

Observation of charge dressing in Bi_4Te_3 topological insulator thin films: A terahertz and infrared spectroscopic study

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Bi_4Te_3 has been first realized as an element of the adaptive series $(\text{Bi}_2)_m(\text{Bi}_2\text{Te}_3)_n$ —deriving from stacking metallic Bi and topological insulator Bi_2Te_3 blocks—and studied for its promising thermoelectric properties. Recently, Bi_4Te_3 has also been found as a *dual* topological insulator hosting a complex band structure with three Dirac cones in k space. Here we report measurements of the optical conductivity of five Bi_4Te_3 thin films with thickness varying from 11 to 32 nm. This extensive study, based on terahertz time-domain spectroscopy and Fourier-transform infrared spectroscopy, provides a broadband optical conductivity. By employing a Kramers-Kronig constrained Drude-Lorentz model, the spectral contributions to the optical conductivity of each sample have been singled out. In contrast to an ideal topological insulator—with perfectly insulating bulk—in Bi_4Te_3 far-infrared and mid-infrared bands indicate the presence of localized massive carriers, which dope the bulk system, as also observed in chemically compensated single crystals of other bismuth-based topological insulators. In addition, the localized bulk charges strongly renormalize the electronic mass. The charge dressing enhances as the thickness increases, indicating a trivial metallic behavior of Bi_4Te_3 thin films. This study elucidates the inner relationship between the optical properties of a *dual* topological insulator like Bi_4Te_3 and its complex band structure, since the detection of the topological massless carriers by optical measurements is prevented by the occurrence of novel and unusual conductive properties.

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I. INTRODUCTION

With the beginning of materials' crystalline growth in the form of thin films on a substrate during the 1980s, made possible through the development of techniques like physical vapor deposition, molecular beam epitaxy (MBE), and pulsed laser deposition, enormous advances have been made in condensed matter physics research. Thin films with a thickness in the nanometer scale opened the way to the miniaturization of devices based on materials, whose physical and chemical properties in the form of films may differ significantly from those of single crystals or polycrystals.

Particularly intriguing is the case of two-dimensional (2D) transition metal dichalcogenides and topological insulators (TIs) [1,2], largely studied in the past few decades for plenty of applications ranging from photonics and optoelectronics to plasmonics [3–9]. Indeed, the conductive behavior of those materials has been investigated when formed as thin films

and many examples of remarkable differences with respect to single crystals have been found. To mention just an example, the benchmark topological insulator Bi_2Se_3 shows a not negligible bulk conductivity in single crystals, deviating from the ideal case of an insulating bulk. Indeed, this bulk contribution hinders the evidence of the topological surface state conductivity, at variance with the case of thin films, where the free electron Drude conductivity is independent of the film thickness, hence proving the 2D character [10,11]. Generally, the conductivity reduces overall in thin films with respect to single crystals, where several conduction channels can be activated due to the presence of bulk carriers, impurities, lattice defects, and polarons. Those particles scatter with electrons, thereby adding different scattering types to the pure electron-electron one.

Bi_4Te_3 has been studied for its promising thermoelectric properties [12] and recently found to be a topological insulator [13]. It can be assembled as an element of the series $(\text{Bi}_2)_m(\text{Bi}_2\text{Te}_3)_n$, deriving from stacking hexagonal metallic Bi bilayers and topological insulator Bi_2Te_3 quintuple layers blocks. In Ref. [14], polycrystalline Bi_4Te_3 has been grown and characterized by x-ray diffraction and thermopower measurements. Here we address the optical conductivity of Bi_4Te_3 thin films. The superlattice $(\text{Bi}_2)_m(\text{Bi}_2\text{Te}_3)_n$ gives a layered

