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ABSTRACT
Near-infrared absorption in transparent conducting oxides (TCOs) is usually caused by electronic intraband transition at high doping levels. Improved infrared transparency is commonly explained by enhanced drift mobility in these TCOs. Here, an alternative cause behind the high infrared transparency of La-doped barium stannate (LBSO) transparent electrodes is presented. Following the Drude model formalism, we reconstructed spectrally resolved dielectric permittivity for a set of thin films with different free electron concentrations. A comparison of optical properties of LBSO with the tin-doped indium oxide thin films with identical carrier concentrations suggests that the redshift of the screened plasma wavelength for LBSO originates from its large high-frequency dielectric constant of 4.4, one of the highest reported for the s-orbital-based TCOs. Moreover, our measurements confirm an optical mobility significantly higher (>300 cm$^2$/V s) than the drift mobility, effectively suppressing the free carrier absorption. These factors enable high infrared transparency of LBSO films and motivate further exploration of LBSO as broadband TCOs for solar cells and nanophotonics.

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I. INTRODUCTION
Transparent conducting oxides (TCOs) combine low optical absorption in the visible spectral range with high electrical conductivity, which is achieved by the combination of their large bandgap energy (>3 eV) and ability to support high doping densities ($N_e$) of $10^{20}$–$10^{21}$ cm$^{-3}$. These properties led to the extensive usage of TCOs as electrodes for optoelectronic applications, which require transparency in visible and/or near-infrared regions (NIR) of the spectrum (e.g., tandem photovoltaics).

For wavelength larger than $\lambda_p$, $\epsilon' < 0$, hence, the TCO becomes reflective. A characteristic TCO thin film for front electrode application in, e.g., solar cells presents a $\lambda_p$ and the onset of a highly reflecting IR region outside the device relevant (300–1200 nm) spectral range. However, the magnitude of $\lambda_p$ additionally affects the range of near-IR transparency, since in its vicinity, there is a rise in the free carrier absorption (FCA) due to electronic intraband transitions.

\[ \lambda_p = \frac{2\pi c}{\omega_p} = \frac{2\pi c}{\sqrt{\epsilon_0 \epsilon_\infty m^* N_e e^2}}, \]  

where $c$ is the speed of light in vacuum, $\omega_p$ is the screened plasma frequency, $\epsilon_0$ is the vacuum permittivity, $\epsilon_\infty$ is the high-frequency dielectric constant of the material, and $m^*$ is the electron effective mass. 

For wavelength larger than $\lambda_p$, $\epsilon' < 0$, hence, the TCO becomes reflective. A characteristic TCO thin film for front electrode application in, e.g., solar cells presents a $\lambda_p$ and the onset of a highly reflecting IR region outside the device relevant (300–1200 nm) spectral range. However, the magnitude of $\lambda_p$ additionally affects the range of near-IR transparency, since in its vicinity, there is a rise in the free carrier absorption (FCA) due to electronic intraband transitions.
Therefore, the device community is striving to mitigate the loss of transparency due to FCA by choosing TCO films with reduced carrier densities. In this case, films with high carrier mobilities (μe) are desired to enable the lateral conductivity required for front electrode applications. Tin-doped In2O3 (ITO) is the most extensively studied TCO for both optoelectronic and nanophotonic applications. Although ITO maintains an optical transparency of >90% in the visible range and provides resistivities below $10^{-4}$ Ω cm, it has moderate μe of around 30 cm²/V s. Hydrogenation of In2O3 or efficient co-doping with other metal atoms such as Mo or Zr enabled the fabrication of thin films with μe exceeding 100 cm²/V s. However, due to the scarcity of In as an element in the earth’s crust,10 its widespread use is limited by the scarcity of In as an element in the earth’s crust,10 and the competitiveness of other metal atoms such as Mo8 and Zr9 enabled the fabrication of thin films with μe exceeding 100 cm²/V s. Moreover, ITO maintains high optical transmittance (75% in the visible range) for both optoelectronic and nanophotonic applications. Although ITO maintains optical transparency of >90% in the visible range and provides resistivities below $10^{-4}$ Ω cm, it has moderate μe of around 30 cm²/V s. Hydrogenation of In2O3 or efficient co-doping with other metal atoms such as Mo or Zr enabled the fabrication of thin films with μe exceeding 100 cm²/V s. However, due to the scarcity of In as an element in the earth’s crust,10 significant effort was dedicated to the development of alternative TCOs primarily consisting of abundant elements.

