Epitaxial Ferroelectric $Ba_{0.5}Sr_{0.5}TiO_3$ Thin Films for Room-Temperature High-Frequency Tunable Element Applications

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Perovskite $Ba_{0.5}Sr_{0.5}TiO_3$ thin films have been synthesized on (001) $LaAlO_3$ substrates by pulsed laser ablation. Extensive X-ray diffraction, rocking curve, and pole-figure studies suggest that the films are *c*-axis oriented and exhibit good in-plane relationship of $<100>_{BSTO}//<100>_{LAO}$. Rutherford Backscattering Spectrometry studies indicate that the epitaxial films have excellent crystalline quality with an ion beam minimum yield χ_{min} of only 2.6 %. The dielectric property measurements by the interdigital technique at 1 MHz show room temperature values of the relative dielectric constant, ε_r, and loss tangent, tanδ, of 1430 and 0.007 with no bias, and 960 and 0.001 with 35 V bias, respectively. The obtained data suggest that the as-grown $Ba_{0.5}Sr_{0.5}TiO_3$ films can be used for development of room-temperature high-frequency tunable elements.

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Thin films of ferroelectric barium strontium titanates $(Ba_{1-x}Sr_x)TiO_3$ (BSTO) have recently become very attractive to the microelectronic industry as good candidates for the applications in high density dynamic random access memories (DRAM) and microwave devices. They exhibit not only high relative dielectric constant (ε_r) values but also tunable ε_r by applying an electric field.[1-7] In the past several years, high density capacity memories have been rapidly shrinking in dimensions and expanding in capacities.[8] Gbit DRAM memories require larger capacitor in a relatively smaller area and short voltage pulses that correspond to frequencies of the microwave. However, traditional SiO_2 based devices are causing many problems in these high-density device fabrications due to the relative low dielectric constant of the SiO_2 materials. BSTO thin films have being considered as the best candidate for this application because of the high ε_r. In addition, the tunability of BSTO thin films also offers unique opportunity for the development of various microwave devices, such as the micro-strip line phase shifters, tunable filters and high-Q resonators.[2,6,9] Thus, the fabrication of high quality BSTO films exhibiting high ε_r and low loss tangent ($\tan\delta$) values have become a very important technological issue for pushing these materials further for practical applications.

Recently, many $Ba_{1-x}Sr_xTO_3$ films have been fabricated with different x values, or different ratios between $BaTiO_3$ and $SrTiO_3$, by various thin film techniques such as sputtering,[3,7] metal-organic solution deposition,[10] laser ablation,[2,11] and sol-gel processing. The ferroelectric properties of BSTO thin films are known to dramatically dependent on the composition x, or ratios between $SrTiO_3$ and $BaTiO_3$ components[12]. The Curie temperature and the room temperature dielectric constant of bulk BSTO vary from 400 to 30 K and few thousands to 300, respectively, for x values between 0.0 to 1.0. To date, no experimental data has been reported that for the room temperature BSTO films the ε_r can reach as high as 1200 and the $\tan\delta$

is as low as 10^{-3}. We have studied the BSTO thin films with x = 0.50 because its Curie temperature close to room temperature and relative large tunable dielectric constant. In this letter, we report the important achievements of the epitaxial growth of $Ba_{0.5}Sr_{0.5}TiO_3$ thin films on low dielectric (001) $LaAlO_3$ by pulsed laser ablation, exhibiting very high dielectric constant, large tunability, and very low loss tangent values.

A XeF excimer pulsed-laser of 248 nm was employed to the deposition of $Ba_{0.5}Sr_{0.5}TiO_3$ thin films on the (001) $LaAlO_3$ substrates. Energy density of about 6 J/cm^2 and the repetition rate of 10 Hz were adopted. The stoichiometric BSTO targets with their densities as high as 95 % were used. We selected single crystal (001) $LaAlO_3$ substrates because of its good lattice matching with the BSTO thin films (<1.3 %) and its low loss tangent ($\tan\delta < 10^{-5}$) values. The dielectric constant of $LaAlO_3$ is about 25. High quality epitaxial BSTO films can be only achieved in relatively narrow temperature range of 750 to 850 °C. Depositions beyond this temperature region have not resulted in epitaxial growth or react with the substrate. The oxygen pressure was an important factor to be considered for the film growth and ranged from 100 to 300 mTorr. The as-grown films were typically about 200 to 500 nm thick. The microstructure, crystallinity, and dielectric properties of the as-grown films were characterized by x-ray diffraction (XRD), Rutherford backscattering spectrometry and channeling (RBS-C), and the interdigital capacitor technique.