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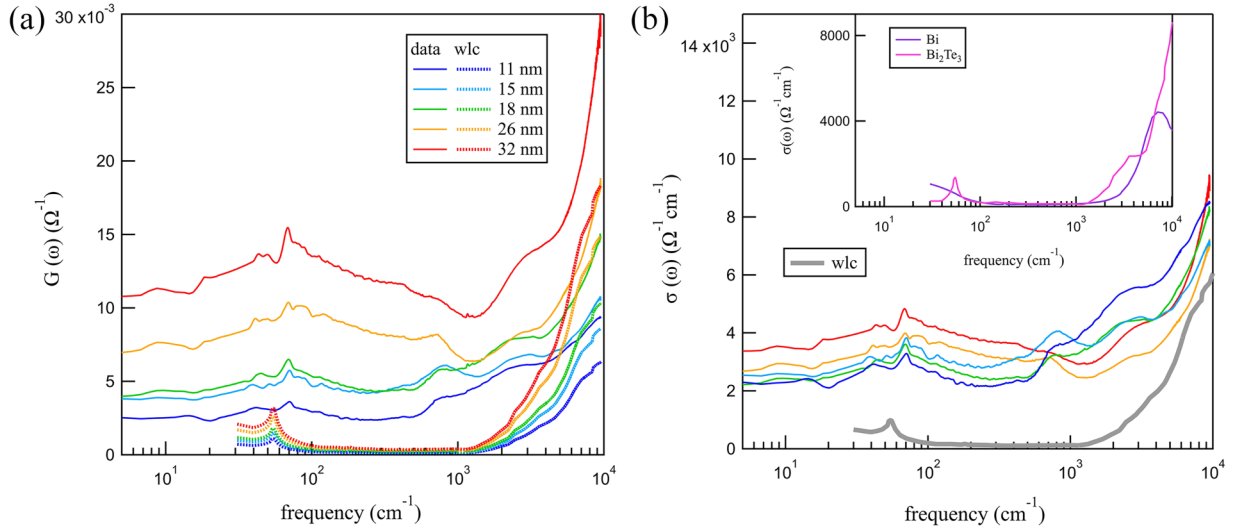


FIG. 1. (a) $G(\omega)$ of the five samples compared to the same quantity for the weighted linear combination (wlc), as extracted by $\sigma_{\text{wlc}}(\omega)$ multiplied by the relative thickness. (b) Corresponding optical conductivities for all samples. Here the gray curve is $\sigma_{\text{wlc}}(\omega)$. Inset: Optical conductivities of Bi and Bi_2Te_3 , as taken from Refs. [22,23] and used to calculate the wlc, according to Eq. (1).

Bi_4Te_3 when $m = n = 3$ [14]. We demonstrate that MBE-grown Bi_4Te_3 films' physical properties differ from those associated with the bare sum of the stacking blocks' properties, unveiling that the growth procedure may introduce interfacial effects with a variation of the low-energy electrodynamics. In particular, we show here that the conductivity enhancement in our thin films is due to localized dressed charges, possibly ascribable to the presence of impurities and/or defects, like Te vacancies [15].

II. METHODS AND MEASUREMENTS

The terahertz (THz) and infrared (IR) conductivity of five Bi_4Te_3 thin films with thicknesses of 11, 15, 18, 26, and 32 nm, grown by MBE on a high-resistivity undoped silicon substrate, have been investigated. The transmittance $T(\omega)$, where ω is the frequency, has been measured for each sample at room temperature in the 5–9500 cm^{-1} frequency range by using the bare silicon substrate as a reference.

The THz transmittance was measured by means of a THz time-domain spectroscopy (TDS) setup, based on photoconductive antennas (BATOP GmbH), available at the TeraFERMI offline facility at FERMI, Elettra Sincrotrone Trieste, in Italy [16,17]. This spectrometer allowed us to cover a frequency range between 0.15 and 2 THz with nitrogen purging (1 THz = 33 cm^{-1}).

