

Optical properties of SrTiO₃: the substrate for interface superconductivity

S. V. DORDEVIC*

Department of Physics, The University of Akron, Akron, OH 44325 USA

The last few years have seen a flurry of interest in the so-called interface superconductivity. Superconductivity with critical temperature as high as 100 K has been reported in FeSe/SrTiO₃ structures. The roles of the two materials and their interface is currently being debated. I report here the optical properties of a SrTiO₃ substrate that is used for these structures. Room temperature spectra are collected over a broad frequency range, from far-infrared to near-ultraviolet. Overall, the transmission is found to be very low, typically below 4 %. The band gap, above which the substrate is completely opaque, is found to be $E_g = 3.21$ eV. The reflectance, on the other hand, is higher and reveals signatures of interband transitions.

(Received October 18, 2016; accepted October 10, 2017)

Keywords: STO, SrTiO₃ substrate, Transmission, Reflection, Interface superconductivity

1. Introduction

Interface superconductivity was predicted more than half a century ago [1], but only recently realized experimentally in bilayers of insulating La₂CuO₄ and metallic La_{1.55}Sr_{0.45}CuO₄ [2, 3]. In the past few years this phenomenon has attracted a lot of attention when it was found that the critical temperature of bulk FeSe can be increased several times [4] (T_c s higher than 100 K have been reported [5]) when grown epitaxially on SrTiO₃. Superconductivity can also be induced in insulating LaAlO₃ when grown on SrTiO₃ [6]. The mechanism of superconductivity in all these systems is currently being debated [7, 8, 9]. Interface superconductivity is obviously a very promising research avenue and a wealth of new discoveries can be anticipated.

Strontium titanate is in itself a fascinating material that has a number of interesting properties, including superconductivity (with $T_c < 0.5$ K when doped with niobium [10, 11]), ferromagnetism (with $T_c < 10$ K when doped with lanthanum [12]), Fermi liquid behaviour [13, 14], persistent photoconductivity [15], etc. SrTiO₃ has also been used for a number of practical applications, such as a building block in oxide electronics, particularly for the formation of two-dimensional electron liquids [16]. For a number of years, SrTiO₃ has been used for high-voltage capacitors, because of its high static dielectric constant ($\epsilon \approx 300$ at room temperature) [17].

In this paper I report on the optical properties of a SrTiO₃ substrate at room temperature. Transmission $T(\omega)$ of a commercially available substrate is measured over a wide range of frequencies, from far-infrared to near ultraviolet. Overall, the transmission of SrTiO₃ substrate is low, below 4 % over most of the measured frequency range. The substrate is found to be completely opaque in the far-infrared part of the spectrum, as well as above the

energy gap, which is located at $E_g = 3.21$ eV. In this range, the transmission is supplemented with the near-normal-incidence reflectance $R(\omega)$. The spectra were analyzed using the so-called Tauc-Lorentz model and from this analyses the parameters of the electronic structure of SrTiO₃ were extracted. Reported results will also be relevant for other spectroscopic measurements involving SrTiO₃ substrates, especially laser-based techniques, such as Raman or time-domain spectroscopies.

2. Experimental results

Single crystal substrates of SrTiO₃ are available commercially from several vendors. In this study a substrate with dimensions 10 mm × 10 mm × 0.5 mm was used. One side of the substrate was mechanically polished by the vendor to an optically flat surface. Transmission measurements were carried out over a wide frequency range from far-infrared to near ultraviolet using several different experimental setups to cover the whole range [18]. In the far- and mid-infrared Bruker 60 FTIR was used, whereas in the near-infrared, visible and near-ultraviolet Varian Cary 300 was employed. Cary 300 was also used for reflection measurements.

Fig. 1 shows the transmission over the whole frequency range. As can be seen, the transmission is below 4 %, except in the region between 1,350 – 3,100 cm⁻¹ where it peaks at about 23 %. In the far-infrared part of the spectrum transmission is zero, due to strong infrared active phonon modes [10]. Phonons in SrTiO₃ have been discussed before [19, 20, 21] and are not the subject of this work. Below the lowest phonon frequency, the transmission increases again towards microwave part of the spectrum.