As described earlier, high quality BSTO thin films can only be synthesized on the (001) $LaAlO_3$ substrates under limited conditions. Figure 1 shows the typical θ-2θ XRD scans and the rocking curve measurements of the as-grown BSTO film of about 200 nm thick, grown at oxygen pressure 230 mTorr and 830 °C. Only the (00l) peaks appear in the θ-2θ scans, indicating that the as-grown film has its c-axis normal to the substrate surface. The rocking curve measurements of the (001) reflections reveal that the full width at half-maximum (FWHM) is less than 0.15°. These results suggest that the BSTO films are basically single-crystalline. The pole figure studies have been done to understand the quality of the epitaxy and to determine in-plane relationship between the BSTO films and the (001) $LaAlO_3$ substrates. The fourfold symmetry reflections from the {111} poles can be clearly seen in the Fig. 2. The very sharp spots with no satellites or broadening have been obtained, suggesting that the films are high degree in-plane orientation. The in-plane orientation relationship have been determined to be $<100>_{BSTO}$ // $<100>_{LAO}$. The lattice misfit of 1.3 %, determined from the in-plane relationship, may result in an increase of interfacial energy between the films and the substrates. However, the existence of plenty of twins in the substrate surfaces may release the strain energy so that the total energy of the system can be reduced during the film growth.[12]

The crystallinity of the BSTO epitaxial films were investigated by using 1.6 MeV He ion RBS- channeling technique. Figure 3 shows the random and aligned spectra for a film deposited with oxygen pressure of 250 mTorr at 820 °C. The RBS signals in the random spectrum reveal the clear steps corresponding to the Ba, Sr, Ti and La surfaces at channel numbers at 230, 219.5, 197, and 150. All of the RBS signals from the aligned spectrum appear as small peaks. To be seen clearly, the aligned spectrum was multiplied by a factor of 2. The minimum yield taken behind Sr surface peak is about 2.6%. This value is comparable with the normal minimum values for a single crystalline Si wafer and suggests the good crystallinity of the BSTO films deposited on (001) $LaAlO_3$ substrates. However, there is a scattering peak near the interface with the minimum yield of about 8.9% in the aligned spectrum. This slight larger value, compared with that of the film, in fact is partially due to the overlap with the La signals from the substrate. The existence of this slightly large scattering peak could result from the slight lattice distortion and/or defects such as dislocations around the interface regions. The defects at the interface may release the lattice mismatch energy, as discussed earlier. The TEM study for the interface morphology and detailed microstructures is currently under way.

The interdigital capacitor technique was applied to study the physical property of the as-grown films with the film thickness of 500 nm. The capacitor consists of 50 fingers of 7 mm long and 20 micron width, and spaced apart by 15 micron. The measurements were done with an HP4192A LF Impedance Analyzer. The ε_r was extracted from the capacitance using the conformal mapping results of Gevorgian, et al.[13] The relative dielectric constant (ε_r) and loss tangent (tanδ) taken as a function of temperature (300 to 80 K) at different values of dc bias (0 to 35 V) are shown in the Fig. 4. It appears from Fig. 4 (a) and (b) that the dielectric constant can be changed as much as 33% and the values of loss tangent can be reduced from 0.007 to 0.001 by applying at E field of 2.33 V/micron at room temperature. The data suggest that the quality of the epitaxial films allow development of high frequency tunable elements operating at room temperature. Higher frequency measurements of the films are being carried out in the NASA Lewis Research Center and will be reported elsewhere.

In summary, perovskite ferroelectric BSTO thin films have been epitaxially grown on the (001) $LaAlO_3$ by pulsed laser ablation. The epitaxial relationship is found to be (001) orientation normal to the substrate surface with in-plane relationships of $[100]_{BSTO}//[100]_{LAO}$. The excellent quality films are evident from relatively small FWHM values of 0.15°, small ion beam minimum yield of 2.6%, small loss tangent tanδ of less than 0.007, and relatively large dielectric constant ε_r of 1430. The dielectric constant can be changed as much as 33% at room temperature for the bias changing from 0 to 35 volts at 1 MHz suggests that the high frequency room temperature tunable elements can be developed using the epitaxial BSTO films.