In this regard, La-doped barium stannate (LBSO) has gained a lot of research attention recently due to extraordinary high μe of 320 cm²/V s reported for single crystals.11 This achievement triggered extensive investigations into carrier transport properties of high-quality LBSO thin films1,2,8,11 and enabled their application as channel layer for oxide transistors.12,13 However, the optical properties of LBSO, which are crucial for its application as efficient transparent electrodes or as low propagation loss nanophotonic components, are widely missing in the literature. Liu14 and Niedermeier15 have reported a high optical transmittance >75% in the visible spectrum and >60% transmittance up to 1500 nm for epitaxially grown 100-nm-thick LBSO films on MgO and SrTiO3 (STO) substrates, respectively. It is noteworthy that the mobility of the films did not exceed 45 cm²/V s and could not fully explain the enhanced IR transmittance for high doping levels (>6 × 10²⁰ cm⁻³) in the former case.

This study presents a quantitative analysis of dielectric permittivity for a set of LBSO thin films with different Nc. For that, we have employed Photothermal Deflection Spectroscopy (PDS) measurements in the 290–1620 nm spectral range. Using the Drude theory, we estimate the high-frequency permittivity of LBSO. The influence of this intrinsic property on IR transmittance is discussed in comparison with ITO.

II. EXPERIMENTAL

Epitaxial films of LBSO of ~120 nm were grown on MgO (001) substrates by pulsed laser deposition (PLD) at 750°C. Substrates were annealed in vacuum at the growth temperature for 1 h prior to deposition. The LBSO target with a La doping concentration of either 4 at.% or 7 at.% was ablated by using an excimer laser (KrF, 248 nm). The oxygen deposition pressure was kept at 0.26 mbar, laser fluence at 1.35 J/cm², and repetition rate at 1 Hz, respectively. Heteroepitaxy was confirmed by high-resolution transmission electron microscopy (TEM) [Fig. 4(a)]. Transmittance was measured using an UV-visible-NIR spectrophotometer (Perkin Elmer Lambda 950S). Electrical transport properties (Nc and μe) were extracted from the Hall effect measurements in van der Pauw geometry using a Quantum Design Physical Properties Measurement System at room temperature.

PDS10 was performed by an in-house developed system based on a 150-W xenon lamp and a three-grating monochromator. Fluorinert FC-72 was used as a temperature-sensitive liquid. The specular transmittance and reflectance were measured simultaneously, and the absorption coefficient was evaluated as described by Ritter and Weiser,16 corrected for second-order terms. The refractive index (n) was fitted by the Drude–Sellmeier model.17 The evaluation was complemented by comparing the modeled and directly measured absorption coefficient (α) in the IR region, where FCA in LBSO thin films dominates. The raw data of absorption and reflectance measurements, fitting and further details of PDS analysis are provided in the supplementary material (Fig. S2).

Reference refractive index (n) and extinction coefficient (k) of sputtered ITO films for different carrier densities measured by spectroscopic ellipsometry were obtained from Holman et al.18

III. RESULTS AND DISCUSSION

A. Optoelectrical properties of epitaxially grown LBSO films

Figure 1 shows the optical transmittance of LBSO thin films grown on MgO substrates with Nc ranging from $1.9 \times 10^{20}$ cm⁻³ to $5.0 \times 10^{20}$ cm⁻³. The NIR transmittance consistently decreases with the increase in Nc associated with stronger FCA following Eq. (3). Noticeably, the LBSO films maintain high IR transmittance (>75% at 1200 nm) even for the highest Nc. As summarized in Table I, such carrier density with μe of 43 cm²/V s leads to Rsh of only 23 Ω/□ for a 120-nm-thick film. In contrast, commercially available ITO thin films deposited on glass with equal Rsh and same thickness are considerably less transparent in the 1000–1800 nm range as indicated by the dashed line in Fig. 1. We note that while the measurements in Fig. 1 are substrate- and thickness-dependent, the optical properties presented and analyzed in the rest of the manuscript are intrinsic to the TCO, i.e., substrate- and thickness-independent.