The IR transmittance was measured by a Fourier-transform infrared (FTIR) spectrometer available at the SISSI-Material Science beamline at Elettra [18]. The beamline is equipped with a Bruker Vertex 70v Michelson interferometer coupled with a Bruker Hyperion 2000 infrared microscope. By changing the internal sources, the beam splitter, and the detector in the setup, we were able to cover an infrared spectral range of 60–9500 cm^{-1} . The transmittances measured by TDS and FTIR techniques have been merged together to obtain $T(\omega)$ in the whole frequency range for each sample. By means of a Kramers-Kronig consistent analysis [19], we extracted the real part of the complex conductivity, i.e., the optical

conductivity $\sigma(\omega)$. The optical conductivity is related to the real part of sheet conductance $G(\omega)$ by the relation $G(\omega) = \sigma(\omega)d$, with d being the thickness of the film.

In Fig. 1(a) we report $G(\omega)$ for all the samples, while Fig. 1(b) shows their $\sigma(\omega)$.

The whole $G(\omega)$, or at least its DC value, increases quasi-monotonically with thickness, like in the cases of trivial semiconducting and metallic samples.

Bi_4Te_3 has been found in the literature to be an element of the infinitely adaptive series $(\text{Bi}_2)_m(\text{Bi}_2\text{Te}_3)_n$, made of alternating metallic Bi bilayers and topological insulator Bi_2Te_3 quintuple layers. When $m = n = 3$, the bismuth-tellurium phase corresponds to the Bi_4Te_3 material [14]. The series can be seen as a superlattice with repetition of building blocks. Following the example of Ref. [20], we can introduce an average optical conductivity $\sigma_{\text{wlc}}(\omega)$, defined as a weighted linear combination of the optical conductivity of the single constituents of a given series of $(\text{Bi}_2)_m(\text{Bi}_2\text{Te}_3)_n$, i.e.,

$$\sigma_{\text{wlc}}(\omega) = \frac{V_{\text{Bi}}\sigma_{\text{Bi}}(\omega) + V_{\text{Bi}_2\text{Te}_3}\sigma_{\text{Bi}_2\text{Te}_3}(\omega)}{V_{\text{Bi}} + V_{\text{Bi}_2\text{Te}_3}}, \quad (1)$$

where $\sigma_{\text{Bi}}(\omega)$ and $\sigma_{\text{Bi}_2\text{Te}_3}(\omega)$ are the optical conductivities of elemental bismuth and of Bi_2Te_3 , respectively, and V_{Bi} , $V_{\text{Bi}_2\text{Te}_3}$ are their corresponding unit-cell volumes. The use of those weights accounts for the differences in thickness and volume of the two elements of the series. The values of the unit-cell volumes have been taken from Ref. [14]. The comparison between the measured conductivity of our five Bi_4Te_3 samples and the calculated weighted linear combination of the conductivity of the two blocks allows us to single out the features directly induced by the samples' growth technique, possibly related to interfacial effects, like charge diffusion and charge accumulation (puddles [21]), strain, and lattice mismatch. These interfacial effects introduce most likely a redistribution of spectral weight as it was observed in the case of $(\text{LaNiO}_3)_n/(\text{LaMnO}_3)_2$ superlattices. In Fig. 1(b), we show the $\sigma_{\text{wlc}}(\omega)$ (gray curve), calculated by using Eq. (1) and the optical conductivities of elemental bismuth and Bi_2Te_3 , found

