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We have developed an interdigited electrode structure for applying a static electric field to a ferroelectric thin film, which enables nearly full transmission of a perpendicularly polarized terahertz wave. This approach has been used to determine the electric field dependence of the complex permittivity of a $SrTiO_3$ thin film on a sapphire substrate up to about 2 THz employing time-domain terahertz spectroscopy. We have demonstrated up to 10% variation of the film permittivity at 300 GHz at room temperature induced by an applied electric field of 100 kV/cm. No dielectric dispersion is observed between 1 MHz and about 500 GHz. The field-induced changes are attributed to soft mode hardening. © *2006 American Institute of Physics*.
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The investigation of dielectric voltage-controlled thin film structures is important for the development of fast-acting frequency agile microwave and sub-THz integrated devices such as phase shifters, tunable filters, voltage tuned oscillators, and amplitude and phase modulators. Ferroelectric materials, which are characterized by a high permittivity up to the THz range[1] offer a possibility of its tuning by means of temperature or external dc (low-frequency) electric field.[2–5] Thin ferroelectric films are especially attractive for tunable applications owing to the low tuning bias, potentially low production cost, and high integrability. In particular, for millimeter wave frequencies semiconducting varactors become very lossy such that ferroelectric films represent a challenging alternative. A review on tunable ferroelectric materials for microwave devices has been published recently.[6]

Strontium titanate (STO) is an incipient ferroelectric material, which means that, on the one hand, its dielectric behavior is fully controlled by the soft mode dynamics,[7] and on the other hand, it remains paraelectric down to the lowest temperatures due to quantum fluctuations.[8] These properties along with the possibility of preparing high quality thin films make STO a material of choice for applications in tunable microwave components. An increased tunability in the microwave range at room temperature can be achieved by chemical substitution of strontium atoms by barium (barium–strontium–titanate solid solutions)[6] or by introducing a tensile strain by a proper choice of substrate for the epitaxial thin film growth.[9]

In this letter we demonstrate an electric-field induced variation of the permittivity of an STO thin film from the MHz up to the THz frequency range at room temperature.

The STO thin film sample was prepared by pulsed laser deposition on a CeO buffered r-cut sapphire substrate. Rutherford backscattering and x-ray analysis reveal a thickness of 313 nm (5% accuracy), a high degree (99%) of (100) orientation, and a lattice constant of 0.3911 nm deviating less than 0.1% from the bulk value. The gold electrodes were prepared by standard photolithography and ion beam etching of a 20 nm thick titanium/300 nm thick gold film deposited by dc magnetron sputtering on top of the STO film. The interdigited capacitor structure of 4×4 mm size was composed of 5 μm wide gold lines separated by 15 μm wide gaps (Fig. 1).

The tuning behavior at a frequency of 1 MHz was determined by measuring the dc bias voltage dependence of the capacitance of the interdigited structure by impedance spectroscopy. It was assumed that the observed relative changes of the capacitance with voltage are equal to the relative changes of the permittivity.

For the THz time-domain experiments we used a Ti:sapphire multipass amplifier (Quantronix, Odin) as a femtosecond laser source. The THz probing pulses were generated via optical rectification in a 1 mm thick [110] ZnTe single crystal. The THz pulses transmitted through the sample were detected using a usual electro-optic sampling scheme with another 1 mm thick [110] ZnTe crystal and a pair of balanced Si photodiodes.[10] The difference signal from photodiodes fed a lock-in amplifier which was synchronized to a suitably chosen modulation (see below). The THz time-

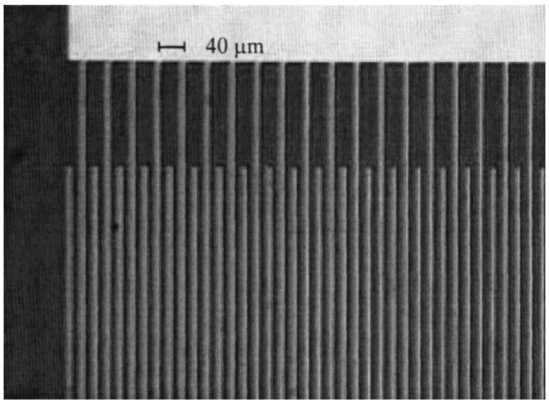

FIG. 1. Scanning electron microscope picture of the interdigited capacitor structure.

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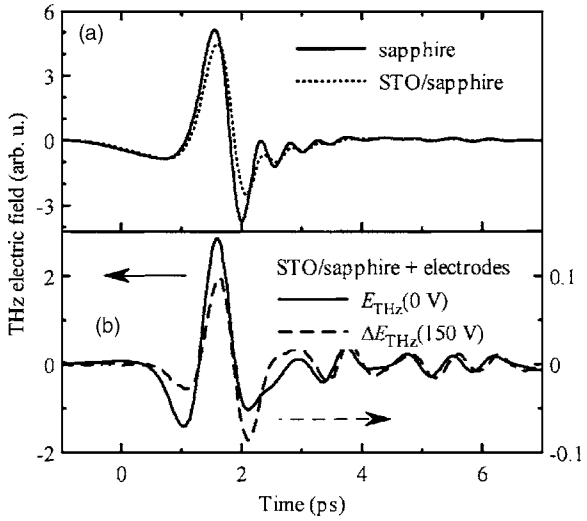

FIG. 2. THz wave forms: (a) sample without electrodes; the data serve for the evaluation of the thin film properties without electric field and (b) sample with electrodes; E_{THz} is a reference wave form (without field) and ΔE_{THz} is a difference signal waveform (field on minus field off).

domain technique probes the in-plane response of the sample, i.e., the possible passive layers between the STO layer and the substrate or the electrodes have almost a negligible effect.

