Terahertz response of SrTiO$_3$ based heterostructures: influence of strain, temperature and electric field

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Abstract—A systematic study of strained SrTiO$_3$/DyScO$_3$ heterostructures was performed using time-domain terahertz spectroscopy in the temperature range from 20 to 294 K. Interdigitated electrodes deposited on top of the structures allowed probing of the terahertz response upon an applied electric field. We explored a possibility to increase the total thickness of SrTiO$_3$ layers while maintaining the epitaxial strain which induces the ferroelectric phase transition. All studied structures underwent the phase transition close to room temperature. Evolution of the complex dielectric spectra with temperature and electric bias was described by a general model involving a harmonic oscillator (soft mode) coupled to a Debye relaxation (central mode). At high temperatures, the soft mode is responsible for nearly all the changes observed in the spectra with temperature and applied electric field. At low temperatures, deep in the ferroelectric phase, the soft mode significantly hardens and loses its importance for the terahertz dynamics; the central mode becomes stronger and it almost completely determines the shape of the measured spectra. The observed variation of the phase transition temperature and of the terahertz response among investigated structures was ascribed to a partial strain relaxation.

I. INTRODUCTION

CONTINUOUS development of the microwave and terahertz (THz) technology cannot be maintained without new functional materials showing a large potential for their applications. Tunable properties of dielectric materials are highly valuable for the development of components enabling a control of the propagation of radiation. Ferroelectric materials are in principle well suited for these applications. They are characterized by a high permittivity value up to far infrared range due to the low-frequency phonon mode connected to the ferroelectric instability. This so-called soft mode gives dominant contribution to the sub-THz permittivity owing to its very low frequency and highly polar character. The latter provides a strong coupling with an external electric field allowing tuning of the material permittivity.

Strontium titanate (STO) is an incipient ferroelectric: on one hand, its sub-THz dielectric behavior is fully controlled by the ferroelectric soft mode; on the other hand, it remains paraelectric down to the lowest temperatures due to quantum fluctuations. Recently, strained epitaxial STO films have been shown$^1$ to undergo the ferroelectric phase transition near 300 K. However, the strain induced by the substrate, DyScO$_3$ (DSO) in our case, gradually relaxes with increasing thickness of the film. To overcome this problem we prepared several heterostructures consisting of n×STO/DSO bilayers (n = 2–8) on (110)-oriented DSO substrates by pulsed laser deposition. The thickness of each individual layer is of 50 nm, since the strain relaxation mainly occurs for thicker STO films.$^2$ The DSO layers in these structures are intended to maintain the
The soft-mode frequency is the key parameter which describes the temperature and electric-field evolution of the permittivity (see Fig. 2). The central mode contributes to the low-frequency part of the spectra at higher temperatures, while it completely governs the shape of the spectra at lowest temperatures.

We estimated the phase transition temperature for all structures from the temperature behavior of the soft-mode frequency (see Table 1). The 2×50 structure has the highest observed phase transition temperature. This can be explained by the fact that the strain distribution is more homogeneous in two separate 50-nm-thick layers of STO than in a 100-nm-thick layer of 1×100 sample. In thicker multilayer systems a gradual reduction of the strain towards the topmost layer occurs. Indeed, x-ray diffraction experiments confirm the above statement.

In our fits of equilibrium spectra (without applied electric field) we assumed that \( \Gamma, \delta, \gamma \) and \( f \) are temperature independent parameters. The values of these parameters do not differ much among samples. The relaxation strength \( g \) acquires nonzero values only at lower temperatures, when the STO thin film is in the ferroelectric phase according to the temperature behavior of the soft-mode frequency. \( g(T) \) grows with cooling. It is interesting to note that the highest values of the relaxation strength were observed in samples with a higher degree of an epitaxial strain relaxation.

The soft-mode frequency was the only electric-field dependent parameter. Its behavior with electric field was interpreted in terms of the Landau-Devonshire theory. We developed the model\(^4\) which includes possible presence of a nonvanishing macroscopic polarization in STO films due to the applied electric bias. The model implies that the soft-mode frequency changes not only due to the applied bias, but that it also can be renormalized by a spontaneous polarization value in the ferroelectric phase. We also approximately estimated the degree of the soft-mode potential anharmonicity value by fitting the electric-field behavior of the soft mode. An increase of the value of the spontaneous polarization was observed with cooling.

III. SUMMARY

We characterized the terahertz dielectric properties and their electric-field tunability of several STO/DSO thin-film heterostructures using the time-domain THz spectroscopy and x-ray diffraction. We estimated the paraelectric-to-ferroelectric phase transition temperatures for all the investigated structures. Evolution of dielectric spectra at different temperatures and applied electric fields was well described by a general model, which includes a damped harmonic oscillator (soft mode) coupled with a Debye relaxation (central mode). The soft-mode frequency was demonstrated to be the key parameter responsible for the behavior of the dielectric spectra.

We did not observe substantial decrease of the electric-field tunability with cooling. This phenomenon is explained by a high value of the spontaneous polarization in the thin films in the ferroelectric phase.

We explain the observed variation of the phase transition temperature and of the THz response by a partial epitaxial strain relaxation in the heterostructures. We correlate the spectroscopic results with the variation of lattice constants of the strained STO films obtained by x-ray diffraction and with effects coming from unintended deviations in the composition stoichiometry of the samples.

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REFERENCES


<table>
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<tr>
<th>Sample</th>
<th>( d_{\text{STO}} ) (nm)</th>
<th>( \omega_0(0) ) (cm(^{-1}))</th>
<th>( T_C ) (K)</th>
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<tr>
<td>1×100</td>
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<td>59.1</td>
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FIG. 2. Soft-mode frequency behavior with temperature and electric field for one of the studied structures. White lines on the surface are lines of equal values of the soft-mode frequency with a step between them of 2 cm\(^{-1}\).

TABLE 1. Selected characteristics of the thin-film structures. The soft-mode frequency \( \omega_0(0) \) is taken at the phase transition temperature \( T_C \) without applied electric field. \( d_{\text{STO}} \) is the total thickness of STO layer in each structure.