The correction of THz-time domain reflection spectroscopy of $SrTiO_3$

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Abstract: The experimental results of $SrTiO_3$ from 0.3THz to 6THz shows that an analytic continuation of the complex reflection can reduce the impact of limited spectra so that a more accurate correction can be acquired.
OCIS codes: (300.6495) Spectroscopy, terahertz; (120.5700) Reflection; (130.2260) Ferroelectrics

1. Introduction

The measurement of THz-time domain reflection spectroscopy (THz-TDRS) is especially useful when samples are opaque in the THz range. However, in the experiment of THz-TDRS, the front surfaces of the reference and the sample should be coincident spatially, typically with a precision of a few microns. It is nearly impossible for operation and the measured phase can be easily distorted by nonnegligible systematic phase error. Various experimental techniques have been suggested to solve this problem [1-3]. However, they still have difficulties and limitations. Therefore, some numerical methods have been proposed: the maximum entropy model (MEM) and Kramers-Kronig (K-K) relations [4-6]. The main problem for MEM is its complication and the concept not relying on physical laws but on information mathematics [4]. Compared to MEM, K-K relations don't have these problems while it is limited by the fact that the measurement can be only within finite spectra experimentally. In order to reduce the influence of limited spectra, modified K-K relations have been proposed. In 2005, Lucarini applied singly subtractive Kramers-Kronig (SSKK) relations to solve this problem, which is just for the positive error [5]. In 2006, based on SSKK dispersion relations, Gornov used a special function satisfied with the causality for the phase retrieval [6].

In this paper, a proper physical model has been considered to determine the permittivity more accurately. It is interesting that the analytic continuation of the complex reflection by the physical model can determine the corrected misplacement about 0.1μm.In addition, the method used here is much simpler than those based on K-K relations and MEM.

2. Experimental setup

Our experiment is carried out in a THz-TDRS system, the high-speed Asynchronous Optical Sampling (ASOPS)-based THz spectroscopy system (HASSP-THZ, Laser Quantum), where two femtosecond lasers are used instead of mechanical delay line. THz pulses are generated by irradiating a biased photoconductive antenna with a 50fs mode-locked Ti: sapphire laser. Then they are collimated and focused onto the sample by off-axis parabolic mirrors with S polarization at an incident angle of 30 degree. The reflected THz pulses are collected and detected using a GaP (110) crystal. The spectrum range of the system is 0.3THz-6THz.

A gold-coating mirror is used as a reference due to its uniform and high reflectivity in the THz range. Compared with THz-time domain transmission spectroscopy (THz-TDTS), the reflective geometry comes with a nonnegligible systematic phase error. The front surfaces of the reference and the sample are fixed to the holder with a hole of 10 mm diameter by their gravity, respectively. This setup made it easy to switch them during the measurement with misplacement less than 3 μm.

3. The analytic continuation of the reflectance and phase

If the experimental misplacement is Δl, the difference of the optical paths between the sample and reference pulse will be equal to $\Delta L = 2\Delta l \cdot cos(\theta_i)$. The measured phase function, $\phi_{exp}(\omega)$, is given by

$$\phi_{exp}(\omega) = \phi(\omega) + \Delta L \cdot |k| \qquad (1)$$

where $\phi(\omega)$ is the true phase without a misplacement, $|k| = \omega/c$ is the modulus of the wave vector and c is the speed of light in vacuum. A correcting phase term, $\beta = 2l_c \cdot cos(\theta_i) \cdot |k|$, is added into the eq. (1) to remove the linear phase error $\Delta L \cdot |k|$. Then we get the corrected phase

$$\phi_c(\omega, l_c) = \phi(\omega) + 2(\Delta l + l_c) \cdot cos(\theta_i) \cdot |k| \quad (2)$$

Eventually, the aim is to find a correcting misplacement term l_c to minimize $\Delta l + l_c$.

The methods based on K-K relations have been developed for many years. However, it's still difficult to determine l_c accurately with the limited spectra. In this paper, we proposed the method below to determine the permittivity more accurately.

The method procedure can be summarized as:

(1) Samples in our experiment are crystals and polymers with flat surfaces. Therefore, based on physical properties of a sample, we could choose a proper physical model to extend the reflectance and phase analytically. For instance, in the THz frequency range the crystal of $SrTiO_3$ should be described by a generalized oscillator mode:

$$\tilde{\epsilon}(\omega) = \epsilon_\infty \prod_j \frac{\omega_{LOj}^2 - \omega^2 + i\omega\gamma_{LOj}}{\omega_{TOj}^2 - \omega^2 + i\omega\gamma_{TOj}} \qquad (3)$$

where ω_{TOj} and ω_{LOj} mark the transverse and longitudinal frequencies of the jth mode, respectively, and γ_{TOj} and γ_{LOj} denote their corresponding damping constants.

