk ferroelectric elements have been fabricated. The test cavity operates in the 400-1600 MHz frequency range with intrinsic Q factor (if loaded with lossless ceramic bar $\varepsilon=260$) in the range of 1000-2000 depending on the mode frequency. The cavity has been used for the characterization of the ferroelectric bars with dielectric constant in the range of ~ 300 and gold metallization deposited on the ferroelectric surfaces.

REFERENCES