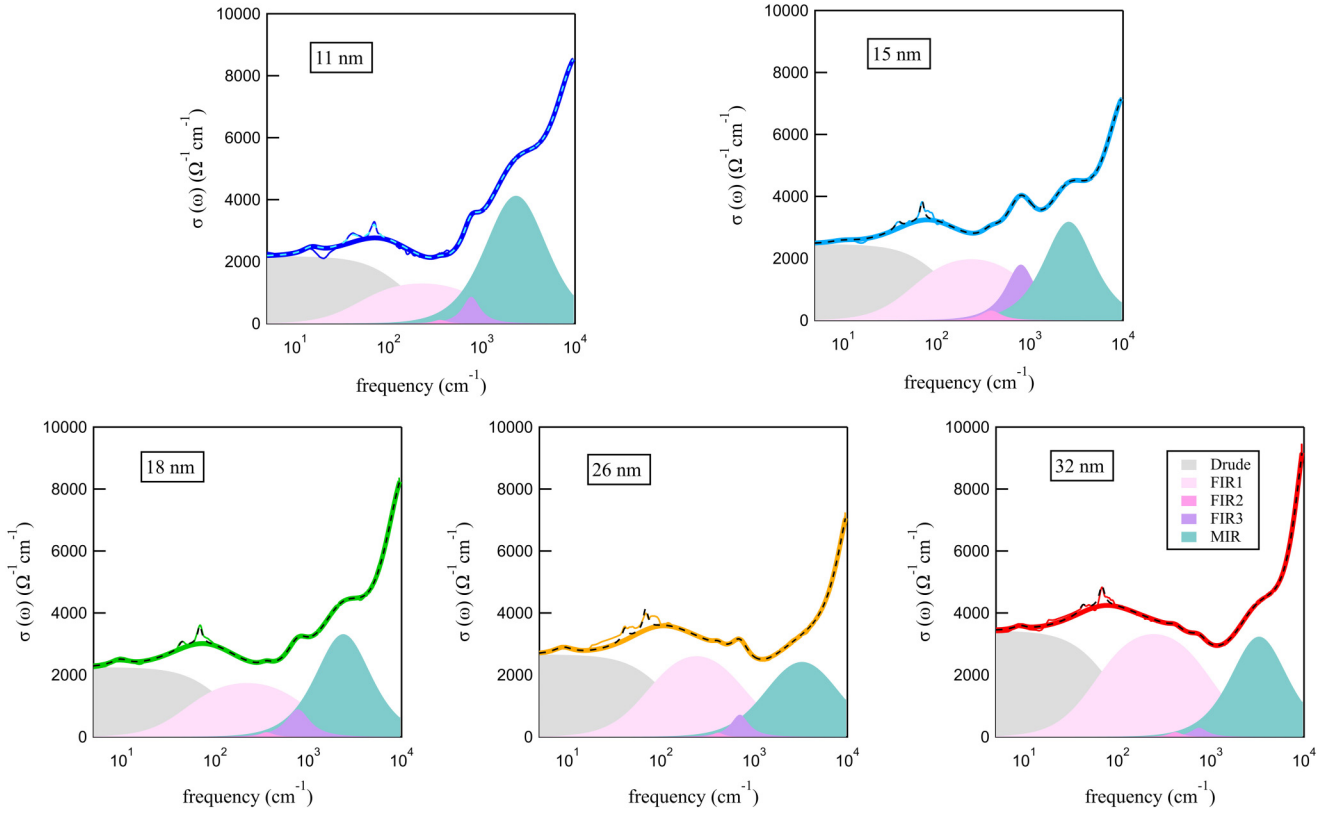


FIG. 2. Optical conductivities (solid curves), fits (dashed curves), and phonon-subtracted optical conductivities (solid bold curves) of the five samples with the intragap Drude-Lorentz fitting components.

in the literature in the same frequency range in Refs. [22] and [23], respectively. Those quantities are also reported in the inset of the same panel. From the weighted linear combination of the single blocks' conductivities, one can easily calculate the related sheet conductances for the corresponding film of thickness d by the formula $G_{\text{wlc}}(\omega) = \sigma_{\text{wlc}}(\omega)d$, which intrinsically makes the calculated sheet conductance increase monotonically with the thickness.

III. DISCUSSION

Bi_4Te_3 is a *dual* topological insulator, being a strong topological insulator with three Dirac cones, and a topological crystalline insulator, where the surface states are protected by crystal symmetries besides the time-reversal symmetry [13,24]. The occurrence of three Dirac cones in k space makes the band structure of Bi_4Te_3 well different from those in Bi_2Se_3 and Bi_2Te_3 , which have only one Dirac cone. Indeed, Bi_2Se_3 thin films have a sheet conductance in which the Drude term remains constant as a function of the thickness, unveiling the presence of surface Dirac carriers as the only contribution to the conductivity [10]. The presence of three Dirac cones in Bi_4Te_3 could instead allow multichannel electronic transport, mirroring novel conductive properties. The comparison with the combined conductivities of the bare stacking of Bi_2 and the well-known topological insulator Bi_2Te_3 [25] may help to single out the optical properties of a real Bi_4Te_3 . Here, indeed, the type of surface termination, either in Bi_2 or Bi_2Te_3 , might play a role in its electrodynamics.