It is noteworthy that the highest μe is achieved for the LBSO sample with the highest Nc of $5 \times 10^{20}$ cm⁻³ (Table I), whereas...
TABLE I. Electrical properties of 120-nm-thick LBSO thin films grown on MgO.

| N_e (10^{20} cm^{-3}) | \(\mu_e\) (cm^2/V s) | \(\sigma\) (S/cm) | \(R_{sh}(\Omega/\square)\) |
|------------------------|----------------------|------------------|------------------------|
| 1.9                    | 15                   | 432              | 230                    |
| 3.8                    | 35                   | 2136             | 39                     |
| 5.0                    | 43                   | 3623             | 23                     |

optimum \(\mu_e\) at lower \(N_e\) of 1–3 \(\times 10^{20}\) cm\(^{-3}\) is demonstrated for many In-based TCOs.\(^{2,24}\) Such a difference was previously reported\(^{25,26}\) and is mainly attributed to the efficient screening of charged impurities in LBSO. This is due to the high static dielectric constant \((\varepsilon = 21)\) of the BaSnO\(_3\)\(^{27}\) host enabling large mobility at high carrier concentrations.

B. Comparison of the high-frequency dielectric constant for different TCOs

For an accurate comparison of optical properties and decoupling reflectance and absorption losses, we analyzed the IR dielectric functions of LBSO and ITO with similar \(N_e\). The complex IR dielectric function \(\varepsilon^*\) of degenerate semiconductor thin films in the spectral range between 800 nm and 1600 nm is described by the Drude model and can be expressed by

\[
\varepsilon^* = (n - ik)^2 = \varepsilon_\infty - \frac{\varepsilon_\infty\omega_p^2}{\omega^2 - i\omega/\tau}.
\]

The \(\lambda_p\) is defined by the crossing at zero of the real part of the dielectric permittivity \((\varepsilon')\). Following this and Fig. 2(a), the \(\lambda_p\) for ITO with \(N_e\) of \(1524\) nm, while for LBSO \(\lambda_p\) is above \(1620\) nm (no crossing determined up to \(1620\) nm). The real part of the dielectric permittivity fitted by the Drude model (shown with dashed lines in the graph) indicates a \(\lambda_p\) at \(1630\) nm for the highest doping of LBSO thin films. Further details of the fitting procedure would be discussed later in the section.

The observed difference in \(\lambda_p\) for films of LBSO and ITO in Fig. 2(a) is attributed to the distinction in the \(\varepsilon_\infty m^*\) following Eq. (1). The effective masses of these TCOs do not differ dramatically and were shown to be 0.3m\(_0\) for LBSO\(^{25}\) and 0.35m\(_0\) for ITO\(^{26}\) for similar \(N_e\) as in this study. Therefore, shifted \(\lambda_p\) originates from the higher \(\varepsilon_\infty\) of LBSO that is shown in Fig. 2(b), where \(\varepsilon_\infty\) can be determined as the intercept of the dielectric function plotted vs \((1/\hbar\omega^2)\)\(^{33,34}\), where \(\hbar\) is the reduced Planck constant. \(\varepsilon_\infty\) of LBSO is found to be 4.4 for \(N_e = 5.0 \times 10^{20}\) cm\(^{-3}\). For ITO, \(\varepsilon_\infty\) is only 3.85 (Table II), which is in good agreement with the reports from the literature for the same \(N_e\).

To validate the accuracy of the determined \(\varepsilon_\infty\) and \(m^*\), we fitted the real and the imaginary parts of the dielectric permittivity following Drude free electron theory utilizing estimated values from Table III for \(N_e = 5.0 \times 10^{20}\) cm\(^{-3}\) and Table S2 of supplementary material. Good agreement between the fitted spectrum [dashed line in Figs. 2(a) and Fig. S4(b)] and the experimental data verifies the correctness of our estimations.

It is important to mention that \(\varepsilon_\infty\) of 4.4 for LBSO film with \(N_e = 5.0 \times 10^{20}\) cm\(^{-3}\) is one of the highest reported for the s-orbital-based TCOs. It is noteworthy that LBSO displays a unique combination of properties, benefiting from small \(m^*\) and large \(\mu_e\) of conventional s-orbital-based TCOs and relatively large \(\varepsilon_\infty\) and \(\varepsilon_e\), which are common for d-orbital-based TCOs. The small \(m^*\) of LBSO originates from a dispersive conduction band mainly composed of Sn 5s bands. Relatively large \(\varepsilon_e\) of LBSO, in its turn, results from the strongly ionic chemical bonds and the ion displacement from the equilibrium lattice in the perovskite structure of LBSO.

