Modulators of THz radiation based on $SrTiO_3$ epitaxial thin films

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***Abstract*—Transmittance of $SrTiO_3/DyScO_3$ thin film multilayers grown on $DyScO_3$ crystals was studied using THz time-domain spectroscopy. By applying an electrical bias of the order of tens of kV/cm, the transmitted power was shown to vary by tens of per cent in a broad frequency range.**

I. Introduction

The research in more efficient THz sources and sensors is accompanied by a growing interest in manipulation of propagating THz beams by means of active elements such as switches, modulators, tunable filters etc. Electrically commanded devices allowing a transfer of information from circuit devices to the THz spectral band are of particular interest [1].

Displacive ferroelectrics represent a class of materials suitable for the modulation of the complex permittivity by controlling the bias electric field or temperature. They exhibit a strong polar soft phonon responsible for the ferroelectric phase transition [2]. The soft mode determines in a decisive way the low-frequency dielectric permittivity, and its dynamics allows the dielectric permittivity to be tuned by means of varying temperature or bias electric field [2]. In particular, so called incipient ferroelectric materials such as strontium titanate ($SrTiO_3$, STO) display a great potential for microwave and THz tuning.

STO crystals remain paraelectric even at low temperatures; however, the ferroelectric phase can be induced and the transition temperature increased by introducing a tensile strain in an STO layer. The latter can be conveniently introduced by epitaxial growth owing to a lattice mismatch between STO and the substrate material [3], such as $DyScO_3$ (DSO). Owing to the tensile strain, there is a significant decrease of the room temperature soft mode frequency and an increase of the static permittivity while the transition temperature shifts close to the room temperature [2]. This modification of properties is advantageous for tuning of STO films and displays a potential for future applications. A further optimization of the properties of the structures depends on the knowledge and understanding of the tuning properties of various strained samples.

II. Experimental details

Four different samples were prepared by pulsed laser deposition onto (110) oriented single crystals of DSO: one $100\,\mu m$ STO film, and three multilayers consisting of $n \times (50\,\mu m\ \text{STO} + 50\,\mu m\ \text{DSO}) + 50\,\mu m$ STO films with $n = 1$, 2, 3. For the purpose of applying electric bias, a $6 \times 6\,mm^2$ interdigitated electrode structure consisting of $5\,\mu m$ wide gold lines and $15\,\mu m$ wide gaps was deposited on top [4]; both this structure and the DSO substrate were, in a broad band, transparent for the THz electric field $E_{THz} \parallel [101]$. The applied dc electric bias attained values of up to about $100\,kV/cm$.

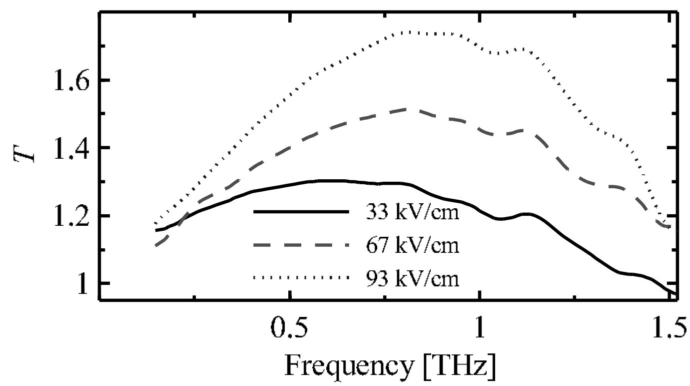

Fig. 1. Relative power transmission of the biased multilayer comprising four $50\,\mu m$ layers of STO separated by three DSO layers, as a function of applied bias, with respect to an unbiased sample.

Linearly polarized THz pulses were generated using femtosecond laser pulses photoexciting a semiconductor-based large-area interdigitated emitter [5]. The samples were placed in a cryostat, biased by voltages up to 120 V and cooled down to 120 K. The details about data acquisition and evaluation can be found in Ref. [4]. From the results, it appears that both the zero-bias spectra and their changes with applied voltage can be explained by a model where the soft mode is coupled to a silent central peak, and the only field-dependent parameter is the bare soft mode frequency; it follows a quadratic and linear regimes at low and high bias, respectively [6]. The temperature dependent measurements confirm that the best tunability occurs near room temperature. We were able to compare the performance of the individual multilayers. An example of the power transmittance depending on the applied electric field is shown in Fig. 1; near 1 THz, one can observe a power modulation of up to 40%.

III. Theoretical description and interpretation

Our data analysis shows that the dielectric behavior of the STO films in the THz range can be described by a general model assuming a highly anharmonic potential of the soft mode. At

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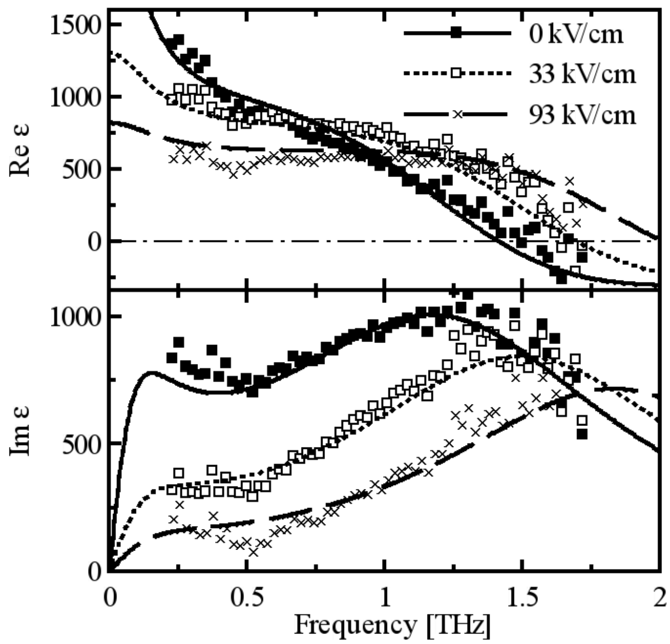