(2) By the physical model above, the amplitude reflectance $r(\omega)$ can be fitted and the relative phase term of the fitting model, $\phi_{fit}(\omega)$, can be acquired by the Fresnel's law.

(3) Finally, the correcting misplacement term l_c is determined by the following norm:

$$Error(l_c) = \int_{\omega_a}^{\omega_b} \left| \phi_c(\omega, l_c) - \phi_{fit}(\omega) \right| d\omega \qquad (4)$$

where ω_a and ω_b is the maximum and minimum frequency, respectively. One may expect that when $Error(l_c)$ reaches its minimum at $l_c = l_{min}$, the value of l_{min} would be close to that of Δl.

4. Experimental Results and Discussions

In our experiment, we measured the complex reflectance of $SrTiO_3$(STO), which is perovskite structure and high-permittivity. STO is a kind of incipient ferroelectrics which is widely used for various microwave applications. In our experiment, $SrTiO_3$ (100) crystal is purchased from Heifei Kejing Materials Technology Co. Ltd and the size is 10mm×10mm with 1mm thick.

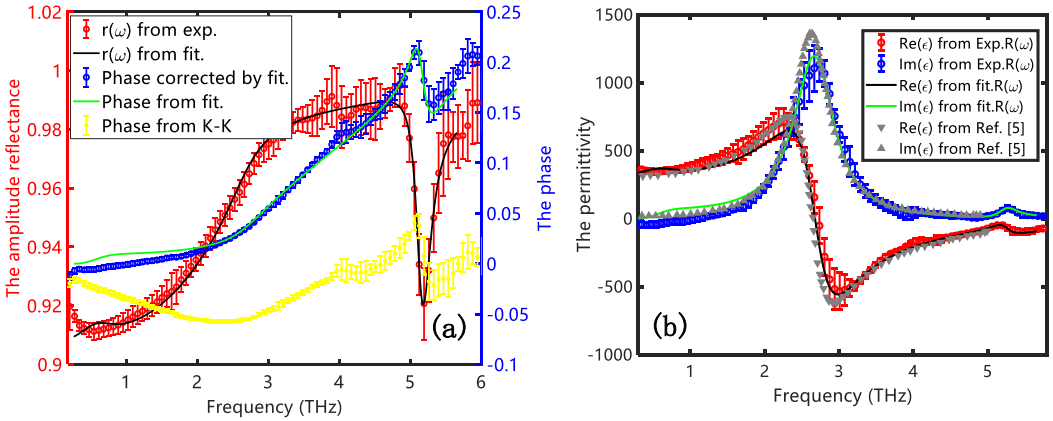

Fig. 1 (a) The complex amplitude reflectance of $SrTiO_3$. The red dots and the black line are the amplitude reflectance of the experiment and the fitting. The blue dots and the yellow ones are the phase terms corrected by the methods of Gornov and us. The green line is the phase term from the fitting physical model. (b) The permittivity of $SrTiO_3$. The experimental permittivity, $Exp.Re(\tilde{\epsilon})$ and $Exp.Im(\tilde{\epsilon})$, is calculated from the complex reflectance corrected by our method. The fitting permittivity, $Fit.Re(\tilde{\epsilon})$ and $Fit.Im(\tilde{\epsilon})$, is obtained from the fitting physical model. The down and up triangles are the permittivity from Ref [8], which is measured by terahertz time-domain spectroscopic ellipsometry.

Fig.1(a) shows the amplitude reflectance and the relative phase of STO from 0.3THz to 6 THz. The amplitude reflectance is beyond 0.9, which results in the sensitivity of permittivity to the error of reflectance $\Delta r(\omega)$. It's due to $\Delta r(\omega)$ inducing an error on the modulus of refractive index as following:

$$\frac{|\Delta\tilde{n}|}{|\tilde{n}|} \approx \frac{2 \cdot \Delta r(\omega)}{1 - r^2(\omega)} \qquad (5)$$

where the small phase term $\phi(\omega)$ is ignored in the derivation. As $r(\omega)$ is larger than 0.9, the influence of $\Delta r(\omega)$ on $|\Delta\tilde{n}|$ would be magnified ten-times at least. In $Fig1\ (a)$ the max error of $r(\omega)$ is about 2%, which is qualified for the determination of permittivity.

