Optical properties of SrTiO$_3$ on silicon(100)

Yao Tian,$^1$ Carolina Adamo,$^2$ Darrell G. Schlom,$^2$ and Kenneth S. Burch$^{1, a}$

$^1$Department of Physics & Institute of Optical Sciences, University of Toronto, Ontario M5S 1A7, Canada
$^2$Department of Materials Science and Engineering, Cornell University, Ithaca, New York 14853, USA

(Received 19 August 2012; accepted 15 January 2013; published online 29 January 2013)

Epitaxial buffer layers enable the many functionalities found in perovskites to be integrated with silicon. However, epitaxial growth of SrTiO$_3$ on silicon is tricky and has so far only been achieved by molecular beam epitaxy. Nonetheless, previous investigations of these films were limited by the amorphous layer occurring at the interface. Through a combination of improved interface quality and an improved model, we report the optical properties of SrTiO$_3$ films on Si(100) investigated by spectroscopic ellipsometry. We find that the data are best described by a model with two different SrTiO$_3$ layers, potentially resulting from variations in the oxygen content.

For a bulk crystal, the Fresnel equations relate the ellipsometric parameters to the optical constants directly without any need for further analysis. However, for anisotropic crystals or layered materials there exists no analytical formula to directly extract the optical constants of the film from $\Psi$ and $\Delta$. In this case, measurements are performed at multiple angles of incidence, then the data are fit to extract the optical constants. Nonetheless, by measuring $\Psi$ and $\Delta$ at various angles of incidence, one can determine the thickness and optical constants of constituent members of a multilayered material. Additionally, all the measurements can be done non-invasively and non-destructively.

The optical constants of the silicon substrate are sensitive to its doping level. Thus, we took data on a silicon substrate from the same batch. Knowing the optical constants of the substrate allowed us to precisely model the SrTiO$_3$/Si system. The SrTiO$_3$ thin film was prepared by MBE via a kinetically controlled growth process. The native surface oxide of the silicon substrate was thermally removed in situ prior to film growth via a strontium-assisted deoxidation process. The details of the growth can be found elsewhere.$^4$ To prevent the interference from the substrate, the back surface of the silicon substrate is roughened intentionally.$^{10}$ Ellipsometric parameters of the SrTiO$_3$/Si film were measured from 0.75 eV to 5.5 eV with 0.05 eV resolution, except in the range 2.4 to 4.6 eV where it was measured with 0.001 eV to properly capture the critical points of Si and SrTiO$_3$. The measured $\Psi$ and $\Delta$ are shown in Fig. 1.

From Fig. 1, we can see there are several features in the experimental data. The feature around 1.4 eV results from interference. The peak at 3.4 eV is the natural result of the $E_1$ critical point of the silicon substrate.$^{11}$ There are also two small features around 3.7 eV and 4.7 eV. The former feature is due to the direct gap of the SrTiO$_3$ film and the latter one is due to the higher energy critical point of the SrTiO$_3$ film at 4.7 eV.

To properly extract the optical constants of SrTiO$_3$ as well as determine the homogeneity of the film, the dielectric function ($\varepsilon(E) = \varepsilon_1(E) + i\varepsilon_2(E)$) of the SrTiO$_3$ film is

\[
\frac{r_p}{r_s} = \tan \Psi e^{i\Delta}.
\]
defined using a sum of Tauc-Lorentz (TL) oscillators that are widely applied to interpret amorphous materials and thin films.\textsuperscript{12} The imaginary part of the dielectric function in this model can be written as:\textsuperscript{13}

\[
\epsilon_2(E) = \frac{A_L E_0 C (E - E_g)^2}{(E^2 - E_0^2)^2 + C^2 E^2} \Theta(E - E_g),
\]

where $E_0$ is the peak transition energy, $A_L$ is the oscillator strength, $E_g$ is the band gap, $C$ is the broadening term, and $\Theta$ is the step function. The real part is obtained from a Kramers-Kronig transformation. From the formula, we see that the imaginary part is the step function. The real part is multiplied by a standard Lorentz oscillator.

From the fit, the thickness of the top (second) layer is 0.35 nm, giving a total thickness of the SrTiO\textsubscript{3} film of 100.5 ± 0.2 nm. This is in good agreement with the nominal thickness determined from in-situ RHEED during the MBE growth of the film.\textsuperscript{15} We present the dielectric function of both layers in Fig. 3. Data for the second layer is only displayed below 3.76 eV since this is the energy where the penetration depth is smaller than the thickness of the top layer (see Fig. 4). As a comparison, the bulk SrTiO\textsubscript{3} is also measured with spectroscopic ellipsometry with resolution 0.05 eV from 0.75 eV to 5.5 eV. The optical constants of the bulk are also extracted by the sum of TL oscillators. A 3.0 ± 0.1 nm-thick Bruggeman effective medium approximation layer was added to account for the surface roughness.\textsuperscript{17} The extracted optical constants are in good agreement with the literatures\textsuperscript{17,18} and plotted in Fig. 3 as well. From Fig. 3, we can see that the onsets of $\epsilon_2(E)$ shift from the bulk value indicating the band gaps of both layers shift to lower energy. The reasons for the shifts will be discussed later.

In Fig. 3(c), we plot $(\epsilon_2(E) \times E^2)^2$, since in the vicinity of direct gap, $\epsilon_2(E)$ has the energy dependence

\[
\begin{align*}
\psi_1(E) = \frac{A_1 E_1 C(E - E_g)^2}{(E^2 - E_1^2)^2 + C^2 E^2} & \Theta(E - E_g), \\
\psi_2(E) = \frac{A_2 E_2 C(E - E_g)^2}{(E^2 - E_2^2)^2 + C^2 E^2} & \Theta(E - E_g), \\
\psi_3(E) = \frac{A_3 E_3 C(E - E_g)^2}{(E^2 - E_3^2)^2 + C^2 E^2} & \Theta(E - E_g),
\end{align*}
\]

where $E_1$, $E_2$, $E_3$ are the peak transition energies, $A_1$, $A_2$, $A_3$ are the oscillator strengths, $C$ is the broadening term, and $\Theta$ is the step function. The real part is obtained from a Kramers-Kronig transformation. From the formula, we see that the onsets of $\psi_i(E)$ shift from the bulk value indicating the band gaps of both layers shift to lower energy. The reasons for the shifts will be discussed later.
and the photon energy. As expected, we observe a linear function of energy whose intercept gives a direct gap of 3.77 ± 0.03 eV close to the bulk gap value of 3.78 eV determined by the same method. Since there is only limited data for the second layer, the direct gap cannot be determined. Also, at the edge of the indirect band gap, \( \epsilon_2(E) \) has the energy dependence given by \( \epsilon_2(E) \propto (E - E_g - E_p)^2 \), where \( E_p \) is the energy of phonon mode. Thus, the optical indirect gap of both layers is determined by a linear fit between \( \epsilon_2(E) \).

In summary, spectroscopic ellipsometry has been performed on SrTiO\(_3\) thin film grown on Si (100). The data are best described by a two layer model. The dielectric function of the second layer changes significantly from bulk. We ascribe this to the effect of oxygen deficiency at the beginning of the growth.

Work at the University of Toronto was supported by NSERC, CFI, and ORF.