Distinction between electron states formed at topological insulator interfaces with the trivial phase and vacuum

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In this paper, we show that electron states formed in topological insulators at the interfaces topological phase–trivial phase and topological phase–vacuum may possess different properties. This is demonstrated on an example of heterostructures based on thick topological Hg1−xCd,xTe films, in which the PT-symmetric terahertz photoconductivity is observed. It is shown that the effect originates from features of the interface topological film–trivial buffer/cap layer. The PT-symmetric terahertz photoconductivity is not provided by electron states formed at the interface topological film–vacuum.

Physics of topological insulators (TI) is one of the hottest topics of the modern solid state physics. In these materials, the conduction and valence bands in the bulk are inverted due to strong spin–orbit interaction. Consequently, two-dimensional electron states are necessarily formed in a close proximity of the TI surface. These 2-D electron states possess Dirac dispersion relation with the spin direction locked perpendicularly to the momentum direction1. Existence of such topological 2-D electron states was first predicted theoretically2,3, and then directly confirmed experimentally through ARPES measurements4,5. Topological 2-D electron states are expected to be formed not only at the TI surface, but in heterostructures, at the interface trivial buffer–topological film as well1,6,7. It was suggested that the 2-D topological electron states formed at the two types of interfaces mentioned above, have analogous features1.

Hg1−xCd,xTe solid solutions reveal certain unusual features among topological insulators. HgTe possesses an inverted band structure and represents the TI phase which was unambiguously confirmed by ARPES measurements8,9. Increasing the cadmium telluride mole fraction leads to the composition-driven transition from the topological to the trivial insulator phase at x = 0.16 (T = 0)10–13. In contrast to most of the other TIs, Hg1−xCd,xTe possess relatively low free carrier concentration14,15, which facilitates measurements of electric transport via the topological surface channel on top of the bulk conductivity. Importantly, the same circumstance allows measurements of photoelectric phenomena including photogalvanic effects16,17, cyclotron resonance18–20 and photoconductivity21–26 in the topological phase of Hg1−xCd,xTe. In particular, the unusual PT-symmetric photoconductivity has been observed in heterostructures based on thick topological Hg1−xCd,xTe films, under terahertz excitation27.

In the experiments described in27, the photoconductivity measurements have been performed at the temperature T = 4.2 K in magnetic fields up to 4 T in the Faraday geometry. The photoconductivity amplitude was not an even function of the magnetic field thus breaking the T (time reversal) symmetry. It was also different for mirror-symmetric potential probe couples of a Hall bar, demonstrating breaking of the P (parity) symmetry. At the same time, simultaneous changing of the magnetic field direction and the potential probe couple kept the photoconductivity intact, revealing therefore the PT-symmetry.

The goal of this paper is to reveal the heterostructure element which corresponds to appearance of the PT-symmetric photoconductivity. We show that this effect originates from features of the interface between the topological and the trivial phase materials. Instead, it does not appear at the interface topological phase–vacuum, for which existence of Dirac spin polarized electronic states was confirmed by ARPES previously8,9.
Results
We have studied photoconductivity induced by terahertz laser pulses in heterostructures based on 4 μm thick Hg$_{1-x}$Cd$_x$Te films. They were grown on a semi-insulating GaAs substrate via ZnTe and CdTe buffer layers followed by a graded-gap Cd-rich Hg$_{1-x}$Cd$_x$Te relaxed layer. In addition, a protecting 10 nm thick trivial phase CdTe cap layer was deposited on top of the structures (Fig. 1). The cadmium telluride content in the film $x = 0.145$ corresponds to the topological phase. The CdTe content $x$ variation at the active layer/trivial buffer interface is smooth, whereas it is sharp at the active layer/cap-layer interface.

To reach the paper goal, two types of structures were studied: the initial one, and the one with the cap layer etched out, so that the Hg$_{1-x}$Cd$_x$Te film surface was open.

