I. INTRODUCTION

The topological insulator MnBi$_2$Te$_4$ (MBT) is currently extensively investigated, since its intrinsic antiferromagnetic order could potentially induce interesting quantum mechanical effects such as quantum anomalous Hall effect (QAH) or axion insulator at low temperatures [1, 2]. The QAH, in particular, plays a crucial role for potential applications in quantum metrology and spintronics [3]. In recent studies, reduced dimensionality proved to realize exotic phenomena in van-der-Waals-type layered materials, which favours the research of thin films/monolayers of this material family [4]. MBT belongs to the group of ternary chalcogenides and its layered crystal structure with space group $R3m$ is built of septime layers, where MnTe blocks are intercalated in Bi$_2$Te$_3$ layers. By exchanging certain elements gradually – in this case bismuth (Bi) by antimony (Sb) – the electronic and magnetic properties can be manipulated [5-8]. Furthermore, an ideal type-II Weyl semimetal can be established through appropriate Sb doping in high magnetic fields [6]. The unit cell parameters are slightly changing in Mn(Bi$_{1-x}$Sb$_x$)$_2$Te$_4$ from $a = 4.33$ Å and $c = 40.93$ Å for $x = 0$ to $a = 4.25$ Å and $c = 40.87$ Å for $x = 1$ [6]. Fig.1(a) displays the unit cell structure of the mixed compound Mn(Bi$_{1-x}$Sb$_x$)$_2$Te$_4$, where the septime layer interaction is of van-der-Waals type [9]. In the antiferromagnetic phase, the out-of-plane Mn spins are aligned parallel within the $ab$ plane and anti-parallel along the $c$ axis. This $A$-type antiferromagnetic state in Mn(Bi$_{1-x}$Sb$_x$)$_2$Te$_4$ orders between $T_N = 25$ K ($x = 0$) and $T_N = 19$ K ($x = 1$) [6]. In case of a high Sb content, also ferrimagnetism has been observed, which possibly results from anti-site mixing of Mn and Sb ions [10].

According to magnetic susceptibility measurements, the effective magnetic moment $\mu_{eff} = 5.3 \mu_B$, originating from the Mn$^{2+}$ ions, is not changed by the Sb doping ratio [8]. However, the magnetic properties of the compounds are significantly affected, namely, the antiferromagnetic ordering temperature, the saturation moment, Weiss constant, and critical fields $H_c$ for the spin flip transition all decrease with increasing Sb substitution ratios. Here, we investigate the effect of magnetic ordering on the bulk electronic structure of Mn(Bi$_{1-x}$Sb$_x$)$_2$Te$_4$ with high Sb content $x = 0.93$ by temperature-dependent reflectivity measurements over a broad frequency range. We observe anomalies in the optical response across $T_N$ when the antiferromagnetic order sets, which suggests a coupling between the magnetic ordering and the electronic structure of the material.
and crosses the former valence bands resulting in mainly $p$-type free charge carriers, and the band gap is reduced compared to the pure compound [13, 14]. Due to the Fermi level crossing of the electronic bands, the MBST sample is expected to show signs of a metallic character with a high reflectivity at low energies, similar to the results for the pure compound [13, 14].

In this work, we investigate the optical excitations in Mn(Bi$_{0.07}$Sb$_{0.93}$)$_2$Te$_4$ by temperature-dependent reflectivity measurements over a broad frequency range, in order to characterize the changes in the electronic structure induced by the magnetic phase transition. The obtained results are compared to the recent reports [13, 14] on the undoped material MBT.

II. METHODS

Single crystals of Mn(Bi$_{0.07}$Sb$_{0.93}$)$_2$Te$_4$ were grown by the self-flux method as reported in Ref. [15]. The plate-like sample had a surface size of app. 0.6 x 0.8 mm and a thickness close to 100 µm. Magnetic susceptibility data have been collected from 1.8 K to 300 K using a superconducting quantum interference device (SQUID, Quantum Design) magnetometer MPMS. According to these measurements, the sample undergoes a magnetic phase transition at $\sim 20$ K, as expected for $x = 0.93$ [see Fig. 1(b)].

The magnetic field $H$ has been aligned parallel of the $ab$-plane, i.e., perpendicular to the antiferromagnetic ordering. For the reflectivity measurements at temperatures between 295 and 5 K, we have used a CryoVac Konti cryostat, which has been connected to a Bruker Hyperion infrared microscope and Bruker Vertex80v FTIR spectrometer. Half of the surface of the freshly cleaved sample was coated with a thin silver layer, which was used as reference for the calculation of the absolute reflectivity. The sample was glued to a sample holder within the cryostat and aligned perpendicular to the incoming beam. The measurements were performed from the far-infrared up to the visible range (100 to 20000 cm$^{-1}$). The measured spectra were extrapolated in the low- and high-frequency range with the help of literature values and volumetric data. Then, the optical functions were calculated through the Kramers-Kronig relations, using programs by David Tanner [10]. The optical spectra were fitted with the Drude-Lorentz model using the software RefFIT [17].