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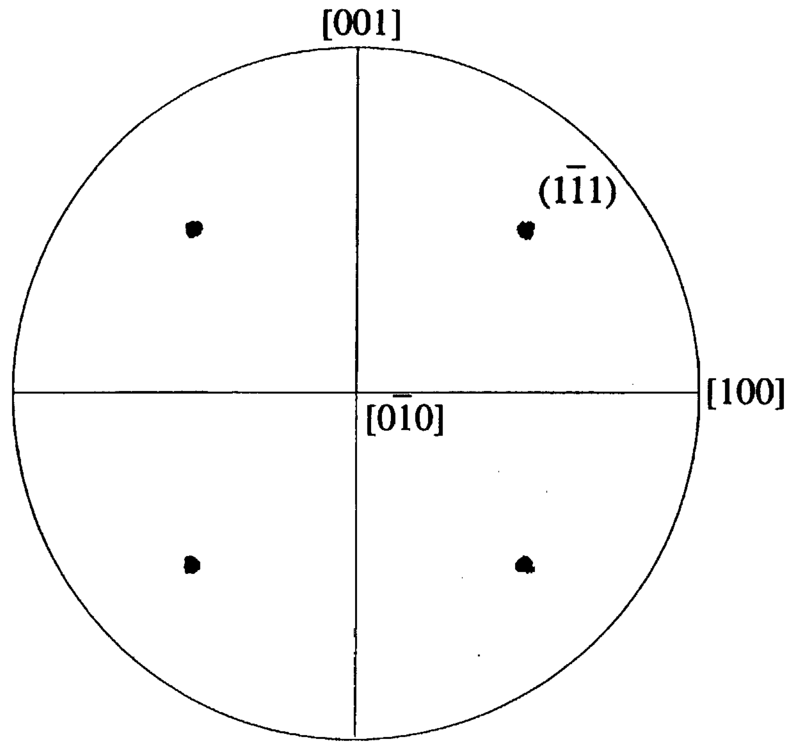
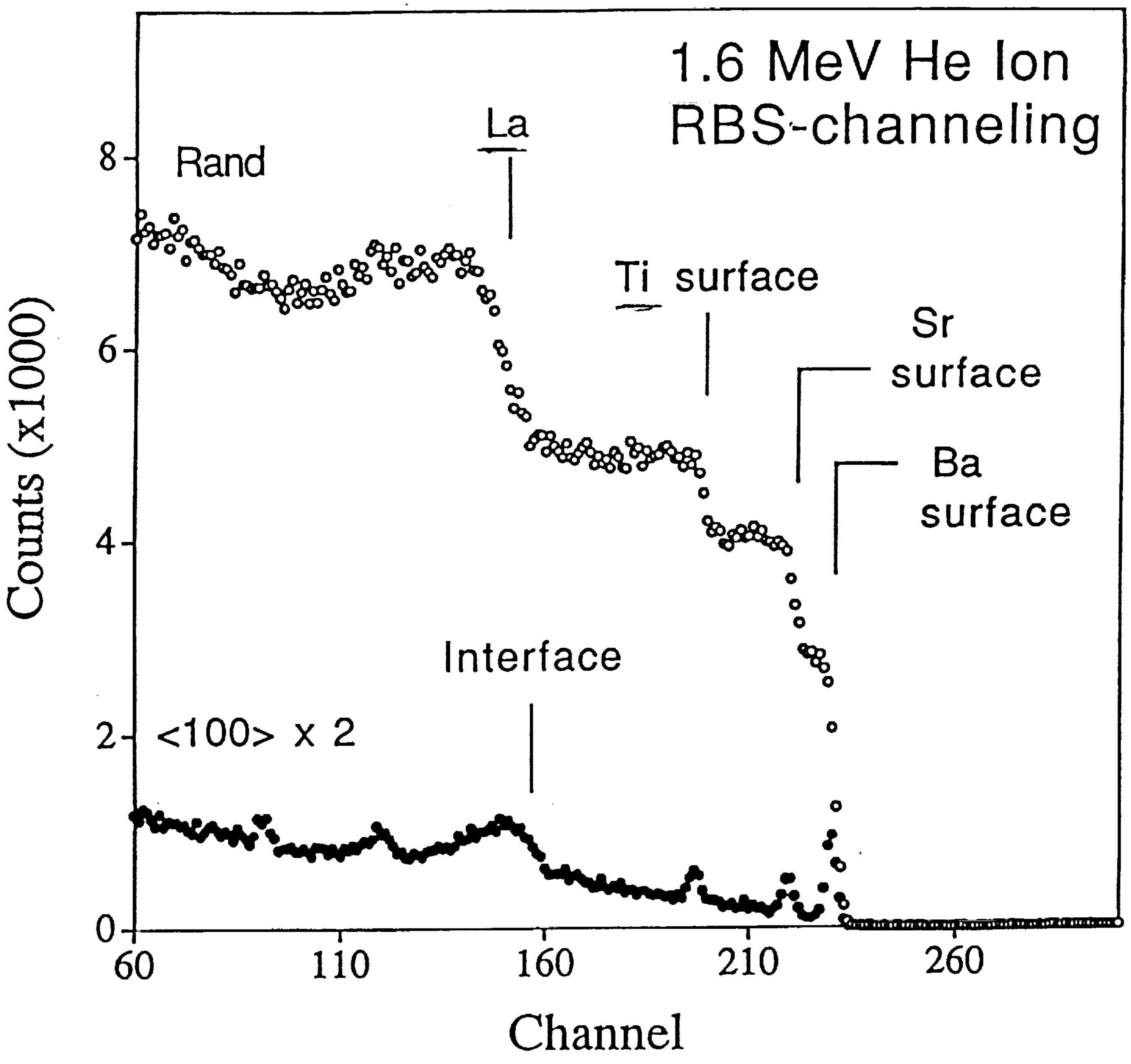


