Terahertz Magnetospectroscopy of Cyclotron Resonances from Topological Surface States in Thick Films of Cd$_x$Hg$_{1-x}$Te

Maximilian Otteneder, Daniel Sacré, Ivan Yahniuk, Grigory V. Budkin, Kilian Diendorfer, Dmitry A. Kozlov, Ivan A. Dmitriev, Nikolay N. Mikhailov, Sergey A. Dvoretsky, Vasily V. Bel’kov, Wojciech Knap, and Sergey D. Ganichev

1. Introduction

The discovery of the quantum spin Hall effect in HgTe quantum wells [1,2] and of topological insulators [3–5] stimulated rapidly growing interest in fundamental properties and possible applications of HgTe-based materials. An attractive feature of these materials is that the band inversion, crucial for the formation of time-reversal symmetry-protected gapless states, can be obtained in several different ways. In particular, the topological phase transition can be realized via the variation of the thickness of HgTe quantum wells, by changing temperature, or by applying strain. This, together with a high quality of materials grown by molecular beam epitaxy (MBE), provides unique opportunities to study helical Dirac fermions (for reviews, see, e.g., refs. [5,6]) as well as induced superconductivity and phase-controlled Josephson junctions.[7,8]

Due to their specific energy dispersion, HgTe quantum wells and strained bulk films have revealed fascinating effects in magnetotransport [2,9] (for recent achievements, see refs. [10–19]), as well as in magneto- and terahertz-spectroscopy.[20–30]

More recently, a considerable attention has been attracted to bulk Cd$_x$Hg$_{1-x}$Te films with the Cd content $x$ equal or lower than the critical one, $x_c$, defining the phase transition from inverted to noninverted band order.[31] This interest has been driven by the observation of 3D massless Kane fermions in films with $x = x_c$,[32] followed by detailed studies using magneto- and terahertz-spectroscopy.[33–35] Topological insulators (TIs) based on bulk films with Cd contents below...
the temperature-dependent critical $x_c$ remain less studied so far.\textsuperscript{[16,17]} These materials, however, have a number of advantages with respect to HgTe TIs. As demonstrated by Tomaka et al.,\textsuperscript{[16]} the topologically protected surface states in Cd$_{x}$Hg$_{1-x}$Te are characterized by 1) two times higher Dirac fermion velocity (approximately the same as in graphene) than in pure HgTe, and 2) a larger band gap and a higher position of the Dirac point on the energy scale than that obtained in strained HgTe films. These significant advantages make TIs based on HgCdTe alloys promising for future applications.

Herein, we present a detailed study of terahertz cyclotron resonances (CRs) of thick undoped films observed in transmission, photocurrent,\textsuperscript{[25,38]} and photoconductivity.\textsuperscript{[39]} The films under study are characterized by different values of the Cd content $x$, corresponding to both normal and inverted band order. In all samples with band inversion, two CRs have been observed in the Faraday configuration, where both magnetic field and laser beam are oriented perpendicular to the sample surface. Evolution of these two distinct resonances with radiation frequency and helicity dependencies of the resonance amplitudes allow us to conclude that they represent CRs produced by two kinds of negatively charged carriers with slightly different masses. The cyclotron masses (below 0.01 $m_0$), obtained from the positions of the CRs, may be attributed to both bulk electrons and electrons in topological surface states, which are expected to exist in films with band inversion.

Our central observations come from CR measurements in Voigt configuration, still with normally incident THz radiation, but with a static magnetic field applied parallel to the sample surface. In this geometry, we obtain qualitatively different results for films with abruptly changing and gradually varying Cd content $x$ at the interfaces between layers with and without band inversion. The combination of measurements with magnetic field applied in-plane and out-of-plane provides an unequivocal experimental test on whether 2D or 3D carriers are responsible for the observed CRs. While samples with smoothly varying $x$ manifest almost identical resonant behavior in Faraday and Voigt configurations, in the sample with an abrupt transition to the cap layer one of the electron CRs disappears in the Voigt geometry. This observation clearly indicates a 2D nature of one group of electrons, apparently located at the abrupt interface and associated with a topological surface state. These conclusions are supported by theoretical modeling of the edge states in the films with abrupt and smooth interfaces. Therefore, the presence of a sharp interface appears to be a crucial technological requirement for studies of fundamental topological properties of such structures.

In samples without band inversion, we detect only one CR, with a CR mass close to that calculated for the bulk material. In all studied films, we also observe an additional broad resonance located in higher magnetic fields. This helicity-independent resonance manifests slower kinetics and decays rapidly with temperature. We assign this resonance to photoionization and magnetic freeze-out of impurity states.

2. Samples

The investigated samples are MBE-grown Cd$_x$Hg$_{1-x}$Te films having a common general layer structure shown in Figure 1.

For each sample, a 30 nm ZnTe buffer layer was grown on top of a (013)-oriented GaAs substrate, followed by a 6 µm layer of CdTe. After that, Cd$_x$Hg$_{1-x}$Te films with different composition (as described later) and thickness ranging between 5 and 12 µm were fabricated. All wafers were cleaved into square 5 × 5 mm$^2$ samples. Ohmic indium contacts have been soldered to the sample edges and corners. In addition, for detailed magnetotransport studies, Hall bar samples were fabricated from several wafers.

Figure 1b–f shows the cadmium content $x$ in Cd$_x$Hg$_{1-x}$Te films selected for the presentation as a function of the distance $d$ from the top of the preceding CdTe layer. In the initial step of growth of the film ($d \leq 1.5$ µm), the cadmium content $x$ was gradually decreased to reduce the strain related to a lattice mismatch, as well as to avoid possible interface disorder effects. Such smooth interfaces with varying $x$ are conventionally used to improve optoelectronic and transport properties of Cd$_x$Hg$_{1-x}$Te films, which are widely used for the detection of infrared radiation. The region of varying $x$ is followed by a wide region with constant $x = 0.15$ (samples #A, #B, and #C), $x = 0.18$ (sample #D),
or $x = 0.22$ (sample #E) which will be called the flat region in the following. After that, a cap layer with gradually increasing Cd content was grown in all samples except for the sample #C. In sample #C, the flat region with $x = 0.15$ was capped by a 30 nm layer of Cd$_{0.85}$Hg$_{0.15}$Te, which resulted in a sharp boundary between the flat region with band inversion and the cap layer without band inversion.