III. RESULTS AND DISCUSSION

The reflectivity spectra of MBST are shown in Fig. 3(a) for selected temperatures. The temperature steps have been decreased close to the phase transition temperature $T_N \approx 20$ K. The high reflectivity at low frequencies and the plasma edge near 1000 cm$^{-1}$ indicate the metallic character of the material. The bumps in the reflectivity spectra above $\sim$1000 cm$^{-1}$ are due to electronic transitions across the optical gap. With decreasing temperature, slight but significant changes can be observed in the low-energy range: During cooling from 295 to 50 K the plasma edge sharpens and the Drude spectral weight, which is associated with the free charge carriers, increases. This trend is, however, reversed for temperatures below 50 K. In the high-frequency range, the temperature-induced effects in the reflectivity spectrum appear to be weak, however, as we will see below, are significant. For further illustration of the temperature-induced changes, we plot in Fig. 3(c) the reflectivity values at 500 cm$^{-1}$, i.e., within the free charge carrier range, as a function of temperature. During cooling, one observes a steady increase of this value down to 40 K, followed by slight deviations down to 20 K and a decrease...
At 18 K one observes a sudden jump to higher values, indicating cooling, an increase of the optical gap occurs down to 75 K, followed by a moderate decrease down to 20 K. At 18 K one observes a sudden jump to higher values, followed by a further steady decrease. This jump might be caused by the magnetic phase transition occurring at this temperature in our sample, since a blue shift of the energy gap can be induced by the onset of magnetic ordering as it has been reported for MnTe \[20\]. We assume, that also the optical gap can be affected by this blue shift, which should be visible in our results.

For a quantitative analysis, we performed a simultaneous fitting of the reflectivity and optical conductivity spectra with the Drude-Lorentz model \[see Fig.3(e)\], where we used the same number of oscillators like for the undoped compound MnBi\(_2\)Te\(_4\) \[13\]. Two Drude terms have been implemented to characterize the response of the free charge carriers, which should be mainly p-type according to Chen et al. \[8\]. Concerning the spectral weight, we have used one strong and one weak Drude term, as shown in Fig.3(e). This can be justified, like for the undoped compound \[13, 14\], by the free carrier contributions of two different conduction bands. In Fig.3 the band structure of MBST is sketched and the crossing of the reflectivity at very low temperatures, which might already be a hint for the magnetic ordering.

The optical conductivity spectrum \(\sigma_1\), which was obtained from the measured reflectivity via KK analysis, is depicted in Fig.3(b). The excitations of the free charge carriers are visible from 0 up to about 2000 cm\(^{-1}\), where we detected the plasma minimum, separating the intraband from the interband transition range. The latter starts from about 400 cm\(^{-1}\), where a steep linear increase in \(\sigma_1\) is visible. This region is followed by a broad maximum at \(\sim 12000\) cm\(^{-1}\). Thus, the Drude spectral weight is rather small compared to the spectral weight of the optical transitions at higher energies, which indicates a relatively weak metallic behavior with \(\sigma_{dc}\) values smaller than 1000 Ω\(^{-1}\)cm\(^{-1}\). The shape of \(\sigma_1\) is comparable to the one we measured for the undoped compound MBT, where the spectral weight of free charge carriers is slightly higher, yet the plasma minimum and the interband transition onset are located at lower energies \[13\].

With the use of linear approximation of the optical conductivity \(\sigma_1\) \[see the example in the inset of Fig.3(b)\] we roughly estimate the onset of the interband transitions at each temperature and associate it with the optical gap \(E_{opt}\). The energy \(E_{opt}\) corresponds to electronic transitions from the highest occupied states below the Fermi level to the next higher lying band across the energy gap \(E_{gap}\), while \(E_{gap}\) corresponds to the smallest energy difference between the valence and conduction band [see Fig.2(b)]. The difference between \(E_{opt}\) and \(E_{gap}\) is due to the Moss-Burstein shift \[19\], which describes the (de)population of states in the conduction (valence) band and the subsequent shift of the Fermi level. The so-obtained values of \(E_{opt}\) are plotted in Fig.3(d): during cooling, an increase of the optical gap occurs down to 75 K, followed by a moderate decrease down to 20 K.
the Fermi level with two different bands is demonstrated. Due to the higher density of states of one band compared to the other, one Drude term has a much larger spectral weight than the other. The temperature-dependent values of the position of the L1 oscillator are summarized to the other, one Drude term has a much larger spectral gap. Consistently, we find similarities between the band gap. From the fitting [see Fig. 3(f)] and calculated according to the equations [22]