Fig. 2

$Ba_{0.5}Sr_{0.5}TiO_3/LaAlO_3$ (100)
A. Brazdeikis/C.L. Chen



○ Counts (x1000)

BST/LAO-001.dat

BST/LAO-001.gra

● <100> x 2

Fig. 3

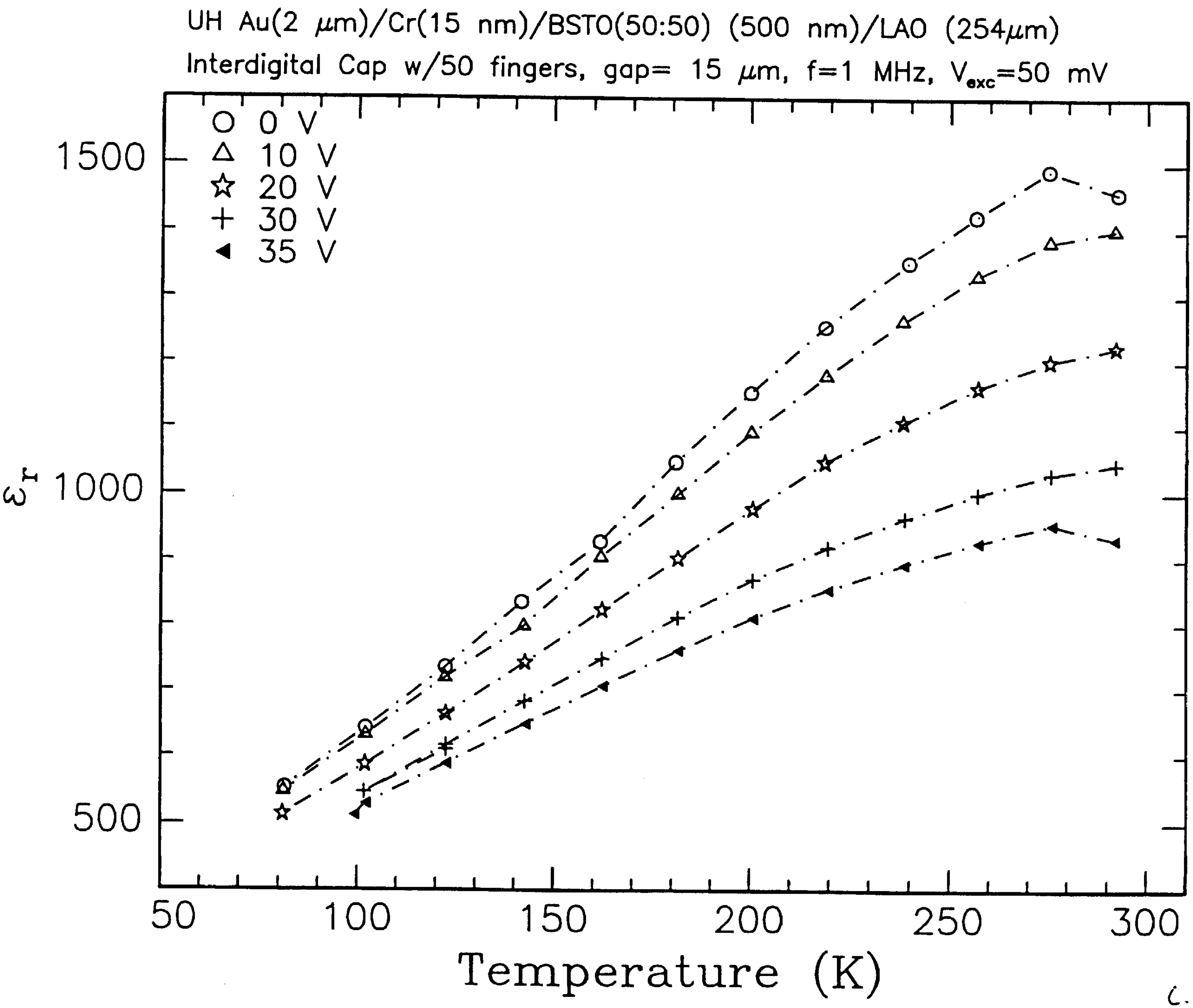