All samples were characterized by transport measurements performed using standard low-frequency lock-in technique in the temperature range from $2$ to $300$ K and an out-of-plane magnetic field up to $10$ T. For wafers with no Hall bars available, the corresponding longitudinal and Hall resistivities have been calculated from data obtained on the van der Pauw samples using permutation method. These studies established that all samples exhibit similar transport behavior that is apparently insensitive to the presence of band inversions and to the type of interfaces. At low temperatures, all samples show strong positive magnetoresistance and a nonlinear Hall effect. A typical example is shown in Figure 2 showing the longitudinal and Hall resistivities obtained from the magnetoresistance data recorded on a Hall bar sample made from wafer #A. The slope of the Hall resistivity at low magnetic field corresponds to negatively charged carriers (electrons), whereas at strong field the Hall slope typically changes to a hole-like. Such behavior is conventional for electron–hole systems\cite{40} in conditions when the hole density exceeds the electron density (although both have the same order of magnitude), whereas electrons are characterized by a significantly higher mobility. The values of sheet densities and mobilities for different types of carriers can be extracted using the two-component classical Drude model, which was successfully implemented for thin HgTe films.\cite{31, 38, 41} The values of sheet electron density $n_s$ obtained from such modeling are shown in Table 1. In this table, we also provide the values of average volume electron density $n$, which are obtained from the corresponding sheet electron densities $n_s$ as $n = n_s/w$, where $w$ denotes the corresponding Cd$_x$Hg$_{1-x}$Te film thicknesses (Figure 1b–f).

It should be emphasized that the aforementioned technique does not allow one to distinguish possible different groups of electrons (or holes) and yields the total electron and hole densities and average mobilities only. We also mention that the agreement between measured transport data and fits using the two-component model was not very good in the region of low magnetic fields. Therefore, in Figure 2d, we present results of an alternative simpler treatment within the single-component Drude model, where the effective sheet density of electrons in the film $n_s$ is obtained from the linear slope of the Hall resistivity at low magnetic fields.

Overall, transport measurements demonstrate that, at low temperatures, the electron sheet density is rather small and lies in a range of $n_s = \frac{2}{3} \times 10^{11} \text{ cm}^{-2}$. The hole sheet density is slightly larger than the electron one. The holes do not contribute to the observed CRs, so their properties will not be discussed in detail. However, they play an important role in keeping the overall electroneutrality. At higher temperatures, both electron and hole densities increase, mainly because of the usual temperature smearing of the energy distribution, but temperature variation of the bulk energy gap may also play an important role.\cite{33} Typically, at $77$ K, the electron sheet density reaches $n_s = 2 \times 3 \times 10^{12} \text{ cm}^{-2}$ for all samples, whereas at $300$ K the density rises to $n_s = \frac{3}{2} \times 10^{13} \times 1.2 \times 10^{14} \text{ cm}^{-2}$. It is worth noting that at liquid nitrogen temperature every sample shows an order of magnitude increase in density in comparison with low temperatures, and, at the same time, the values of $n_s$ are very similar for all samples independent of the value of the band gap in the flat region. Such behavior suggests that at $T < 77$ K, the bulk carriers do not play any essential role in transport and points to the formation of inversion/accumulation quasi-2D layers at the interfaces of the film, or, alternatively, to the formation of topological surface states with similar properties in samples with band inversion. Note that in both cases the bulk holes play an important role in maintaining overall charge neutrality in the sample.

![Figure 2](image.png)

**Figure 2.** Magnetoresistivity obtained on a Hall bar made from wafer #A with $x = 0.15$ at different temperatures, $T = 1.7$, 10, 15, 20, 25, 30, 40, 50, 70, 100, 150, and 300 K. a,b) The longitudinal sheet resistivity $\rho_{xx}$ in linear and logarithmic scale, respectively. c) The Hall resistivity $\rho_{xy}$. d) The effective sheet electron density $n_s(T)$ obtained from the linear slope of $\rho_{xy}$ at $B = 0$ in part (c).

**Table 1.** Basic parameters of the investigated samples #A–#E including the Cd content $x$, the corresponding type of the band order in the flat region, type of the top interface, the effective sheet electron density $n_s$ obtained from low-frequency magnetotransport measurements at $T = 4.2$ K, and the corresponding average volume density $n$ (obtained as $n_s$ divided by the total Cd$_x$Hg$_{1-x}$Te film thickness).

| Sample | Cd content, $x$ | Band structure | Top interface | $n_s[10^{11}\text{cm}^{-2}]$ | $n[10^{14}\text{cm}^{-2}]$ |
|--------|----------------|----------------|---------------|------------------|------------------|
| #A     | 0.151          | Inverted       | Smooth        | 2.9              | 4.8              |
| #B     | 0.150          | Inverted       | Smooth        | 2.2              | 3.7              |
| #C     | 0.151          | Inverted       | Sharp         | 3.3              | 6.7              |
| #D     | 0.179          | Normal         | Smooth        | 4.2              | 4.2              |
| #E     | 0.223          | Normal         | Smooth        | 2.4              | 1.9              |

\*Note: All samples were grown in all samples except for the sample #C. The Cd content was grown in all samples except for the sample #C.