Standard Hall bars with the dimensions 5·0.5 mm$^2$ were prepared by photolithography. Both films were of the $n$-type with the free electron concentration $\sim 10^{15}$ cm$^{-3}$ at the liquid helium temperature. The photoconductivity measurements were performed at $T = 4.2$ K in the Faraday geometry in magnetic fields up to 0.5 T. Further on, we denote the magnetic field direction coinciding with the incident radiation flux as $B^+$, and the opposite direction as $B^-$. Let us consider first the initial structure. The respective photoconductivity kinetics is presented in the Fig. 2a. In the zero magnetic field, the photoconductivity is negative in the beginning of a laser pulse, then it becomes slightly positive. As the magnetic field $B^+$ grows, the positive photoconductivity is strongly enhanced.
Its amplitude $A$ (see Fig. 2a for the definition) reaches the maximum at $B^+ \approx 0.15$ T and drops in higher fields (Fig. 3a, solid red points). For the opposite magnetic field direction $B^-$, the positive photoconductivity amplitude continuously decreases as the $B^-$ value grows. It should be noted that for the opposite mirror-symmetric couple of potential probes (see the inset in Fig. 3a), the positive photoconductivity behaves in the opposite way: it is enhanced for the $B^-$ field direction, and reduces for the $B^+$ one (Fig. 3a, open red points). Therefore, the $PT$-symmetric behavior of the positive photoconductivity is observed, as in 27.

Strikingly, etching out a very thin 10 nm cap layer results in drastic changes of the photoconductivity kinetics and amplitude. Now the photoconductivity is positive in the zero field, the contribution of the negative photoconductivity starts to appear only in non-zero magnetic fields (Fig. 2b). The positive photoconductivity remains $PT$-symmetric, but its amplitude rises by several times compared to the initial structure (Fig. 3, black points).

The key features of the effects observed do not depend on the laser radiation frequency from 1.1 to 3.3 THz, as well as on the radiation polarization, neither linear, nor circular.

**Discussion**

The photoconductivity observed in the structures studied consists of several components. The first one is the negative photoconductivity that is almost even in magnetic field and practically does not differ for two mirror-symmetric potential probe couples of a Hall bar. Previously, it was demonstrated that this contribution to the photoconductivity results from heating free electrons by incident THz radiation pulses. Such a heating results in a mobility drop and, consequently, in the negative photoconductivity25,26.

The second component is the positive photoconductivity, which is not symmetric neither in magnetic field, nor for the potential probe couples located at the opposite sides of a Hall bar. Phenomenologically, the positive photoconductivity amplitude $A$ may be considered as a sum of the even $A_e$ and odd $A_o$ in magnetic field contributions. The respective decomposition of the $A$ value into the even $A_e$ and odd $A_o$ components for the two structures studied is presented in the Fig. 3, right upper and lower panels, respectively. It can be seen that while the even in magnetic field contribution $A_e$ is very close for the potential probe couples at the opposite Hall bar sides, the odd contribution $A_o$ for the two Hall bar sides has the same amplitude, but the opposite sign.

Such a decomposition is not just a way to present the results, but has a definite physical meaning. As it was demonstrated in 27, the odd in magnetic field contribution is due to appearance of the unusual chiral non-local photoconductivity. In the non-local photoconductivity measurement geometry, when the bulk conductivity is excluded, photocurrents induced by the incident terahertz radiation flow along the sample edge around a sample. The photocurrent chirality changes every time the magnetic field direction or the electric bias polarity is reversed. The photocurrents are absent if the magnetic field or the external bias is not applied. Apparently, in the standard Hall bar geometry used in the present study, the chiral non-local photocurrent adds up to the usual bulk photocurrent at one edge of the Hall bar, and is subtracted from it at the opposite edge providing appearance of the odd in magnetic field photoconductivity contribution and giving rise to the $PT$-symmetric photoconductivity.