In a first series of experiments we have determined the complex dielectric spectrum of the STO layer (without an interdigited electrode pattern). For this purpose the optical beam generating of the THz pulses was chopped by a mechanical chopper at 166 Hz. As the $(1\bar{1}02)$-oriented sapphire substrate is birefringent we used a pair of crossed wire-grid polarizers to carefully align the principal axes of the substrate with respect to the THz beam polarization. The experiment consisted of two consecutive measurements: (i) measurement of a signal wave form E_s with the thin film on a substrate in the path of the THz beam; and (ii) measurement of a reference wave form E_r with a bare substrate [see Fig. 2(a)]. The ratio of the Fourier transformations of the time-domain wave forms provides complex THz transmittance spectrum of the film: $t(\omega)=E_s(\omega)/E_r(\omega)$.

The technique allows us to determine the dielectric response $\varepsilon(\omega)=\varepsilon_1(\omega)+i\varepsilon_2(\omega)$ of the thin film in the range from 8 to 70 cm^{-1} by numerically inverting the expression for $t(\omega)$:[1]

$$t=\frac{2N(n_s+1)\exp[i\omega(N-1)d/c]}{(1+N)(N+n_s)+(1-N)(N-n_s)\exp(2i\omega Nd/c)}, \quad (1)$$

where $N=\sqrt{\varepsilon}$ is the complex refractive index of the STO film, n_s is the refractive index of the sapphire substrate, $d=313$ nm is the film thickness, and c is the speed of light in vacuum. The resulting dielectric spectrum is shown in Fig. 3. The error of the determination of the permittivity value is about 30%. Note that the low-frequency value of the permittivity (≈ 370) and the soft mode frequency (≈ 90 cm^{-1}) of the thin film are close to the values found in STO single crystals,[11] giving evidence of a good quality of our film. The damping of the soft mode is found to be rather strong (95–115 cm^{-1}); this is probably due to the inhomogeneous broadening which is frequently observed in thin films.[1,6]

In a second series of experiments we used the sample with the electrode pattern deposited onto the STO layer. An

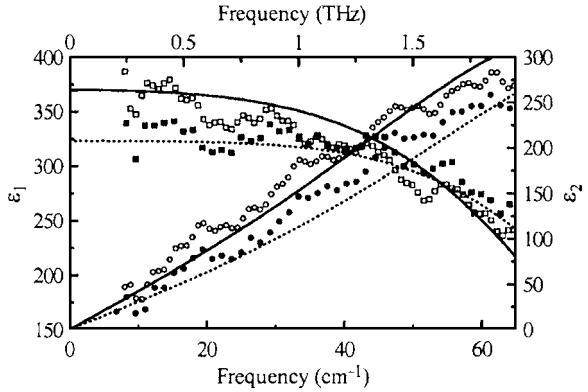

FIG. 3. Measured dielectric spectra of STO thin film in the THz range (symbols) and their fits by a single damped harmonic oscillator model (lines). ε_1: squares, ε_2: circles; open symbols and full lines: zero field, closed symbols and dotted lines: $V_{eff}=150$ V.

ac sinusoidal voltage $V(t)=\sqrt{2}V_{eff}\sin(2\pi ft)$ ($f=166$ Hz) was applied to the sample with effective voltage values up to $V_{eff}=150$ V. We assumed, and the experiments confirmed this hypothesis, that the dielectric modulation would show a quadratic behavior upon the applied field. The depth of the dielectric modulation then should be proportional to $\langle V^2(t)\rangle=V_{eff}^2$. In order to increase the sensitivity of the measurements, the mechanical chopper was removed from the optical beam path and the lock-in amplifier was locked to the frequency $2f$. This allowed us to measure directly the difference wave form $\Delta E_{THz}(V_{eff})=E_{THz}(V_{eff})-E_{THz}(0)$. Figure 2(b) shows a comparison of the reference zero-field wave form E_{THz} and the difference wave form ΔE_{THz} recorded for the highest voltage applied. Note that ΔE_{THz} has the same sign as E_{THz}, which means that the transmitted THz signal increases when the field is applied. Such a behavior can be accounted for by an improved impedance matching at the film surface (i.e., by a decrease of the real part of the permittivity) and/or by a decrease of the propagation losses of the film.