\*Note: All samples were grown in all samples except for the sample #C. The Cd content was grown in all samples except for the sample #C.
3. Methods

For optical excitation, we used linearly polarized radiation from a continuous wave molecular gas laser operating at frequencies \( f = 2.54 \text{THz} \) (wavelength \( \lambda = 118 \, \mu\text{m} \), photon energy \( h\omega = 10.5 \, \text{meV} \), 1.63 THz \( (\lambda = 184 \, \mu\text{m} , h\omega = 6.74 \, \text{meV} ) \), and 0.69 THz \( (\lambda = 432 \, \mu\text{m} , h\omega = 2.87 \, \text{meV} ) \). The laser provided radiation power ranging between 40 and 120 mW depending on the wavelength. The radiation was modulated by an optical chopper operating at a frequency of about 80 Hz. The samples were placed in a temperature controllable optical cryostat with z-cut quartz and poly(methylpentene) windows. To block visible and near-infrared radiation, the windows were additionally covered by black polyethylene foil. The laser beam was focused using off-axis parabolic mirrors and controlled by a pyroelectric camera.\(^{[43]}\) It had an almost Gaussian shape with the spot diameter ranging from 1.5 to 3 mm depending on the wavelength. Room-temperature lambda plates made of x-cut quartz were used to modify the state of polarization, in particular, to obtain circularly polarized right-handed \((\sigma^+)\) and left-handed \((\sigma^-)\) radiation. For time-resolved measurements, we additionally used a pulsed optically pumped THz laser with a pulse duration of about 100 ns, a repetition rate of 1 Hz, and a peak power \( P \) of up to 60 kW.\(^{[44]-[46]}\) The laser emitted frequency lines in the range between 0.6 and 2.6 THz.

A split coil superconductive magnet was used to study magnetic field dependencies of radiation transmission and photocurrents. Most of the experiments were performed with normally incident THz radiation and magnetic field oriented either perpendicular to the film surface (Faraday geometry) or in-plane (Voigt geometry) (Figure 3a,b). In addition, experiments in Faraday geometry with the sample tilted by an angle \( \theta \) were conducted (Figure 3c).

For experiments on radiation transmission, a pyroelectric detector was placed behind the sample (Figure 3). Photovoltage and photoconductivity measurements were conducted using a standard lock-in technique. Photovoltage was measured using the phase-locked voltage drop across the sample under the influence of the chopper-modulated THz radiation in the absence of external DC bias. To measure the photoconductivity, an external DC bias was applied and photoresponses for positive and negative bias polarities were subtracted to exclude photogalvanic contributions and extract the photoconductivity signal. Alternatively, in some photoconductivity measurements, we applied a low-frequency AC bias and used a double-modulation technique described in previous studies.\(^{[39,47]}\)

Because main conclusions of this work are based on the comparison of results obtained using the three setups in Figure 3, it is important to recall the basic distinctions between them in application to 3D and 2D electron systems. In a 3D system, CR is expected to emerge in all three setups; the orientation of both the laser beam and static magnetic field with respect to the sample surface is immaterial for a 3D isotropic system; the dynamics of carriers is different for parallel (Faraday) and perpendicular (Voigt) relative orientation of the magnetic field and THz radiation propagation directions. This different dynamics may lead to a substantial shift of the resonance position in Voigt geometry with respect to Faraday geometry (see ref. [48] and Section 5 for details).

For a 2D system localized at an interface parallel to the sample surface, the decisive role is played by the orientation of the magnetic field with respect to the surface. Only the normal component of the magnetic field affects the in-plane dynamics of carriers and leads to the development of the cyclotron motion and the formation of Landau levels. In contrast, the in-plane static magnetic field can only slightly modify the confinement energy and the corresponding common wave function describing the localized 2D electron states. In the case of normal orientation of magnetic field (as in Faraday geometry in Figure 3a), one thus expects a usual CR dip in transmission for both 2D and 3D electrons. On the contrary, for in-plane magnetic field (as in Voigt geometry in Figure 3b) 2D electrons should manifest no CR, whereas 3D electrons can still experience CR.

4. Results

4.1. Cyclotron Resonances in Cd\(_{0.15}\)Hg\(_{0.85}\)Te Films with Band Inversion and Smooth Interfaces

We begin with the presentation of results of magnetotransmission and photovoltage measurements performed on almost identical Cd\(_{0.15}\)Hg\(_{0.85}\)Te films #A and #B (Figure 1b,c). The flat layer in both samples has a Cd content \( x \approx 0.15 \), corresponding to inverted bands for temperatures below \( T \approx 90 \text{K} \).\(^{[13]}\) On both sides of the flat layer, the Cd content gradually increases, thus providing smooth interfaces to regions without band inversion. Magnetotransmission data recorded in Faraday geometry (Figure 3a) for samples #A and #B are presented, correspondingly, in Figure 4 and Figure 5. The data here and later generally manifest three resonant dips for both negative and positive values of the out-of-plane magnetic field \( B \). The positions of observed resonances at \( B = \pm B_{\text{CR1,2}} \), \( B = \pm B_{\text{CR}} \), and \( B = \pm B_1 \), marked by arrows in Figures 4 and 5, are always symmetrically offset from the origin \( B = 0 \).
The main focus of this work concerns the sharp dips at low B which, as shown later, originate from the CR of free conduction electrons. In contrast, the broad minima at $B = \pm B_i$ are attributed to photoionization of impurities and will be addressed separately in Section 4.4. The data in Figures 4 and 5a show that the low-field resonances are sensitive to the radiation helicity. Although, for the right-handed circular polarization $\sigma^+$, they appear only at positive $B$, for the left-handed circular polarization $\sigma^-$, they are present at negative $B$ only. The position of these resonances scales linearly with the radiation frequency (Figure 5b). All these features indicate that the low-B sharp dips are indeed caused by the CR of negatively charged free carriers.

