

Supplementary Material

Growth, Structure and Electronic Properties of Epitaxial Bismuth Telluride Topological Insulator Films on BaF₂ (111) Substrates

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(Dated: January 9, 2013)

PACS numbers:

OPTICAL PROPERTIES

The optical constants of the Bi_2Te_3 and BiTe epilayers were determined by combining spectroscopic ellipsometry and reflectivity measurements. For this purpose a mid-infrared ellipsometer attached to a Bruker IFS55 EQUINOX Fourier spectrometer as well as a Woollam M-2000 and Jobin Yvon UVISEL ellipsometer for the near-infrared, visible and ultraviolet range were employed. Reflectance measurements in the 0.01–0.2 eV range were done using a Bruker IFS 66v/S spectrometer and far-infrared ellipsometric measurements at the NSLS synchrotron in Brookhaven. Figure 1(a) presents the near-normal incidence infrared reflectivity of the $\text{Bi}_2\text{Te}_3/\text{BaF}_2$ sample (solid line). Since the layer is transparent in this energy range, the spectrum exhibits also features from the substrate – for example the structure near 0.02 eV is due to the substrate phonon. The sequence of minima and maxima at higher energies corresponds to the interference in the layer. We have analyzed the spectrum with the standard model of coherent interferences within a layer on a substrate [1] assuming the dielectric function of the layer in the Drude-Lorentz form

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_{\text{pl}}^2}{\omega(\omega + i\omega\gamma)} + \sum_k \frac{\Omega_{\text{pl},k}^2}{\Omega_{0,k}^2 - \omega^2 - i\omega\Gamma_k} \quad (1)$$

where ϵ_∞ stands for the electronic interband contribution, ω_{pl} and γ denotes the plasma frequency and broadening of the Drude term and $\Omega_{\text{pl},k}$, $\Omega_{0,k}$ and Γ_k the plasma frequency, frequency and broadening of the Lorentz terms, respectively. The dielectric function of the BaF_2 was determined by fitting the reflectivity of a bare substrate in the far-infrared range assuming the 3-term Lorentz model similar to Eq. (1), from which we obtain in units of cm^{-1} except for ϵ_∞ : $\epsilon_\infty = 2.16$, $\Omega_{0,1} = 186$, $\Omega_{\text{pl},1} = 402$, $\Gamma_1 = 6.1$, $\Omega_{0,2} = 270$, $\Omega_{\text{pl},2} = 57$, $\Gamma_2 = 51$, $\Omega_{0,3} = 330$, $\Omega_{\text{pl},3} = 52$, $\Gamma_3 = 53$ at $T = 300$ K

The model spectrum of the whole layer–substrate structure presented as dashed line in Figure 1(a) fits very well to the measured data up to the energy of about 0.1 eV. At higher energies, the substrate is transparent and the quantitative analysis of the spectra is complicated due to the incoherent reflections from the backside of the sample. The model dielectric function involved also a Lorentz oscillator at $\Omega_0 = 5.6$ meV corresponding to the so-called alpha phonon [2], which lies outside the frequency range of our spectrometer. The resulting best-fit parameters for the epilayer are: $\epsilon_\infty = 45 \pm 2$, $\omega_{\text{pl}} = (0.33 \pm 0.02)$ eV, $\gamma = (0.07 \pm 0.01)$ eV. In addition, we obtained the thickness of the layer as $d = (830 \pm 40)$ nm.

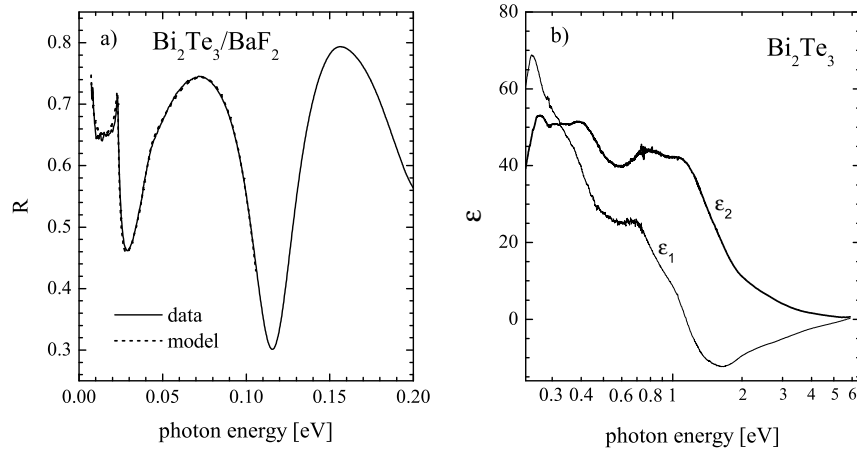


FIG. 1: (a) Far-infrared reflectivity of the $\text{Bi}_2\text{Te}_3/\text{BaF}_2$ epilayer (solid line) and a model spectrum (dashed line) for electric vector perpendicular to the c -axis, $E \perp c$. (b) Real and imaginary part of the dielectric function of Bi_2Te_3 obtained from ellipsometric measurements up to 5 eV.