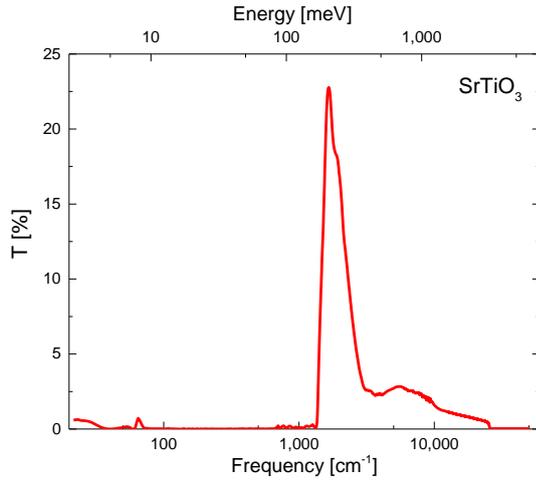


Fig. 1. Room temperature transmission spectrum of SrTiO₃ substrate. Transmission is shown over a broad frequency (energy) range, from the far-infrared to near-ultraviolet

In the near-infrared and visible part of the spectrum transmission is also very low and vanishes completely above 3.21 eV. I identify this value as the energy of the indirect band gap, which is about 1.2 % smaller than a previously reported value (3.25 eV) [22]. Note that the direct band gap in SrTiO₃ is higher than the indirect, and cannot be observed in transmission spectra. Energy of the direct gap (3.75 eV [22]) is marked with a vertical dashed line in Fig. 2(a). In this part of the spectrum around the gap, the transmission measurements were supplemented with the reflection of the same substrate. These measurements were performed in the slab geometry [23], to allow for free reflection from the back side of the substrate. The results are shown in Fig. 2(a), which for comparison also includes part of transmission spectrum from Fig. 1, on an expended scale. As can be seen, in the region above the gap, the reflectance is dominated by interband transitions and its values are higher than transmission, typically between 15–30 %. We also note that, unlike the transmission, the reflection spectrum does not reveal any signatures of band gaps (neither direct nor indirect).

3. Analysis and discussion

In this section I analyze the data to gain a better insight into absorption and dispersion properties of SrTiO₃ around the band gap. For that purpose, I *simultaneously* fit the reflectance and transmission data (Fig. 2(a)) using the so-called Tauc-Lorentz model [24, 25]. This is a modified version of the Lorentz model [23], which takes into account the presence of a gap in the excitation spectrum. The model is frequently used to account for absorption across a band gap, especially in (amorphous) semiconductors and insulators [24]. Within this model, the imaginary (dissipative) part of the dielectric function is given as:

$$\varepsilon_2(\omega) = \frac{1}{\omega} \frac{\omega_p \omega_0 \gamma (\omega - \omega_g)^2}{\omega (\omega^2 - \omega_0^2)^2 + \gamma^2 \omega^2} \Theta(\omega - \omega_g) \quad (1)$$

and the corresponding real part $\varepsilon_1(\omega)$ can be calculated using a KK transformation:

$$\varepsilon_1(\omega) = 1 + \frac{2}{\pi} \int_0^\infty \frac{\omega' \varepsilon_2(\omega')}{\omega'^2 - \omega^2} d\omega' \quad (2)$$

In Eq. 1 ω_0 , ω_p and γ are the central frequency, oscillator strength and the width of the mode, whereas ω_g is the gap frequency. $\Theta(x)$ is the Heaviside step function, which assures that there is no absorption below the gap. It is easy to see that for $\omega_g = 0$ Eq. 1 reduces to a Lorentz mode [23].