More specifically, magnetotransmission traces in Figure 4a that were recorded at the highest radiation frequency $f = 2.54$ THz show two distinct sharp dips at $B = \pm B_{CR1}$ and $B = \pm B_{CR2}$, which clearly reveals the presence of two kinds of electrons with different cyclotron masses (the corresponding values are shown in Table 2). It is worth mentioning at this point that the corresponding cyclotron masses below 0.01 $m_e$ may be attributed both to bulk electrons and electrons in topological surface states, which are expected to exist in the films with inverted bands. At lower frequency $f = 1.63$ THz, the CR dips start to merge, and become indistinguishable at the lowest frequency $f = 0.69$ THz where a single merged CR dip $B_{CR}$ is observed (Figure 4b,c). The positions of the resolved CR dips for $f = 2.54$ THz and $f = 1.63$ THz are found to be almost identical for both samples #A and #B (Figures 4 and 5 and Table 2).

Solid lines in Figure 4 show magnetotransmission fits obtained using a two-component Drude model with account for interference effects due to multiple reflections in the substrate. Specifically, the complex THz conductivity $\sigma$ is modeled as a sum of contributions $\sigma_j$ from each electron transport channel $j$, with individual $\sigma_j$ given by the Drude formula

$$\sigma_j = \frac{en_j}{\mu_j^{-1} + i(B_{CRj} \pm B)}$$

(1)

Here, $e$ is the elementary charge, $n_j$ and $\mu_j$ are the electron sheet density and mobility in the transport channel $j$, $B_{CRj} = m_j2\pi f/e$ defines the position of CR for a given THz frequency $f$ through the cyclotron mass $m_j$, and the $+(-)$ sign before $B$ corresponds to the left-handed (right-handed) circular polarization of the THz radiation. The fraction of transmitted power in Faraday geometry is then expressed through $\tilde{\sigma} = \sigma/2\mu_0 c$ as [49,50]

$$T(B) = \left| (1 + \tilde{\sigma}) \cos \phi - i \frac{1 + n_i^2 + 2\tilde{\sigma}}{2n_i} \sin \phi \right|^2$$

(2)
where \( \phi \) is the interference phase accumulated after single reflection in the substrate, \( n_r \) is the refractive index of the substrate, \( \varepsilon_0 \) the permittivity of free space, and \( c \) the speed of light. Using Equation (2), we were able to precisely fit all magnetotransmission traces. This demonstrates that the model based on Drude approximation is capable to reproduce the experimental observations. At the same time, the magnetotransmission traces do not provide sufficient data to convincingly extract all relevant parameters of the model. The fits to experimental data required an additional constant offset accounting for nonresonant contributions of other transport channels. Two identical Lorentzians centered at \( B = \pm B_e \) were included into the model to account for the impurity resonances discussed in Section 4.4.

The CRs were also detected in photovoltage experiments. A typical photovoltage trace, shown in the inset of Figure 6a, features two peaks coinciding with the positions of the CR dips in magnetotransmission as indicated by vertical dashed lines. Similar to the transmission dips, the position of the peaks scales linearly with the radiation frequency (Figure 6a). Note that, in the photovoltage response, two CR peaks are clearly resolved even for the lowest frequency \( f = 0.69 \) THz, in which case the CR dips in magnetotransmission get merged (Figure 4c).

Strikingly, applying a tilted magnetic field (see the experimental setup in Figure 3c), we obtain that the CR positions are independent of the tilt angle \( \theta \) (Figure 6b). This observation provides a first clear evidence that the resonances are excited in a 3D electron gas. Indeed, for a 2D system, one would expect a significant up-shift of the resonance position with the increase in the tilt angle because in this case the CR should be sensitive only to the normal component of the magnetic field. Here, it is important to stress that the tilted field measurements are performed in Faraday geometry, in which case the resonance position for an isotropic 3D system is expected to be independent of the tilt angle.

Measurements of the magnetotransmission in Voigt geometry (Figure 3b) confirm the aforementioned conclusions showing that even for a magnetic field oriented in-plane both CRs are still present (Figure 7). This observation rules out the possibility that any of the observed resonances originates from 2D electrons.

We note that, as expected, in Voigt geometry, the CR dips become helicity-independent and show up symmetrically for both polarities of magnetic field. The CR field values detected for sample #A are found to be slightly smaller in Voigt geometry than in Faraday geometry, which could be attributed to the 3D

![Figure 6](image-url)

**Figure 6.** a) Resonance field values \( B_{\text{CR1}}, B_{\text{CR2}}, \) and \( B_i \) (as marked) extracted from photovoltage measurements on sample #A at \( T = 4.2 \) K in Faraday configuration. The low-\( B \) part of a typical photovoltage trace, obtained at radiation frequency \( f = 2.54 \) THz, is shown in the inset. b) Dependence of the resonance field values on the tilt angle obtained from photovoltage measurements on sample #A at \( T = 4.2 \) K under \( f = 1.63 \) THz radiation in the Faraday configuration with tilted sample (Figure 3c). Dashed lines are linear fits to the data.
plasmonic shift (see ref. [48] and Section 5). Surprisingly, such shifts were not detected for other samples (Table 2).

4.2. Cyclotron Resonances in Cd$_{0.15}$Hg$_{0.85}$Te Film with Band Inversion and Sharp Top Interface

For sample #C, the behavior of resonances becomes qualitatively different despite the fact that this sample has almost the same design and that the Cd content in the flat region remains the same as in samples #A and #B. The only difference is that, instead of a smooth top interface as in samples #A and #B, sample #C has an abrupt boundary between the Cd$_{0.15}$Hg$_{0.85}$Te film and the Cd$_{0.85}$Hg$_{0.15}$Te cap layer (Figure 1d).

Similar to samples #A and #B, in Faraday geometry, sample #C clearly manifests two CRs in both magnetotransmission (see, e.g., blue curve in Figure 8a) and photovoltage (Figure 8c). However, in sharp contrast to samples with two smooth interfaces, in Voigt configuration only one CR dip remains (Figure 8b).