Fig. 2. Complex permittivity of the heterostructure composed of four STO and three DSO 50-nm thick layers depending upon the applied bias. Symbols: experimental results, lines: fits using a model of dielectric function according to Eq. (1).

the same time, it has been suggested recently that near the ferroelectric transition temperature, the displacive ferroelectrics exhibit a central mode in the 10–100 GHz range [7]. Therefore, the THz permittivity is written as:

$$\varepsilon(\omega) = S_i S_j G_{ij}(\omega) + \varepsilon_\infty \qquad (1)$$

where ε_∞ is a constant denoting the permittivity above the THz range; $\mathbf{G}$ is a Green function matrix in the form

$$\mathbf{G}^{-1}(\omega) = \begin{pmatrix} 1 - \mathrm{i}\omega/\gamma & \delta \\ \delta & \omega_0^2 - \omega^2 - \mathrm{i}\omega\Gamma \end{pmatrix} \qquad (2)$$

where ω_0 is the eigenfrequency of the polar soft mode at the given value of the external electric field, Γ denotes its bare damping and there is another low-frequency decay channel corresponding to a Debye relaxator with a bare frequency γ. The contribution of the relaxator is clearly visible in the spectra of unbiased samples (see Fig. 2) The vector $\mathbf{S}$ in Eq. (1) has the form $\mathbf{S} = (0, \sqrt{f})$; consequently, the relaxator only plays a role owing to the non-zero value of the coupling constant δ.

The model was successfully used to reproduce the data obtained experimentally. From our analysis it appears that the THz dielectric behavior is entirely controlled by the soft mode. The same values of parameters, $f = 2.3 \times 10^6\,\mathrm{cm}^{-2}$, $\gamma = 10\,\mathrm{cm}^{-1}$ and $\delta = 33\,\mathrm{cm}^{-1}$, can be used to describe correctly the dielectric spectra of all the samples at any value of the electrical bias (see Fig. 2); the values of the soft mode damping differ slightly among samples with values $\Gamma \approx 50\,\mathrm{cm}^{-1}$ which we attribute to an inhomogeneous broadening caused by a distribution of strain within the heterostructures.

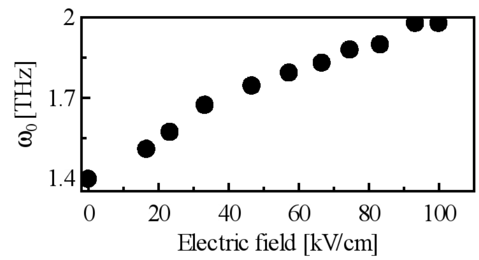

Fig. 3. Soft mode frequency ω_0 of the same sample as in Fig. 2, plotted as a function of applied electric field.

The parameter determining principally the shape of the spectra is the soft mode eigenfrequency ω_0 which can be influenced by the applied voltage. As an example, Fig. 3 shows the electric field dependence of the soft mode frequency obtained for the sample containing four 50 nm thick layers; a hardening by as much as 0.6 THz is observed for this sample. Note that the room-temperature eigenfrequency of the soft mode in unstrained STO is 2.7 THz. This pronounced shift of the soft mode is at the origin of the tunable transmittance depicted in Fig. 1.

IV. Conclusion

We have investigated the dielectric properties of different STO thin film heterostructures and shown their prominent tuning capabilities in the THz range. The tuning behavior of the samples can be explained by hardening of the soft mode which is coupled to a low-frequency relaxator. Further technological progress of the tunable structures based on strained STO films requires preparing multilayers with a higher number of bilayers without a decrease in the tensile strain within the ferroelectric films.

References

[1] H.-T. Chen, W. J. Padilla, J. M. O. Zide, A. C. Gossard, A. J. Taylor, and R. D. Averitt, "Active terahertz metamaterial devices," *Nature*, vol. 444, p. 597, 2006.

[2] P. Kužel and F. Kadlec, "Tunable structures and modulators for the THz light," *Comptes Rendus de l'Académie des Sciences – Physique*, vol. 9, pp. 197–214, 2008.

[3] J. H. Haeni, P. Irvin, W. Chang, R. Uecker, P. Reiche, Y. L. Li, S. Choudhury, W. Tian, M. E. Hawley, B. Craigo, A. K. Tagantsev, X. Q. Pan, S. K. Streiffer, L. Q. Chen, S. W. Kirchoefer, J. Levy, and D. G. Schlom, "Room-temperature ferroelectricity in strained $SrTiO_3$," *Nature*, vol. 430, p. 758, 2004.

[4] P. Kužel, F. Kadlec, J. Petzelt, J. Schubert, and G. Panaitov, "Highly tunable $SrTiO_3$/$DyScO_3$ heterostructures for applications in the terahertz range," *Appl. Phys. Lett.*, vol. 91, p. 232911, 2007.

[5] A. Dreyhaupt, S. Winnerl, T. Dekorsy, and M. Helm, "High-intensity terahertz radiation from a microstructured large-area photoconductor," *Appl. Phys. Lett.*, vol. 86, p. 121114, 2005.

[6] P. Kužel, C. Kadlec, F. Kadlec, J. Schubert, and G. Panaitov, "Field-induced soft mode hardening in $SrTiO_3$/$DyScO_3$ multilayers," *Appl. Phys. Lett.*, 2008, submitted.

[7] E. Buixaderas, S. Kamba, and J. Petzelt, "Lattice dynamics and central-mode phenomena in the dielectric response of ferroelectrics and related materials," *Ferroelectrics*, vol. 308, p. 131, 2004.