Since the reflectivity was measured at near-normal incidence, the retrieved dielectric function corresponds to the in-plane direction, $\vec{E} \perp c$. The derived dielectric constants were included in the dielectric functions depicted in Fig. 7 of the manuscript.

Figure 1(b) displays the real and imaginary part of the dielectric function of the Bi_2Te_3 layer above 0.2 eV, where the layer is opaque. The spectra were obtained from the ellipsometric measurements at the angle of incidence of 70 degrees (0.6–6) eV, and at 75 degrees (0.2–0.6) eV. The displayed (pseudo)dielectric function was obtained assuming that the layer is isotropic. As usual, the pseudo-dielectric function very close to the in-plane contribution. This is particularly true for spectra acquired at low angles of incidence, and for large values of the index of refraction, with the in-plane orientation of the electric vector inside the layer. The pseudo-dielectric function measured in the 0.6–6 eV range did not exhibit any detectable angular dependence. Consequently, it represents the in-plane response, $\vec{E} \perp c$. At these photon energies the reported anisotropy of bulk Bi_2Te_3 is small [3]. The characteristic energies of the spectral features seen in Fig. 1(b) are in good agreement with those of bulk reported in Ref. [3]. However, the magnitude of our spectra is by a factor 1.5-2 smaller and our value $\epsilon_\infty = 45 \pm 2$ is smaller by a factor of ~ 2 than that obtained in Ref. [2]. The discrepancy is probably caused by errors in the absolute values of the measured reflectivity in the

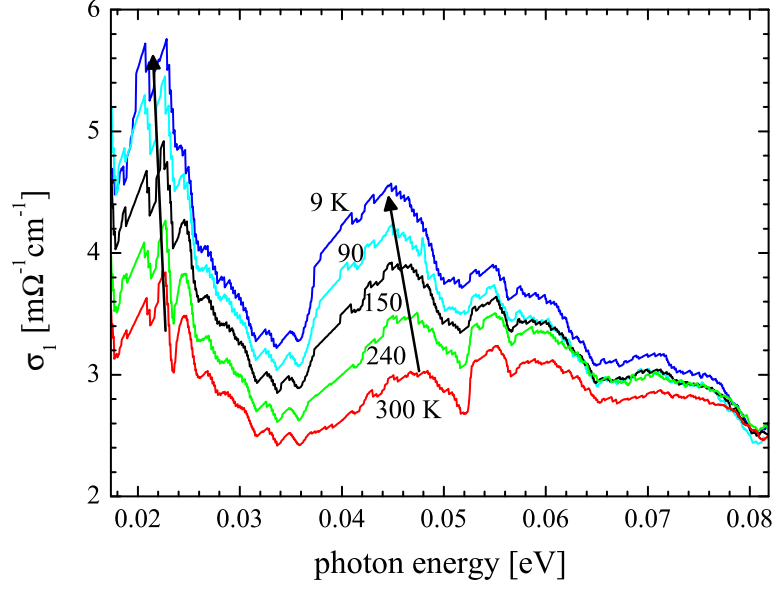


FIG. 2: Temperature dependence of the real part of conductivity of the BiTe epilayer in the far-infrared regime determined by ellipsometry at the NSLS synchrotron in Brookhaven.

cited works, or by the errors induced by extrapolations necessary for the Kramers-Kronig analysis.

As indicated in Fig.7 of the manuscript(a), the infrared conductivity of BiTe exhibits two bands near 0.02 and 0.05 eV that are superimposed on the Drude contribution. In order to inspect these bands in a greater detail, we have performed low-temperature far-infrared ellipsometric measurements at the NSLS synchrotron in Brookhaven. Figure 2 displays the resulting real part of conductivity from 9 to 300 K. The sharper structures in the spectra are artefacts due to the inhomogeneity of the polarizer. Both bands at around 0.02 and 0.05 eV gradually sharpen and soften with decreasing temperature.

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