The fact that one of the resonances disappears in Voigt geometry provides a clear evidence that it originates from 2D confined electron states. Figure 8d shows the temperature dependence of the dip amplitude $\Delta T$. For resonances at $B_{CR1}$ (Faraday geometry) and $B_{V}$ (Voigt configuration), we observe that $\Delta T$ is almost independent of temperature. In contrast, the strength of the CR resonance at $B_{CR2}$, which is present in Faraday geometry only, strongly increases with rising temperature. Such temperature evolution is strikingly similar to that found recently for topological surface states in strained 80 and 200 nm HgTe films.[25,38] Taken together, the aforementioned observations clearly indicate the formation of 2D surface states at the abrupt interface between materials with and without band inversion.

4.3. Cyclotron Resonances in Films without Band Inversion

We proceed to results obtained on films #D (with $x = 0.22$) and #E ($x = 0.18$) which are expected to have normal band order. Indeed, the data we are going to discuss were obtained at liquid helium temperature at which the critical Cd content marking the transition from normal to inverted band order is $x_c \approx 0.17$.[33,51] Thus, in both samples, the Cd content in the flat region is higher than $x_c$, meaning that they should behave as conventional narrow-gap semiconductors.

In contrast to previously discussed materials with $x = 0.15$, for these samples, we observe a single CR in both Faraday and Voigt geometries. Magnetotransmission data for samples #D and #E is shown in Figure 9a,b and Figure 10a. The data in Figure 9a,b are obtained with linearly polarized THz radiation. Correspondingly, the CR dip is present for both polarities of magnetic fields. For circularly polarized radiation, the CR dip appears either in positive ($\sigma^+$) or negative ($\sigma^-$) $B$ (Figure 10a).

It is worth mentioning that the CR positions and the corresponding cyclotron masses obtained on samples #D and #E (see Table 2 and insets in Figures 9 and 10) are substantially
different from those obtained on the other samples with $x = 0.15$. This is consistent with our expectations: the largest cyclotron mass is obtained for sample #D with $x = 0.22$ which is supposed to have the largest band gap, whereas the lowest mass is obtained for sample #D with $x = 0.18$, closest to the critical Cd content $x_c \approx 0.17$ corresponding to a gapless material with a linear spectrum. We also mention that a strong asymmetry of the CR feature in Figure 10a is well captured by our fitting model and manifests significant role of interference effects included into Equation (2). Indeed, according to this formula, a symmetric shape of the transmission dip is expected either in conditions of constructive interference ($\sin \phi = 0$) or destructive interference ($\cos \phi = 0$). For intermediate values of $\phi$, the shape can be highly asymmetric\cite{Note1} and the observation of such strong asymmetry is a clear indication of the importance of interference effects.

The CRs were also detected in the photovoltage response. Under linearly polarized radiation as in Figure 9c, two symmetric CR peaks emerge in positive and negative $B$, whereas the circularly polarized radiation in Figure 10b gives rise to a single pronounced CR peak for the corresponding active magnetic field polarity only. In this particular case, an additional peak with substantially smaller magnitude was detected for the CR passive magnetic field $B > 0$, most probably caused by parasitic antenna effect produced by contact wires.

### 4.4. Impurity-Related Resonances

Apart from the sharp CRs at low $|B|$ which constitutes the main subject of this work, all samples manifested broad resonant features at higher magnetic fields which, as discussed later, can be naturally attributed to photoionization of impurities and thus are referred here as impurity resonances. The main properties of the impurity resonances, observed in magnetotransmission as well as in photovoltage and photoconductivity, can be summarized as follows. First of all, impurity resonances appear symmetrically with respect to $B = 0$ at $B = \pm B_i$ in both Faraday and Voigt geometries, and are insensitive to the radiation helicity (Figure 4). This fact alone provides a clear evidence that they may not be caused by the CR. The resonance positions $B = \pm B_i$ scale linearly with radiation frequency, as exemplified for samples #A and #B in Figure 5b. In contrast to the CR, in which case the amplitude of dips in transmission is either independent of temperature or increases with rising $T$, the amplitude of dips at $B = \pm B_i$ decreases quickly with growing $T$ and vanishes at $T \approx 30$ K (Figure 11a). The position $B_i$ of impurity resonances shifts to lower $|B|$ as $T$ grows (Figure 11b), whereas the CR positions are $T$-independent. The photoconductivity signal, as shown in Figure 11c, significantly increases around the position of the impurity resonance. The positions of impurity resonances for a given radiation frequency are substantially
different for samples with different cadmium content \( x \) (Table 2). In samples with \( x = 0.15 \) in the flat region (with band inversion), the impurity resonances were detected both in transmission and photovoltage, whereas in samples with \( x > x_c \) (without band inversion) they were present in photovoltage but not visible in magnetotransmission.

On a qualitative level, the aforementioned features are consistent with the photoionization mechanism if one considers that the activation energy of shallow impurities can significantly increase in magnetic field, an effect well-known as magnetic freeze-out of impurities.\(^{[52]}\) In this scenario, in the limit of low \( B \), the activation energy is so small that all impurities are thermally ionized making the photoionization processes impossible.\(^{[53]}\) In stronger \([B]\), the activation energy of impurities increases such that at sufficiently low \( T \) the majority of impurities are in the neutral state and, at the same time, the photon energy is high enough to promote electrons into the unbound states in the conduction band. Under the same conditions, the total occupation of the conduction states is lowered making the relative contribution of the photoexcited carriers more pronounced.