On the other hand, d-orbital-based TCOs such as the doped STO and TiO\(_2\) possess even larger \(\varepsilon_\infty\) enabling a further redshift of \(\lambda_p\) as summarized in Table II. However, transition-metal based TCOs with large \(\varepsilon_\infty\) exhibit significantly lower \(\mu_e\) (<10 cm\(^2\)/V s) due to their large \(m^*\) (>1m\(_0\)) and would require sufficient increase in the thickness for obtaining highly conductive films. \(\lambda_p\) for films with device-relevant \(N_e > 10^{21}\) cm\(^{-3}\) is below 2000 nm and comparable with films with \(N_e = 5.0 \times 10^{20}\) cm\(^{-3}\) in this study. However,
TABLE II. Screened plasma wavelength, drift mobility, high-frequency dielectric constant, static dielectric constant, and effective mass for s-orbital-based and d-orbital-based TCOs.

| Material                  | $\lambda_p$ for $N_e = 5.0 \times 10^{20}$ cm$^{-3}$ (nm) | $\mu_e$ (cm$^2$/V s) | $\varepsilon_\infty$ | $\varepsilon_s$ of host oxide | $m^*/m_0$ |
|--------------------------|----------------------------------------------------------|----------------------|----------------------|-------------------------------|-----------|
| Sn:In$_2$O$_3$ (ITO)/glass$^*$ | 1524                                                    | 28                   | 3.85                 | 8.5                           | 0.27      |
| La:BaSnO$_3$ (LBSO)/MgO—this work | 1629                                                    | 43                   | 4.40                 | 20                            | 0.27      |
| Nb:TiO$_2$ (NTO)/SrTiO$_3$(STO)$^b$ | 3735                                                    | 8                    | 6.25                 | 173                           | 1.0       |
| La:SrTiO$_3$ (LSTO)/SrTiO$_3$ (STO)$^c$ | 4575                                                    | 8                    | 5.20                 | 310                           | 1.8       |

$a$ Reference 23.
$b$ Reference 28.
$c$ References 29–31.

the increased $N_e$ would also lead to the enhanced parasitic absorption (as discussed in Sec. III C) and would benefit mitigates of larger $\lambda_p$ for d-orbital-based TCOs. Therefore, we would further focus on comparing the optical properties of LBSO with conventional ITO films.

C. Free carrier absorption and its relation to optical mobility

As mentioned earlier, the IR transparency of films in the vicinity of $\lambda_p$ is also limited by FCA. The absorption coefficient ($\alpha$) for highly doped semiconductors is expressed by

$$\alpha = \frac{\lambda^2 e^3 N_e}{4\pi^2 \varepsilon_0 c^3 n (m^*_e)^2 \mu_{opt}}$$

where $\lambda$ is the photon wavelength and $\mu_{opt}$ is the optical mobility of the material. It is important to mention that $\mu_{opt}$ in Eq. (3) is typically attributed to the in-grain mobility$^7$ since optical excitation is assumed to deflect free electrons over a distance shorter than the grain size. Therefore, $\mu_{opt}$ may differ from drift mobility $\mu_e$ extracted from the Hall effect as would be discussed below.

As indicated in Fig. 3(a), the absorption coefficient of ITO is higher than $5 \times 10^4$ cm$^{-1}$ at its $\lambda_p$ (1524 nm), whereas $\alpha$ of the LBSO thin films remains lower than $5 \times 10^3$ cm$^{-1}$ in the whole range of measurement. Given same $m^*$ for LBSO and ITO (Table II), this difference originates from differences in $n$ and/or $\mu_{opt}$ following Eq. (3). The comparison of $\alpha$ and $n$ for a telecommunication wavelength (1550 nm) is given in Table III. Slightly higher $n$ of LBSO (0.71 vs 0.53) and $\mu_e$ compared to ITO (43 cm$^2$/V s vs 28 cm$^2$/V s) are expected to decrease FCA for LBSO films but cannot be accountable for an order of magnitude difference in $\alpha$. It is plausible that the higher crystal quality of epitaxially grown film [Fig. 4(a)] causes differences between $\mu_e$ and $\mu_{opt}$ due to distinct carrier transport at grain boundaries, since the mean free path of electrons (<2 nm)$^{35}$ at such high $N_e$ is smaller than the typical grain sizes in both ITO$^4$ and LBSO films.$^{34}$