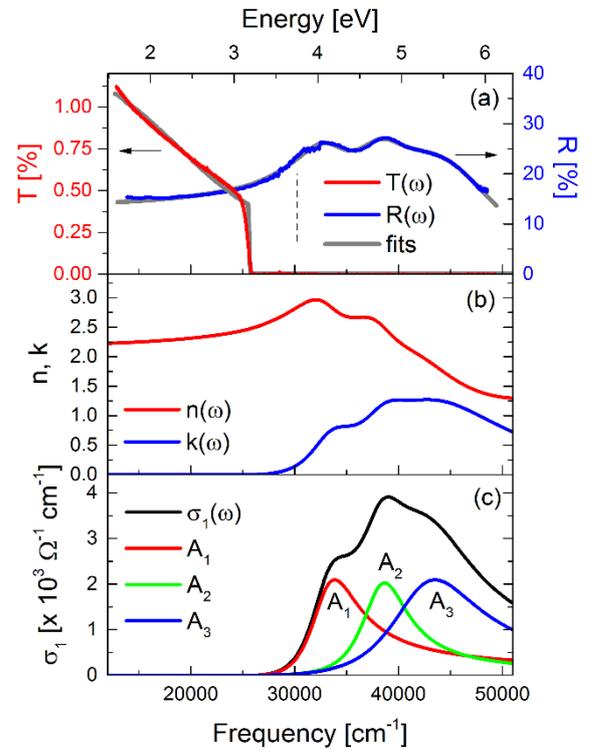


Fig. 2. Optical functions of SrTiO₃. (a) The reflection spectrum, along with a part of transmission spectrum from Fig. 1. The best fits to the Tauc-Lorentz model are shown with gray lines, generated from the simultaneous fits of reflectance and transmission. Vertical dashed line marks the value of the direct band gap in SrTiO₃ (3.75 eV). (b) Real and imaginary parts of the complex index of refraction $n(\omega)$ and $k(\omega)$. (c) Real part of the optical conductivity $\sigma_1(\omega)$ generated from the best fits in panel (a). Also shown are the three components of the fit, previously labeled as A_1 , A_2 and A_3 [22].

Modeling of the data was done with Ref FIT [25] which has the Tauc-Lorentz model implemented. The simultaneous fits of both reflectance and transmission provide stringent constraints on the fitting parameters.

Satisfactory fits are achieved with four oscillators. Three of them are clearly visible in reflectance (Fig. 2(a)), and therefore three Tauc-Lorentz modes are used in the fits. In addition, a (very) weak Lorentz oscillator is used to account for a small, but finite (. 1 %) transmission below the gap. The best fits are shown in Fig. 2(a) with gray lines, and as can be seen, the model is capable of capturing all the essential features of both transmission and reflection data.

From the best fit one can generate all other optical functions of interest, such as the complex index of refraction ($N(\omega) = n(\omega) + ik(\omega)$) and complex optical conductivity ($\tilde{\sigma}(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$) [23]. The $N(\omega)$ spectra are shown in Fig. 2(b), and are qualitatively similar to the previously reported [22] obtained using different experimental techniques. Fig. 2(c) shows the real part of optical conductivity $\sigma_1(\omega)$. This panel also shows the three individual components of the fit, which were previously labeled as A1, A2 and A3 [26, 22]. The fitting procedure employed here allows one to extract not only the energy of the transition ω_0 , but also its strength ω_p and width γ (which is related to the lifetime of the electronic transitions). Table 1 gives the values of all parameters, as well as the values of ω_0 from Ref. [22]. Comparing these values, one can see that the values reported here are approximately 2.5 % smaller than those from Ref. [22]. I also point out that the intensity of the fourth mode is much smaller than the three modes shown, and consequently this mode cannot be seen in Fig. 2(c).

Table 1. Parameters of the best fits from the simultaneous fits of transmission and reflection. ω_0 , ω_p and γ are from Eq. 1. Also shown are the values of ω_0 from Ref. [22]. All parameters are reported in eV.

oscillator	ω_0 (eV)	ω_p (eV)	γ (eV)	ω_0 from Ref. [22] (eV)
A ₁	4.10	43.44	0.67	4.2
A ₂	4.78	19.18	0.68	4.9
A ₃	5.28	24.54	1.32	5.4

4. Summary

In summary, optical properties of SrTiO₃ substrate are reported over a broad frequency range. Overall, transmission is found to be low, and in some parts of the spectrum equal to zero. In particular, the transmission is vanishing above 3.21 eV, which is identified as the energy of the indirect band gap. The reflectance on the other hand is higher, and reveals signatures of interband transitions. From the fits with the Tauc-Lorentz model, parameters of the electronic structure of SrTiO₃ are extracted and compared with previously reported values. These results will be relevant for all spectroscopic studies, especially laser-based, involving SrTiO₃ substrate. Depending on the required frequency range, those experiments will have to be done either in reflection or transmission geometry.