This is the parametric region of the impurity resonance where one expects most remarkable influence of the photoionization processes on magnetotransmission and photovoltage signals. In still stronger \([B]\) and low \( T \), the photoionization is blocked as soon as the energy distance between the ground level of impurity and available unbound states exceeds the photon energy. Photoionization is thus possible in an intermediate range of \([B]\) centered at a certain \( B_i \), and should be effective at sufficiently low temperatures only. Furthermore, the position of \( B_i \) should increase with the radiation frequency, and should also be highly sensitive to temperature and to the Cd content \( x \), as both of them modify the spectrum and, therefore, the position of impurity levels. On top of that, the temperature dependence can be affected by the processes of photoexcitation between the hydrogen-like impurity levels with subsequent thermal excitation into the continuum of unbound conduction states.\(^{[54,55]}\)

Assuming that the resonance at \( B_i \) is related to the ionization of impurity states, it is also natural to expect different kinetics of the photoresponse with respect to that at CR. These expectations were confirmed by time-resolved photocconductivity measurements performed using a pulsed THz laser. Figure 12 shows that indeed the decay of the photoresponse at the position \( B = B_i \) of the impurity resonance is several times longer than that measured at the position \( B = B_{CR} \) of CR. In the latter case (black trace in Figure 12), the photosignal essentially reproduces the form of the THz laser pulse (data not shown), meaning that kinetics of free carriers responsible for the CR is fast on the time scale of pulse duration, given by the full width at half maximum \( \tau_{\text{full}} \approx 100 \text{ ns} \). In contrast, the photosignal measured at \( B = B_i \) (red trace in Figure 12) features a significant long-time tail, which can be well fitted by an exponential function \( \exp(-t/\tau) \) (blue line in Figure 12) with a decay time \( \tau = 350 \text{ ns} \).

![Figure 11](image-url)  
**Figure 11.** a) Temperature dependence of the amplitude of Lorentzian dips in magnetotransmission around the impurity resonance at \( B = B_i \), extracted from data measured on sample #A at \( f = 2.54 \) THz as illustrated in the inset. b) Temperature dependence of the impurity resonance position \( B_i \) extracted from the same data. c) Photocconductivity \( \Delta \sigma \) normalized to the dark DC conductivity \( \sigma \) showing a resonant peak at position \( B = B_i \) of the impurity resonance. These data were obtained at \( T = 4.2 \) K on sample #A under \( f = 2.54 \) THz radiation with an applied current of 100 \( \mu \text{A} \).

![Figure 12](image-url)  
**Figure 12.** Time-resolved photocconductivity response \( U \approx \Delta \sigma \) (normalized to its maximum value) obtained on sample #A at liquid helium temperature using a pulsed THz laser operating at \( f = 2.03 \) THz. The black curve demonstrates a fast photoresponse at magnetic field \( B_{CR} = 0.29 \) T corresponding to the CR. The full width at half maximum of the CR photoresponse, \( \Delta \sigma_{\text{FWHM}} \approx 100 \text{ ns} \), coincides with the corresponding width \( \tau_{\text{pulse}} \) of the laser pulse profile (data not shown). In contrast, the photoresponse recorded at magnetic field \( B = 1 \) T, corresponding to the impurity resonance (red curve), shows a much slower decay which can be well fitted by an exponential function \( \exp(-t/\tau) \) (blue line) with a decay time \( \tau = 350 \text{ ns} \).
5. Discussion

We now return to the main observation: a qualitatively different behavior of one of the CRs in samples with sharp and smooth interfaces between materials with and without band inversion (Sections 4.1 and 4.2). The fact that one of the CRs disappears in the Voigt geometry for sample #C with a sharp interface is a clear indicator of the true 2D nature of carriers causing this resonance. The emergence of at least one surface state is indeed guaranteed by the topologically nontrivial transition from inverted to normal band order at the interface. In Faraday geometry, the same resonance exhibits a strong temperature dependence, which additionally distinguishes it from other resonances, which are present in both Voigt and Faraday geometries and presumably originate from carriers localized at smooth interfaces. These results indicate that smooth interfaces lead to the emergence of multiple surface states, known as Volkov–Pankratov states (VPS).\cite{16,19,56,57} As the number \( N \) of these states is not fixed by topology (apart from the requirement \( N \geq 1 \)), their number and properties should be sensitive to a particular choice of materials, growth conditions, form of interfaces, strain profile, and so on. The dynamics of carriers occupying multiple VPS can be effectively 3D, similar to conventional quasi-2D electrons occupying multiple sub-bands in a wide quantum well, which would immediately explain our observations.

Before presenting model calculations supporting this interpretation of the distinct behaviors at smooth and sharp interfaces, we first shortly comment on the observed frequency dependencies of the CRs and on the expected difference of resonance positions in Faraday and Voigt geometries. Both aspects deserve further systematic studies but do not influence our qualitative results and conclusions, and thus are of secondary importance for the purposes of the current work.

An inspection of the results presented in Figures 5, 6, 9, 10 and Table 2 shows that the magnetic field positions of CRs in all cases fairly well follow a linear frequency dependence. These observations seemingly conflict the well-known fact that in systems with linear spectrum the energies of individual transitions between Landau levels scale as \( B^{l/2} \).\cite{58} In fact, even for massless Dirac quasiparticles, the linear dependence should approximately restore in the quasiclassical limit of high chemical potential, such that many Landau levels are occupied, and the distance between them in the vicinity of the chemical potential is well described by the cyclotron mass.\cite{59,60} Such a classical description works well for all frequencies used in our work. Unfortunately, the presence of surface states and several types of carriers makes a reliable determination of the actual position of the chemical potential in our structures problematic. We expect that a reliable experimental evidence for the quantum square root scaling would require CR measurements in a broader frequency range.

As mentioned in Sections 3 and 4.1, for 3D electrons, the position of CR in Voigt geometry \( B_{V}^{CR} \) is expected to be down-shifted with respect to the CR position \( B_{CR} \) in Faraday geometry. Specifically, the resonance condition is given by \( \omega_{r}^{2} = \omega_{b}^{2} + \omega_{V}^{2} \) in Voigt geometry, instead of the usual \( \omega_{r}^{2} = \omega_{c}^{2} \) in Faraday geometry.\cite{61,62} Here, \( \omega_{b}^{2} = \frac{ne^{2}}{m_{e}} \) is the square of the plasma frequency of a 3D electron gas. Using for a rough estimate, the values \( \omega_{r} = \epsilon / \epsilon_{0} \approx 10 \) for the relative permittivity and \( m_{e}/m_{0} = 0.004 \) for the effective mass (Table 2) in the expression

\[
B_{V}^{CR} / B_{CR} = (1 - \omega_{0}^{2} / \omega^{2})^{1/2},
\]

for sample #A one finds that the observed relative shifts \( B_{V}^{CR} / B_{CR} \approx 0.8 \) correspond to the electron volume density \( n \sim 5 \times 10^{14} \text{ cm}^{-3} \), in a reasonable agreement with estimates obtained from the analysis of magnetotransport data (Table 1). At the same time, this agreement can be a coincidence because for other samples we generally do not observe any sensible difference between the CR positions in both geometries (Table 2). These surprising findings require further analysis and focused experiments. In particular, it would be interesting to understand how such plasmonic shifts are modified in the relevant case of a quasi-2D system involving several populated 2D sub-bands at the interface.