In order to better understand the electrodynamic behavior of our samples, we analyze the data with a Drude-Lorentz model [26], which allows us to distinguish the spectral components of the optical conductivity and to finally understand the origin of its overall enhancement with respect to that of the model, i.e., the stack of Bi_2 and Bi_2Te_3 layers.

In Fig. 2, we show the five optical conductivities of Fig. 1 together with the corresponding Drude-Lorentz fits. The fitting parameters are three for each Drude or Lorentzian component, i.e., an oscillator strength S_i , a central frequency $\omega_{i,0}$, and a scattering rate γ_i . The index i varies from 0 to 7, where each number corresponds to a specific Drude or Lorentzian component (see Table I). Being the Drude component, a Lorentzian centered at 0 frequency, we refer to its oscillator strength S_0 as ω_p (plasma frequency) and to its scattering rate γ_0 as γ_{Drude} . As reported in Table I, the indexes $i = 1-7$ indicate the four Lorentzian components, namely, the bands FIR1, FIR2, FIR3 (far-infrared bands, centered between 100 and 1000 cm^{-1}), and MIR (mid-infrared band, centered between 1000 and 5000 cm^{-1}), the two peaks falling in the range of 50–70 cm^{-1} and an intergap band with $\omega_0 > 10\,000\text{ cm}^{-1}$, which corresponds to our upper limit of the investigated frequency range. The two peaks in the 50–70 cm^{-1} range can be assigned to two phonons, resulting from vibrational modes of Bi and Te atoms, as occurs in Bi_2Te_3 where a clear phononic mode is visible at 55 cm^{-1} [27] [see the inset in Fig. 1(b)]. In Fig. 2 we report also, together with the intragap Drude and Lorentzian components, the conductivities reconstructed from the fitting parameters by excluding the two phonon

TABLE I. The Drude-Lorentz components together with the corresponding fitting parameters are reported here for all the samples. The components with $i = 1-4$ are related to intragap bands, while those with $i = 5$ and 6 are the two phonon modes not discussed in this work. The last component is the intergap band. All the values are in cm^{-1} .

i		11 nm			15 nm			18 nm			26 nm			32 nm		
		S_i	$\omega_{i,0}$	γ_i	S_i	$\omega_{i,0}$	γ_i	S_i	$\omega_{i,0}$	γ_i	S_i	$\omega_{i,0}$	γ_i	S_i	$\omega_{i,0}$	γ_i
0	Drude	ω_p		γ_{Drude}	ω_p		γ_{Drude}	ω_p		γ_{Drude}	ω_p		γ_{Drude}	ω_p		γ_{Drude}
		4472	0	153	4300	0	125	4149	0	128	4400	0	121	4213	0	86
1	FIR1	9287	229	1112	11203	237	1061	10342	221	1026	11574	241	857	14605	245	1072
2	FIR2	1035	358	151	2115	386	234	1156	357	147	1042	411	131	1256	418	170
3	FIR3	4588	772	406	8960	804	744	5456	774	559	4050	705	382	2572	750	375
4	MIR	34475	2332	4803	28317	2620	4202	29302	2374	4317	34897	3264	8389	34542	3299	6149
5	ph1	414	41	12	309	39	7	300	44	7	390	41	7	313	43	7
6	ph2	575	70	13	545	70	8	565	69	9	641	69	10	651	68	11
7	Inter gap	98808	11676	20345	87058	11667	18136	93310	11850	17575	82296	12816	15500	84489	12043	12163

contributions (solid bold curves), the investigation of which is beyond the aim of this work. Observing the Drude-Lorentz components of the five phonon-subtracted conductivities one can assess that the main spectral contribution comes from the Drude, the FIR1, and the MIR bands. Performing a Drude-Lorentz fit to the weighted linear combination conductivity gives us a Drude term and an intergap band. No FIR and/or MIR bands contribute to the weighted linear combination conductivity. This qualitative difference guided us into a spectral weight analysis, which significantly helps us to better understand the charge contribution to the conductivity of the samples. Indeed, the spectral weight (SW) is defined as