References

- [1] V. L. Ginsburg, Physics Letters **13**, 101 (1964).
- [2] A. Gozar, G. Logvenov, L. Fitting Kourkoutis, A. T. Bollinger, L. A. Giannuzzi, D. A. Muller, I. Bozovic, Nature **455**, 782 (2008).
- [3] I. Bozovic, C. Ahn, Nature Physcis **10**, 892 (2014).
- [4] Q. Y. Wang, Z. Li, W.-H. Zhang, Z.-C. Zhang, J.-S. Zhang, W. Li, H. Ding, Y.-B. Ou, P. Deng, K. Chang, J. Wen, C.-L. Song, K. He, J.-F. Jia, S.-H. Ji, Y.-Y. Wang, L.-L. Wang, X. Chen, X.-C. Ma, Q.-K. Xue, Chinese Physics Letters **29**, 037402 (2012).
- [5] J.-F. Ge, Z.-L. Liu, C. Liu, C.-L. Gao, D. Qian, Q.-K. Xue, Y. Liu, J.-F. Jia, Nature Materials **14**, 285 (2015).
- [6] N. Reyren, S. Thiel, A. D. Caviglia, L. Fitting Kourkoutis, G. Hammerl, C. Richter, C. W. Schneider, T. Kopp, A.-S. Ruetschi, D. Jaccard, M. Gabay, D. A. Muller, J.-M. Triscone, J. Mannhart, Science **317**, 1196 (2007).
- [7] D.-H. Lee, Chinese Physics B **24**(11), 117405 (2015).
- [8] Z.-X. Li, F. Wang, H. Yao, D.-H. Lee, Science Bulletin **61**(12), 925 (2016).
- [9] K. Zou, S. Mandal, S. D. Albright, R. Peng, Y. Pu, D. Kumah, C. Lau, G. H. Simon, O. E. Dagdeviren, X. He, I. Bozovic, U. D. Schwarz, E. I. Altman, D. Feng, F. J. Walker, S. Ismail-Beigi, Charles H. Ahn, Physical Review B **93**(18), 180506(R) (2016).
- [10] J. L. M. van Mechelen, D. van der Marel, C. Grimaldi, A. B. Kuzmenko, N. P. Armitage, N. Reyren, H. Hagemann, I. I. Mazin, Physical Review Letters **100**(22), 226403 (2008).
- [11] J. Ruhman, P. A. Lee, Physical Review B **94**, 224515 (2016).
- [12] P. Moetakef, J. R. Williams, D. G. Ouellette, A. P. Kajdos, D. Goldhaber-Gordon, S. J. Allen S. Stemmer, Physical Review X **2**, 021014 (2012).
- [13] D. van der Marel, J. L. M. van Mechelen, I. I. Mazin, Physical Review B **84**, 205111 (2011).
- [14] X. Lin, B. Fauque, K. Behnia, Science **349**(6251), 945 (2015).
- [15] M. C. Tarun, F. A. Selim, M. D. McCluskey, Physical Review Letters **111**(18), 187403 (2013).
- [16] M. Yang, M. Pierre, O. Toressin, M. Goiran, W. Escoffier, S. Zeng, Z. Huang, H. Kun, T. Venkatesan, Ariando, M. Coey, Applied Physics Letters **109**, 122106 (2016).
- [17] A. A. Demkov, A. B. Posadas, Integration of Functional Oxides with Semiconductors, Springer 25 (2014).
- [18] S. V. Dordevic, L. W. Kohlman, N. Stojilovic, R. Hu C. Petrovic, Physical Review B **80**(11), 115114 (2009).
- [19] A. S. Barker, J. J. Hopfield, Physical Review **135**(6A), 1732 (1964).
- [20] K. Kamaras, K.-L. Barth, F. Keilmann, R. Henn, M. Reedyk, C. Thomsen, M. Cardona, J. Kircher, P. L. Richards, J.-L. Stehle, Journal of Applied Physics **78**(2), 1235 (1995).

- [21] J. L. M. van Mechelen, D. van der Marel, C. Grimaldi, A. B. Kuzmenko, N. P. Armitage, N. Reyren, H. Hagemann, I. I. Mazin, *Physical Review Letters* **100**(22), 226403 (2008).
- [22] K. van Benthem, C. Elsasser, R. H. French, *Journal of Applied Physics* **90**(12), 6156 (2001).
- [23] M. Dressel, G. Gruner, *Electrodynamics of Solids*, Cambridge University Press, Cambridge (2001).
- [24] P. Yu, M. Cardona, *Fundamentals of Semiconductors: Physics and Materials Properties*, Springer (2010).
- [25] A. B. Kuzmenko, *Review of Scientific Instruments* **76**(8), 083108 (2005).
- [26] D. Bauerle, W. Braun, V. Saile, G. Sptissel, E. E. Koch, *Zeitschrift für Physik B* **29**(3), 179 (1978).

*Corresponding author: dsasa@uakron.edu