The surface states in systems with sharp interfaces are well studied for strained HgTe films.\cite{9,25,61,62} These topologically protected states are caused by the inversion of \( \Gamma_{6} \) and \( \Gamma_{8} \) bands at the interface. The energy of the surface states depends on the wave vector \( k_{f} \) in the interface plane and lies in the interval between the corresponding energy levels of the light and heavy holes.\cite{9,61} If the crystal is strained, there is a gap between the light- and heavy-hole sub-bands; the energy of the lowest \( k_{f} = 0 \) topological surface state coincides with the top of the heavy-hole sub-band, whereas in the limit \( |k_{f}| \rightarrow \infty \) the dispersion of the high-energy surface states approaches that of the light holes. As a result, ideally, the low-temperature transport properties of a strained HgTe film with the Fermi energy lying in the gap between the light- and heavy-hole sub-bands should be solely determined by the topologically protected surface states because

\[

to the quantum square root scaling would require CR measurements in a broader frequency range.

As mentioned in Sections 3 and 4.1, for 3D electrons, the position of CR in Voigt geometry \( B_{V}^{CR} \) is expected to be down-shifted with respect to the CR position \( B_{CR} \) in Faraday geometry. Specifically, the resonance condition is given by \( \omega_{r}^{2} = \omega_{b}^{2} + \omega_{V}^{2} \) in Voigt geometry, instead of the usual \( \omega_{r}^{2} = \omega_{c}^{2} \) in Faraday geometry.\cite{61,62} Here, \( \omega_{b}^{2} = \frac{ne^{2}}{m_{e}} \) is the square of the plasma frequency of a 3D electron gas. Using for a rough estimate, the values \( \omega_{r} = \epsilon / \epsilon_{0} \approx 10 \) for the relative permittivity and \( m_{e}/m_{0} = 0.004 \) for the effective mass (Table 2) in the expression

\[
B_{V}^{CR} / B_{CR} = (1 - \omega_{0}^{2} / \omega^{2})^{1/2}, \quad \text{for sample #A one finds that the observed relative shifts } B_{V}^{CR} / B_{CR} \approx 0.8 \text{ correspond to the electron volume density } n \sim 5 \times 10^{14} \text{ cm}^{-3}, \text{ in a reasonable agreement with estimates obtained from the analysis of magnetotransport data (Table 1). At the same time, this agreement can be a coincidence because for other samples we generally do not observe any sensible difference between the CR positions in both geometries (Table 2). These surprising findings require further analysis and focused experiments. In particular, it would be interesting to understand how such plasmonic shifts are modified in the relevant case of a quasi-2D system involving several populated 2D sub-bands at the interface.}

The surface states in systems with sharp interfaces are well studied for strained HgTe films.\cite{9,25,61,62} These topologically protected states are caused by the inversion of \( \Gamma_{6} \) and \( \Gamma_{8} \) bands at the interface. The energy of the surface states depends on the wave vector \( k_{f} \) in the interface plane and lies in the interval between the corresponding energy levels of the light and heavy holes.\cite{9,61} If the crystal is strained, there is a gap between the light- and heavy-hole sub-bands; the energy of the lowest \( k_{f} = 0 \) topological surface state coincides with the top of the heavy-hole sub-band, whereas in the limit \( |k_{f}| \rightarrow \infty \) the dispersion of the high-energy surface states approaches that of the light holes. As a result, ideally, the low-temperature transport properties of a strained HgTe film with the Fermi energy lying in the gap between the light- and heavy-hole sub-bands should be solely determined by the topologically protected surface states because

\[

\]
all bulk states in this case are gapped. Here, it is important to mention that, despite the very existence of a topological surface state stems from the inversion of light-hole band and conduction band at the interface, the dispersion of the surface states is strongly affected by hybridization with the heavy holes. CdHgTe films with band inversion share the aforementioned qualitative properties of strained HgTe films, but usually feature much smaller band gaps which are controlled by the Cd content of the alloy.

For a smooth interface between materials with and without band inversion, the situation is different. The calculations below show that instead of a single topologically protected state, the interface now can host multiple VPS.\(^{[16,19,56,57]}\) Similar to the case of the sharp interface, the energies of VPS lie in between the energies of the strain-split light and heavy holes. Figure 13 schematically shows a sharp interface (as in sample #C, Figure 13a) and a smooth interface (as in sample #A or #B, Figure 13b). The left parts in Figure 13a,b present the flat Cd\(_{0.15}\)Hg\(_{0.85}\)Te regions, whereas the middle parts show the transition to the top cap layer without band inversion and with a larger band gap sketched in the right parts. For the sharp interface, the spatial extension of the topologically protected surface state is determined by an exponential decay of the wave function inside both the film and the barrier. In contrast, the extension of VPS localized at a smooth interface is governed by the spatial gradient of the band gap near the band inversion point. The spatial distribution of the probability densities for the corresponding wave functions is illustrated in Figure 13. If one increases the width of the transition region between the topological insulator and normal insulator, both the total number of the VPS and their widths increase. This explains why two samples #A and #C with an almost identical design, but different interface structures, show different behavior in the Voigt configuration. Sample #C hosts 2D topologically protected states which do not experience CR in an in-plane of the interface magnetic field, whereas sample #A exhibits behavior typical for a quasi-2D system with many occupied sub-bands.