One can estimate optical relaxation time by correlating the real and the imaginary part of the dielectric permittivity following the derivation (Eqs. S2–S7 in supplementary material) of optical conductivity,$^5$

$$\varepsilon' = \varepsilon_{\infty} - \frac{\omega}{\tau} \varepsilon''.$$

![FIG. 3. (a) The absorption coefficient for thin films of LBSO and ITO with $N_e = 5.0 \times 10^{20}$ cm$^{-3}$. (b) Real part ($\varepsilon'$) of dielectric permittivity as a function of frequency times the imaginary part ($\varepsilon''$). The slope of the curve equals to the carrier relaxation time ($\tau$) and intercept equals to the high-frequency dielectric constant ($\varepsilon_{\infty}$).](attachment:image.png)
Equation (4) shows that plotting $\varepsilon'$ as a function of $\omega\varepsilon''$ should yield a straight line with the slope of the curve equal to the carrier relaxation time ($\tau$) and intercept equal to $\varepsilon_\infty$ [Fig. 3(b)]. In its turn, the carrier mobility depends on $\tau$, according to $\mu = \varepsilon_0/\tau$. The extrapolated $y$-intercept values of $\varepsilon_\infty$ for LBSO and ITO are equal to $\varepsilon_\infty$ obtained from the intercept in Fig. 2(b).

As summarized in Table III, $\mu_{opt}$ and $\mu_\infty$ for ITO thin films exhibit similar values ($28 \text{ cm}^2/\text{V s}$ and $33 \text{ cm}^2/\text{V s}$, respectively). This indicates that the carrier transport is not primarily hindered by scattering at grain boundaries in agreement with the discussion of a small potential barrier height for electron tunneling at enhanced carrier concentrations by Fujiwara and Kondo.33

Conversely, the optical and the drift mobility for the LBSO films differ significantly: $\mu_{opt}$ is found to be $337 \text{ cm}^2/\text{V s}$, which is almost an order of magnitude higher than the $\mu_\infty$ of $43 \text{ cm}^2/\text{V s}$. The estimated $\mu_{opt}$ is in good agreement with the calculations of the intrinsic LO-phonon-limited mobility of $3.0 \pm 0.6 \times 10^2 \text{ cm}^2/\text{V s}$ by Niedermeier et al.36 and demonstrated single crystal mobility of $\approx 320 \text{ cm}^2/\text{V s}$ by Kim et al.1 The trend of $\mu_{opt} > \mu_\infty$ implies that dislocation scattering is pronounced for the investigated LBSO films and may be further supported by columnar microstructures observed in the cross-sectional image by TEM [Fig. 4(b)]. Hence, improving the crystal quality should lead to the enhancement of LBSO conductivity. On the other hand, it proves that LBSO films even with moderate $\mu_\infty$ are suitable candidates for broadband transparent electrodes with high conductivity ($\sigma > 3 \times 10^3 \text{ S/cm}$) and for plasmonic applications due to intrinsically high $\varepsilon_\infty$ and $\mu_{opt}$.

### IV. CONCLUSIONS

In summary, epitaxially grown LBSO thin films on MgO substrates maintain high transparency in the NIR (>70% at 1200 nm), even for a high free carrier density of $5.0 \times 10^{20} \text{ cm}^{-3}$. We have demonstrated that the screened plasma wavelength of LBSO thin films exhibits a redshift with respect to the ITO thin film with the same carrier density due to the large high-frequency dielectric constant (4.40 against 3.85). This, together with the suppressed free carrier absorption due to the large optical mobility of LBSO, enables its enhanced infrared transparency. This work suggests that LBSO is a promising low-loss plasmonic material and In-free electrode for optoelectronic applications.

### SUPPLEMENTARY MATERIAL

See the supplementary material for a comparison of the microstructure and electrical properties of LBSO thin films grown on MgO (001) and STO (001) substrates, details about PDS measurements, fitting parameters for the Drude model, and elaborated derivation of the relation between real and imaginary parts of the dielectric constant.

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### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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