The modulation of the THz spectrum remains relatively small even for the highest applied field, i.e., $\Delta E_{THz} \ll E_{THz}$. The field-induced dielectric properties of the thin film then can be evaluated using the following formula:

$$\Delta N=\left(\frac{dt}{dN}\right)^{-1}\Delta t=\left(\frac{dt}{dN}\right)^{-1}\frac{\Delta E_{THz}(\omega)}{E_{THz}(\omega)}, \quad (2)$$

where the derivative dt/dN can be calculated using Eq. (1). The experimental protocol allows us to obtain rather precisely the field-induced changes of the real and imaginary part of the permittivity $\Delta\varepsilon=\Delta\varepsilon_1+i\Delta\varepsilon_2$: $\Delta\varepsilon=2N\Delta N$. The spectra of these quantities for several values of the voltage are shown in Fig. 4. As expected, both $\Delta\varepsilon_1$ and $\Delta\varepsilon_2$ are negative. The modulation of the permittivity reaches 11% for 150 V (i.e., for 100 kV/cm). This corresponds well to the predictions based on the Landau theory for STO yielding a tunability of 9% for the same electrical field applied.[6]

The results obtained in the THz range can be directly compared to those measured at 1 MHz (see Fig. 4). The values indicate that there is no additional dielectric contribution in between these spectral ranges. On the other hand, one observes a pronounced dispersion of the measured curves in the THz range where the soft mode contribution dominates the behavior of the permittivity and losses.

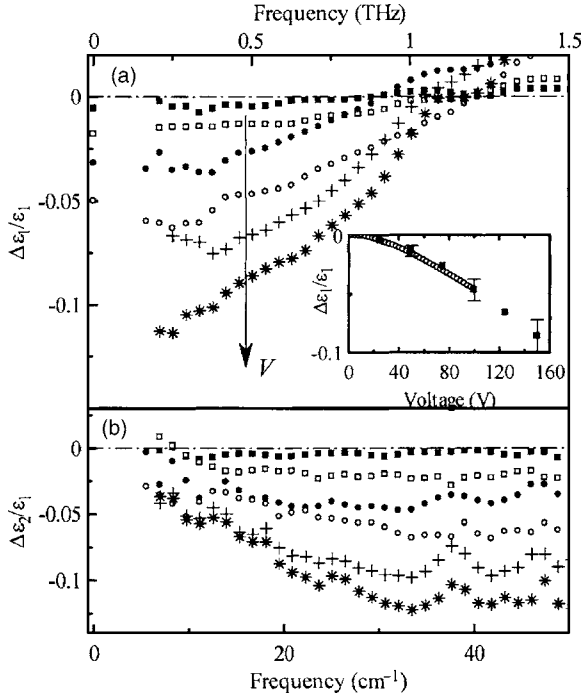

FIG. 4. Electric field induced modulation of the permittivity and losses. Applied voltages: 25, 50, 75, 100, 125, and 150 V. Values at 1 MHz were measured by impedance spectroscopy. (Inset) Comparison of the permittivity variation at 0.5 THz (closed symbols) and 1 MHz (open symbols).

Figure 3 shows a comparison of the zero-field dielectric behavior with that observed at 150 V. Note that, while the determination of the permittivity modulation $\Delta\varepsilon$ is quite accurate, the curves in Fig. 3 are rather noisy due to the experimental errors in the zero-field permittivity determination: $\varepsilon(150\ \mathrm{V})=\varepsilon(0\ \mathrm{V})+\Delta\varepsilon$. We fitted the spectra by a dielectric function of a single damped harmonic oscillator (soft mode):

$$\varepsilon = \varepsilon_\infty + \frac{B}{\omega_0^2(V) - \omega^2 - i\omega\gamma(V)}, \tag{3}$$

where ε_∞ is the high-frequency permittivity and B is the oscillator strength. Neither of these parameters is assumed to depend on the applied field. In contrast, both the soft mode frequency ω_0 and its damping γ may be tuned by the applied voltage. In fact, it is not possible to determine unambiguously all the parameters from the experimental data. On the other hand, we have performed a large number of simultaneous trial fits of $\varepsilon(0)$ and $\varepsilon(150)$ with some of the parameters (ε_∞, B, and/or γ) fixed to particular values. All these fits lead to a conclusion that the soft mode frequency hardens by 6 cm^{-1} when the field is applied. Similar frequency shift of the soft mode was observed in the temperature dependence upon heating by 60 K.[11] The soft mode is heavily damped ($\gamma \approx 95-115$ cm^{-1}) and the fit is slightly improved if the damping decreases with the field (by 2–5 cm^{-1}). Other parameters ($\omega_0 \approx 90$ cm^{-1}, $B \approx 2.5\times10^6$ cm^{-2}) are in reasonable agreement with their values reported for single crystals.[7,11]

To summarize, we have demonstrated the field-induced tunability of the dielectric properties of STO thin films in the MHz and THz frequency ranges. We have shown that the observed modulation is due to the soft mode hardening upon applied electric field. No additional contribution was found below the THz range. At 100 kV/cm the permittivity decreases by 11%. Using such electrically controllable thin films would considerably enhance the operation speed and performance of highly tunable photonic crystal filters as recently reported in Ref. 12.

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