Based on the fitting of STO's amplitude reflectance (the black line In Fig.1(a)) by eq. (5), the fitting phase term $\phi_{fit}(\omega)$ of STO's complex reflection (the green line In Fig.1(a)) is obtain. At $l_c = l_{min}$, the corrected phase term $\phi_c(\omega, l_c)$ (the blue dots in Fig.1(a)) can overlap the fitting phase term $\phi_{fit}(\omega)$ well. Fig.1(b) shows the permittivity calculated by the corrected and fitting phase terms in Fig.1(a). The corrected permittivity (the red and blue dots in Fig.1(b)) matches that measured by terahertz time-domain spectroscopic ellipsometry (the up and down triangles) [8]. However, the method coming from K-K relations obviously gives a wrong corrected phase as indicated by the yellow dots in Fig.1(a). The corrected misplacement term l_c of yellow dots in Fig.1(a) is $0.8\mu m$ deviated from that of blue dots. This deviation causes a complete error of STO's permittivity.

There are two possible reasons responsible for the deviation of K-K relations:

(1) The absorption coefficient of the $SrTiO_3$ is very big over 2 THz. As the error of the calculated refractive index is approximately proportional to the absorption coefficient [9], the misplacement has a strong impact on the permittivity of STO.

(2) In the limited spectrum of 0.3THz to 6 THz, the corrected misplacement of K-K relation is $0.8\mu m$ deviated from the right result, which means the influence of K-K relations on STO is serious.

In general, there's no problem fitting the amplitude reflectance by a physical model. However, when the corrected phase $\phi_c(\omega, l_c)$ deviates from the fitting phase $\phi_{fit}(\omega)$ or the horizontal axis seriously, the physical model should be considered again. In Fig.1 (a), the lines of $\phi_c(\omega, l_c)$ and $\phi_{fit}(\omega)$ coincide well with each other, which means a nice analytic continuation of the complex reflection.

5. Summary

In this paper, we proposed that the analytic continuation of the complex reflectance by a proper physical model is a more accurate and simpler method than that based on K-K relations. For comparison, the complex reflectance of $SrTiO_3$ was corrected by the methods of Gornov and us. Due to the limited spectra in our experiment, the method of Gornov which was based on K-K relations didn't correct the complex reflectance accurately. Nevertheless, our method determined an accurate and reasonable permittivity for the analytical extension of the complex reflection. At the same time the dielectric property and the physical mechanism were researched, which was beneficial for the application of materials in THz gap.

6. References

[1] Max Khazan, Reinhold Meissner, and Ingrid Wilke, "Convertible transmission-reflection time-domain terahertz spectrometer," Rev. Sci. Instrum. 72, 3427-3430 (2001).

[2] A. Pashkin, M. Kempa, H. Nemec, F. Kadlec, and P. Kuzel, "Phase-sensitive time-domain terahertz reflection spectroscopy," Rev. Sci. Instrum. 74, 4711-4717 (2003).

[3] S. Nashima, O. Morikawa, K. Takata, and M. Hangyo, "Measurement of optical properties of highly doped silicon by terahertz time domain reflection spectroscopy," Appl. Phys. Lett. 79, 3923-3925 (2001).

[4] Erik M. Vartiainen, Yusuke Ino, Ryo Shimano, Makoto Kuwata-Gonokami, Yuri P. Svirko, and Kai-Erik Peiponen, "Numerical phase correction method for terahertz time-domain reflection spectroscopy," J. Appl. Phys. 96, 4171 (2004).

[5] Valerio Lucarini, Yusuke Ino, Kai-Erik Peiponen, and Makoto Kuwata-Gonokami, "Detection and correction of the misplacement error in terahertz spectroscopy by application of singly subtractive Kramers-Kronig relations," Phys. Rev. B 72.125107 (2005).

[6] E. Gornov, K.-E. Peiponen, Y. Svirko, Y. Ino, and M. Kuwata-Gonokami, "Efficient dispersion relations for terahertz spectroscopy," Appl. Phys. Lett. 89, 142903 (2006).

[7] K-E Peiponen and J J Saarinen, "Generalized Kramers–Kronig relations in nonlinear optical- and THz-spectroscopy," Rep. Prog. Phys. **72** 056401 (19pp) (2009).

[8] Naoki Matsumoto, Takashi Fujii, Keisuke Kageyama, Hiroshi Takagi, Takeshi Nagashima, and Masanori Hangyo, "Measurement of the Soft-Mode Dispersion in SrTiO3 by Terahertz Time-Domain Spectroscopic Ellipsometry," Jpn. J. Appl. Phys. 4, 09KC11 (2009).

[9] Maxime Bernier, Frédéric Garet, and Jean-Louis Coutaz, "Precise Determination of the Refractive Index of Samples Showing Low Transmission Bands by THz Time-Domain Spectroscopy," IEEE Trans. THz Sci. Techn., 3, 295–301 (2013).