$$\text{SW}(\Omega) = \int_0^{\Omega} \sigma(\omega) d\omega, \quad (2)$$

where Ω is a frequency cutoff, which can be arbitrarily chosen. The frequency-dependent SW of each sample is reported in Fig. 3, together with the SW calculated for the σ_{wlc} with the same cutoff corresponding to the limit of the accessed spectral range (i.e., 9500 cm^{-1}). The main difference between the five measured SWs and the estimated SW for a linear combination of stacking layers lies in the range between 100 and 4000 cm^{-1} , which corresponds to the FIR1 and MIR bands' maximum contributions.

From the spectral weight one can calculate the number of electrons per unit cell. This number depends on the electronic mass m_e , which changes in a solid with respect to that of a free electron, due to many-body effects, in an *effective* mass m^* . To the best of our knowledge, the effective mass in Bi_4Te_3 is not known, so we can define an *effective* number of electrons N_{eff} that corresponds to the number of electrons which the system would have if its $m^* = m_e$, i.e. [26,28,29],

$$N_{\text{eff}}(\Omega) = \frac{2m_e}{\pi e^2} \text{SW}(\Omega). \quad (3)$$

We show in Fig. 4(a) the values of N_{eff} related only to the Drude free carriers (blue dots), calculated from the Drude SW, compared with N_{eff} calculated from the sum of the SWs of the FIR1 and MIR bands (green diamonds), which lie in the spectral range where the measured SW differs from the expected one (Fig. 3). We highlight here that the SW of the FIR1 and MIR bands can be simply calculated as $S_{1,4}^2$ (see Table I), which mathematically corresponds to Eq. (2)

integrated over the FIR1 and MIR bandwidths. Here the choice of considering only the first band (FIR1) among the three FIR ones is given by the fact that FIR2 and FIR3 have negligible spectral weights with respect to FIR1, although phenomenologically necessary for the best fit of the optical conductivity. N_{eff} deriving from FIR1 and MIR bands' SW provides the amount of localized charges, due to the presence of impurities and/or defects in the system. The difference between the two quantities in Fig. 4(a), about 2 orders of magnitude, is a clear indication that our samples are far from perfect stoichiometry and chemical compensation, most likely due to the presence of Te vacancies, as was found in Ref. [15]. The localized charges may be due even to the additional presence of impurities. In this case, the low-frequency absorption bands (FIR1 and MIR) of the optical conductivity correspond to the hydrogenlike $1s \rightarrow np$ transitions of isolated impurities

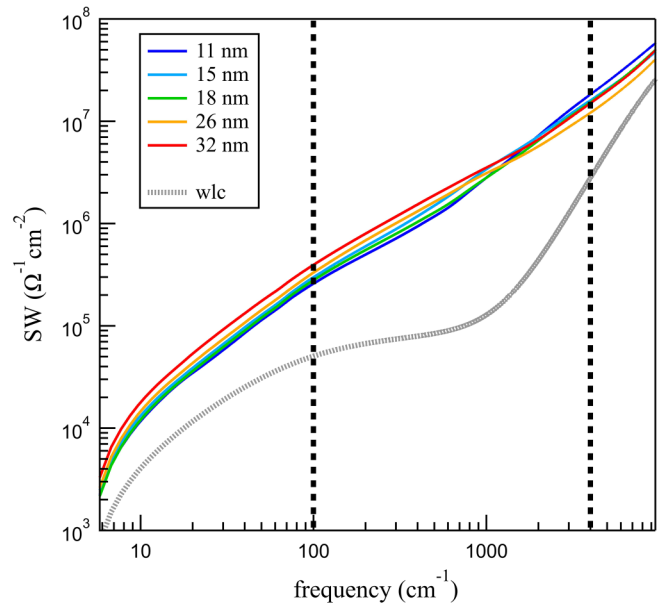


FIG. 3. Spectral weight of the five samples compared to that of the weighted linear combination (wlc). The spectral range between the two vertical dashed lines highlights the region in which the difference between the data and the calculated SW from the weighted linear combination is higher.