\[ \omega_{\text{pl}} = \sqrt{\omega_{\text{pl},1}^2 + \omega_{\text{pl},2}^2} \]  
\[ D = \frac{\omega_{\text{pl},1}^2 + \omega_{\text{pl},2}^2}{8\pi^2 c \sigma_{dc,x}} . \]  

The plasma frequency can also be expressed by the charge density \( N \) and the effective mass \( m^* \) with the formula \( \omega_{\text{pl}} = \sqrt{4\pi Ne^2/m^*} \) [22]. \( \omega_{\text{pl}} \) is increasing steadily

Figure 4. (a) \( \varepsilon_1 \) and (b) loss function of Mn(Bi0.07Sb0.93)2Te4 at selected temperatures. (c) and (d) show the temperature-dependent values of the screened plasma frequency \( \omega_{\text{scr}}^\text{pl} \) resulting from the zero-crossing of \( \varepsilon_1 \) and the peak position of the loss function, respectively. (e) and (f) display the temperature dependence of the plasma frequency \( \omega_{\text{pl}} \) and damping of the combined two Drude terms from the Drude-Lorentz fitting.
from 295K down to 18K, whereas below 18K it decreases, which symbolizes a weakening of the metallic character. This could either originate from a change in the charge carrier density or in the effective mass due to band profile modifications [see Fig.3(e)]. The damping is decreasing from 295K to 18K, and below 18K we find a strong increase. Accordingly, the lowering of temperature causes a growth in the metallic characteristics of the sample, but below $T_N$ this trend is reversed. Thus, also the parameters $\omega_{pl}$ and $D$ show an anomaly at approx. 20K, which agree with the values from Figs.1(c) and (d). We also point out that the temperature behaviors of $\omega_{pl}^{\text{scr}}$ and $\omega_{pl}$ are in good agreement with each other. Yet, the absolute values differ by a factor, since $\omega_{pl}^{\text{scr}}$ is affected by high-energy excitations and, hence, shifted to lower values, while $\omega_{pl}$ is solely determined by the Drude contributions. Quantitatively, the relation $\omega_{pl}^{\text{scr}} = \omega_{pl}/\sqrt{\varepsilon_{\infty}}$ holds [23]. At the lowest temperature (5K) we obtain $\omega_{pl}^{\text{scr}} = 844$ cm$^{-1}$ (from the loss function) and $\omega_{pl} = 5167$ cm$^{-1}$, which gives the value $\varepsilon_{\infty} = 37.5$. We extract a similar value ($\sim 43$) from the function $\varepsilon_1$ close to its maximum at $\sim 6000$ cm$^{-1}$. Hence, $\varepsilon_{\infty}$ characterizes the interband transitions at higher energies.

The observed weakening of the free charge carrier density at $T_N$ could be related to the opening of a gap in the electronic surface states when the antiferromagnetic order sets in. He et al. [23] discussed an opening of the topological surface gap in the case of pure MBT when the magnetic moments start to align in an A-type antiferromagnetic order. Although infrared spectroscopy is mainly sensitive to bulk electronic properties and less to surface states, this contribution should be minor, which applies to our findings. Eventually, this might explain the weakening of the metallic character of Mn(Bi$_{0.97}$Sb$_{0.03}$)$_2$Te$_4$ below $T_N$, as revealed by the temperature dependence of $\omega_{pl}^{\text{scr}}$ and $\omega_{pl}$.

IV. CONCLUSION

In conclusion, we studied the temperature-dependent optical functions of the magnetic insulator Mn(Bi$_{0.97}$Sb$_{0.03}$)$_2$Te$_4$ by reflectivity measurements, in order to characterize the effect of the antiferromagnetic ordering on the electronic structure near the Fermi level. Similar to the topological insulator MnBi$_2$Te$_4$, we have detected an anomaly in the profile of the spectra and several optical parameters at $T_N = 20$ K. From our findings, we conclude an interplay between the magnetic ordering and the electronic structure in Mn(Bi$_{0.97}$Sb$_{0.03}$)$_2$Te$_4$. The anomalous behavior might be caused by the opening of an exchange gap at the surface Dirac point, due to the breaking of the time reversal symmetry when the antiferromagnetic ordering sets in.

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