For numerical calculations, we implemented the six-band Kane model taking into account a static strain of the crystal which causes a splitting of the heavy- and light-hole sub-bands. In the basis of the \(\Gamma_6\) and \(\Gamma_8\) states, the Kane Hamiltonian has the form\(^{[61]}\)

\[
H = \begin{pmatrix}
E_c I_2 & H_{cv} \\
H_{cv} & E_c I_4 + H_{BP}
\end{pmatrix}
\]

where \(I_n\) is the \(n \times n\) identity matrix and

\[
H_{cv} = P_{cv} \begin{pmatrix}
-k_x - ik_y \\
\sqrt{2} k_z \\
-k_x + ik_y \\
0
\end{pmatrix}
\]

Here \(P_{cv}\) is the Kane parameter and \(H_{BP}\) is the Bir–Pikus Hamiltonian. To phenomenologically model the gap between light and heavy holes, we introduce a nonzero \(\alpha_{zz}\) component of the strain tensor, in which case the Bir–Pikus Hamiltonian has the form \(H_{BP} = b u_{zz} \text{diag}(-1, 1, 1, -1)\), where \(b\) is the valence band deformation potential and diag stands for a diagonal matrix. In the following, we set \(u_{zz}\) such that the heavy-hole sub-band is below the light-hole sub-band. This Hamiltonian for the bulk CdHgTe has three double-degenerate eigenstates: the light-hole sub-band states \(\epsilon_\ell(k)\) on top, the dispersionless states in the heavy-hole sub-band \(\epsilon_h(k)\) below them, and the low-lying conduction band states \(\epsilon_c(k)\), explicitly given by

\[
\epsilon_\ell(k) = E_0 + \sqrt{\delta^2 + 2P_{cv}^2 k^2/3}
\]

and

\[
\epsilon_h(k) = E_c - b u_{zz}
\]

with \(2E_0 = E_c + b u_{zz} + E_c\) and \(2\delta = E_c + b u_{zz} - E_c\).

To calculate the VPS spectrum and wave functions for a smooth interface, we take into account the dependence of \(E_c\) and \(E_r\) on the coordinate. Using the parameters \(2m_0(P_{cv}/\hbar)^2 = 18.8\ \text{eV}\)\(^{[62]}\) the band gap \(E_r - E_c = -32\ \text{meV}\) for Cd\(_{0.13}\)Hg\(_{0.87}\)Te\(^{[63]}\) and the dependencies of \(E_r(z)\) and \(E_c(z)\) found from the profile of compound distribution shown in Figure 1a, we calculated the VPS for sample #A. In this calculation, we set \(u_{zz} = 2\ \text{meV}\) and add a static electric field \(E = 2\ \text{kV cm}^{-1}\) at the band closing. At \(k_0 = 0.02\ \text{nm}^{-1}\), such calculation yields three VPS with energies 3, 5, and 8\,meV with respect to the heavy-hole energy at the \(\Gamma\) point in the Cd\(_{0.15}\)Hg\(_{0.85}\)Te film. The probability density of the VPS is distributed over hundreds of nanometers, which is greater or of the order of the cyclotron radius \(r_c\) of charge carriers in the bulk CdHgTe film. The latter can be found as \(r_c = v/\omega\) using \(\omega = 2\pi f\) and an estimate \(v = \sqrt{2/3 P_{cv}/\hbar} \approx 10^8\ \text{cm s}^{-1}\) for the carrier velocity. This yields the values of \(r_c = 65, 100,\) and 240\,nm for \(f = 2.54, 1.63,\) and 0.69\,THz, respectively. We also find that the number of VPS emerging at such a smooth interface is highly sensitive to variation of the band gap, as well as to the energy splitting between light- and heavy-holes caused by deformation and the static electric field. The latter parameters are not known precisely and are introduced here as phenomenological parameters. Taken together, the presented calculations confirm that smooth interfaces in our samples can indeed host many VPS leading to a quasi-2D behavior of the CRs, which are thus present for both in-plane and out-of-plane orientations of the magnetic field.

6. Conclusion

To summarize, the presented low-temperature THz spectroscopy studies of CRs in thick films of Cd\(_{x}\)Hg\(_{1-x}\)Te unambiguously show that 2D surface states are present only in structures with a sharp interface separating the internal layer with band inversion \((x < 0.17)\) and a cap layer without band inversion. In contrast, conventional smooth interfaces between materials with and without band inversion, involving regions of Cd\(_{x}\)Hg\(_{1-x}\)Te with gradually changing Cd content \(x\), make the surface states effectively 3D such that the corresponding CRs are clearly visible even for an in-plane orientation of the applied magnetic field. Our observations, supported by the calculations describing the formation of the surface states in films with sharp
and smooth interfaces, clearly demonstrate that future studies of topologically protected surface states in Cd$_{1-x}$Hg$_x$Te films require the structures with sharp interfaces. Apart from the CRs, all samples manifest broad helicity-independent resonances which emerge at higher magnetic fields and show slower kinetics and higher temperature sensitivity. These broad resonances are attributed to photoionization and magnetic freeze-out of impurity states.

Acknowledgements

The authors thank S.A. Tarasenko for fruitful discussions. The support from the DFG priority program SPP 1666 “Topological Insulators: Materials – Fundamental Properties – Devices” (project no. GA501/12-2) and the IRAP program of the Foundation for Polish Science (grant no. MAB/2018/9, project CENTERA) and TEAM project POIR.04.04.00-00-3D76/16 550 (grant no. TEAM/2016-3/25) are gratefully acknowledged. MAB/2018/9, project CENTERA) and TEAM project POIR. Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

CdHgTe films, cyclotron resonances, magnetospectroscopy, surface states, terahertz, topological Volkov–Pankratov states

Received: January 13, 2020
Revised: March 27, 2020
Published online: May 4, 2020