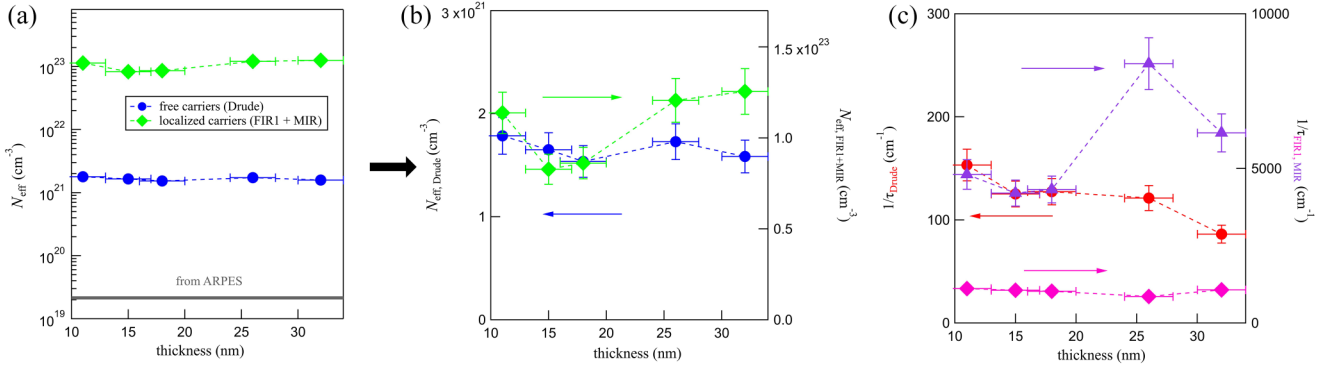


FIG. 4. (a) N_{eff} calculated from Drude SW (free carriers) and from the sum of SWs from FIR1 and MIR bands (localized carriers) vs thickness. The gray line represent a volume carrier concentration which accounts only for the topological massless surface Dirac electrons (see main text). (b) Same thickness-dependent quantities of panel (a) but reported on different scales: N_{eff} of free carriers (left scale) and N_{eff} of localized carriers (right scale). (c) Drude scattering rate (left scale) compared with the FIR1 and MIR scattering rates (right scale) vs thickness.

or, at higher frequencies, to the transitions from the impurity bound states to the continuum [11]. Those FIR and MIR bands have been already observed in Bi₂Se₃ and Bi-based TIs single crystals with increasing chemical compensation. Their total spectral weight was found to be several order of magnitudes higher than that of Bi₂Se₃ thin films, where the SW accounts only for Drude surface topological carriers [11].

In Fig. 4(a), the gray line represents the volume carrier density that the Bi₄Te₃ samples would have if they had only the Dirac massless carriers of the topological surface state with no contribution of massive bulk charges, i.e., in the ideal case of perfect insulating bulks. This volume carrier density, corresponding to $2.14 \times 10^{19} \text{ cm}^{-3}$, has been calculated by the carrier concentration of topological surface found by angle-resolved photoemission spectroscopy [15,24]. This quantity is obviously independent of the thickness of the samples.

In Fig. 4(b) we better compared the same quantities (blue and green symbols) of Fig. 4(a). We observe that the number of localized dressed charges (right scale) slightly increases with the thickness, while the free carriers' contribution (left scale) remains almost constant.

In order to provide further evidence of the presence of localized electrons in thicker samples, in Fig. 4(c) the Drude (free carriers) scattering rate is compared with the FIR1 and MIR scattering rates, namely, γ_1 and γ_4 (as reported in Table I), which account for localized carriers. Here we use the Eq. (1) $1/\tau_{\text{Drude},1,4} = \gamma_{\text{Drude},1,4}$, where τ is the carrier lifetime. While γ_1 remains almost constant with respect to the thickness, the MIR scattering rate increases with sample thickness higher than 18 nm. This behavior may signify that the localized charges, likely related to Te vacancies, corresponding to the MIR contribution to the conductivity, are the most responsible for the mass renormalization, at least as the samples are thicker.