Fig 4a
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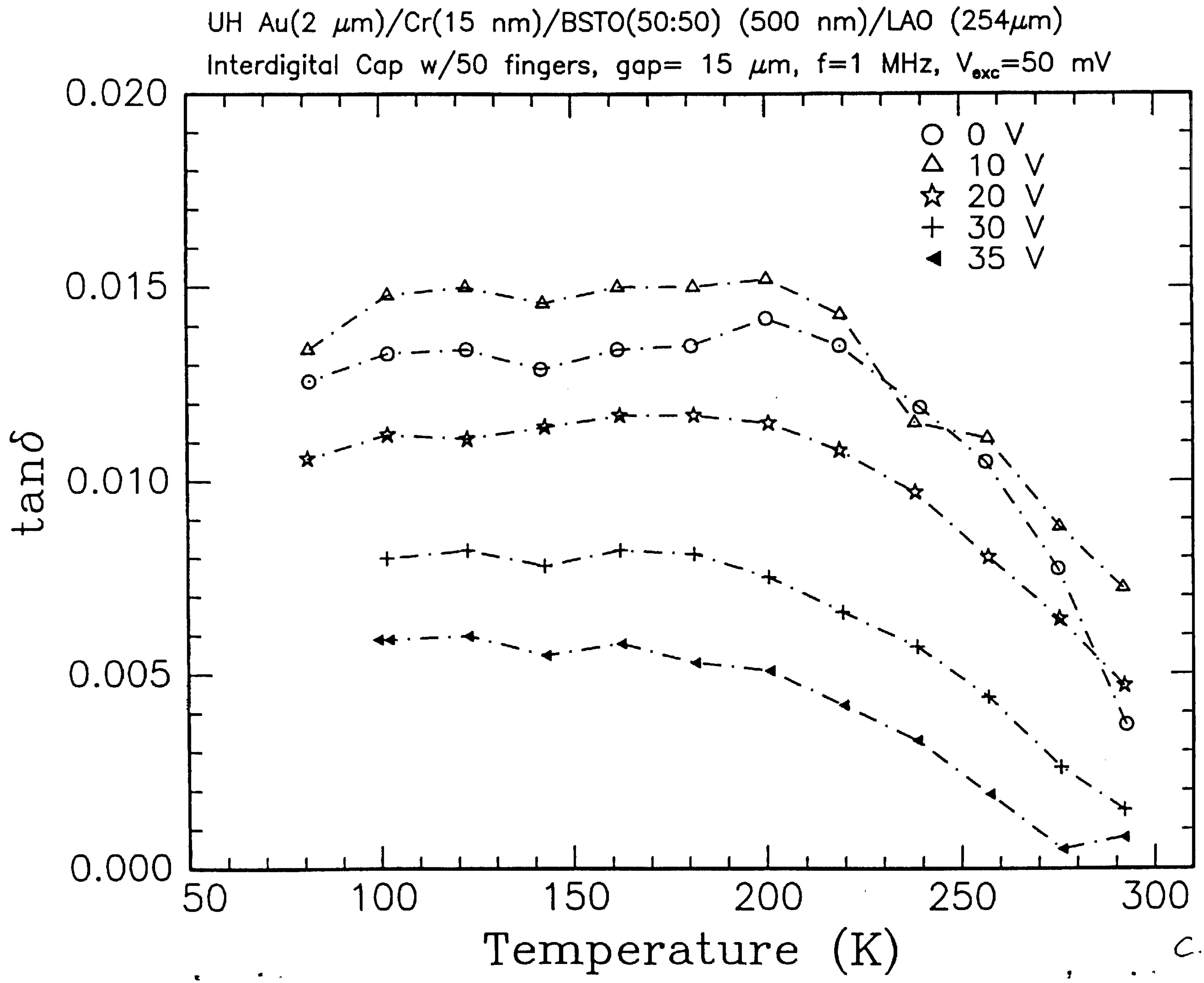

Fig 4b
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