Figure 5 shows the DC resistivity calculated from our optical data. Indeed

$$\rho_{\text{DC,optical}} = \frac{1}{\sigma_{\text{DC,optical}}} = \frac{4\pi\gamma_{\text{Drude}}}{\omega_p^2}. \quad (4)$$

The optical DC resistivity decreases with thickness. This phenomenologically confirms a tendency towards a more metallic behavior for thicker samples, for which the number of bulk carriers (whatever is their origin) increases as well.

The optical DC resistivities of Bi₄Te₃ films are also similar to those found by transport measurements in single crystals of other topological insulators, like Bi₂Te₃ and Bi₂Se₃ [30].

IV. CONCLUSIONS

In conclusion, we addressed the optical properties of MBE-grown Bi₄Te₃ thin films through a terahertz and infrared spectroscopic study. We showed that their optical conductivity has a slight metallic behavior with a strong contribution of localized charges. The presence of localized bulk carriers leads to a renormalization of the electronic mass and hinders the evidence of the contribution purely coming from the topological surface carriers. This behavior follows very well that found on chemically compensated bismuth-based

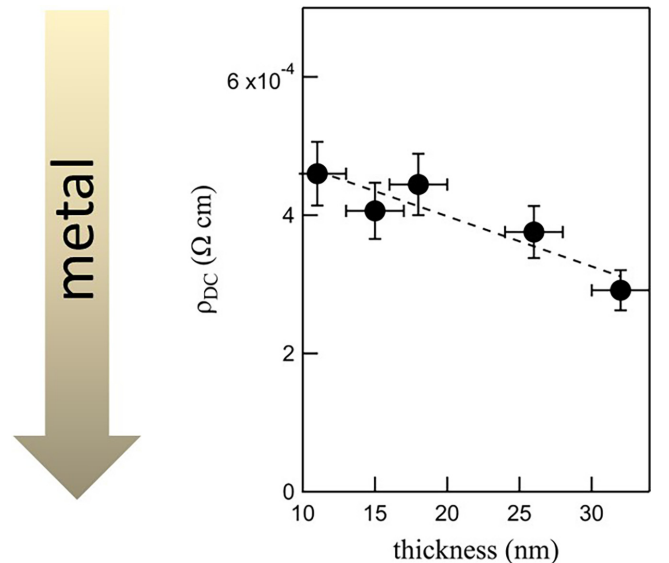


FIG. 5. Optical DC resistivity vs thickness. The dashed line is a guide to the eye.

topological insulator single crystals, where far- and mid-infrared bands in the optical conductivity indicate the presence of impurities and/or defects. Indeed, by analyzing the Drude-Lorentz spectral weights, we found that our samples behave differently from the ideal case of Bi_4Te_3 calculated as a weighted linear combination of alternating metallic Bi and topological insulator Bi_2Te_3 , in which impurities and/or defects due to the growth process are not considered. The difference is particularly effective in thicker samples where the bulk contribution to the conduction is higher, due to imperfect chemical compensation that does not occur in the ideal, perfectly stoichiometric case. We demonstrate here that, although the ideal topological insulator has an insulating bulk with charge contribution coming only from the massless Dirac electrons hosted in the topological surface state, the real case is much more complex and far from this situation. We assess that our finding elucidates the inner

relationship between the low-energy electrodynamics of a dual topological insulator like Bi_4Te_3 and its complex band structure, where multichannel electronic transport allows the occurrence of novel and unusual conductive properties. Our results will help researchers to better understand the future direction to undertake, in order to realize novel topological insulator thin films, not only with a simple growth recipe but also with potentially full suppression of bulk conduction contribution. Bulk-sensitive THz and IR spectroscopies, by unveiling the presence of renormalized massive charges, represent a powerful tool for that aim.

DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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