The symmetric in magnetic field photoconductivity part $A_e$ may have a traditional origin and correspond to photoexcitation in the sample bulk. The answer to the question on what is the heterostructure element that is responsible for appearance of the odd in magnetic field photoconductivity component $A_o$, is not so straightforward.
First, it is clear that the bulk of the film may not be such an element since the respective photocurrent flows along a sample edge. Therefore, only the side surface of the film and the edges of the film–buffer layer and film–cap layer interfaces may be candidates for this element (Fig. 4). Let us consider these options.

If we suppose that the $A_o$ photocurrent flows along the side film surface representing the topological film–vacuum interface (Fig. 4a), then etching out the cap layer would lead to disappearance of the effect. Indeed, after removal of the cap layer, the top film surface becomes the topological film–vacuum interface as well. Then the respective photocurrent would flow not only along the sample edge, but via the top surface, too, thus shunting the edge photocurrent contribution. Experimentally, however, the $PT$-symmetric photoconductivity is strongly enhanced upon removal of the cap layer (see Fig. 3), to the contrary of the expected behavior. Therefore, the topological film–trivial buffer and topological film–trivial cap layer interfaces are the only remaining options.

The second important point is the following. Apparently, the $A_o$ photocurrent components corresponding to the film–buffer and film–cap layer interfaces, to a great extent compensate each other (Fig. 4b). Indeed, when the film–cap layer interface disappears in the etched structure, the antisymmetric photocurrent component $A_o$ grows by several times (see Fig. 3). The reason for such a behavior may be the following. As it was suggested in26, the positive photoconductivity in the zero magnetic field is due to diffusion of electrons heated in the film bulk, to the heterojunction area, where the topological layer is located. When entering the topological layer, the diffusing electrons acquire higher mobility thus providing appearance of the positive photoconductivity. In the non-zero magnetic field, such a diffusion occurs along or against the magnetic field direction for the two different interfaces. This could be a reason for the fact that the two interfaces act towards each other and compensate the effect significantly. The effect amplitude apparently depends on the details of the interface structure, so the film–buffer and the film–cap layer interfaces do not compensate each other exactly.

Figure 4. Possible options for the element of the structure responsible for the odd photoconductivity component appearance. (a) The chiral photocurrent flows along the topological film–vacuum interface; (b) chiral photocurrents flow along the edges of the topological film–trivial buffer/cap-layer interfaces.
As it has been shown in\textsuperscript{19} for analogous structures based on thick topological Hg\textsubscript{1−x}Cd\textsubscript{x}Te films, the electronic states located at smooth and sharp interfaces between the topological and trivial phases have different structures. While the sharp interface forms 2D electron states, the smooth interface produces 3D-like states. It was suggested that the sharp interface forms true 2D electron states, whereas for the smooth interface, a series of Volkov–Pankratov interface states are formed\textsuperscript{19}. In the latter case, these electronic states are effectively 3D, in an analogy to conventional quasi-2D electron states in a wide quantum well occupying multiple subbands.

The results obtained in the present study suggest that both true 2D interface states at the topological film–cap layer interface and quasi-3D interface states at the topological film–trivial buffer interface provide appearance of the chiral non-local edge photocurrents and, consequently, of the PT-symmetric photoconductivity. Moreover, it seems that the effect is more strongly pronounced for the smooth interface since the amplitude does not change sign upon removal of the cap layer. The reasons for such a behavior may be the following.

The Volkov–Pankratov interface states, while being quasi-3D, are located at the interface and still possess higher mobility compared to the film bulk, which is an important condition for appearance of the positive photoconductivity in the zero magnetic field. Another possible option is related to the alloy composition fluctuations at the smooth interface. In such a situation, the 2D topological state may be not flat, as it is for the sharp interface, but rippled. Then the effective surface of the 2D boundary between the TI film and the trivial buffer at the smooth interface may be larger than for the sharp interface between the film and the cap layer providing higher chiral photocurrent.

It should be stressed that the microscopic origin of the chiral non-local edge photocurrents providing appearance of the PT-symmetric photoconductivity is still unclear. Moreover, it contradicts apparent symmetry arguments\textsuperscript{27,28}. It means that there exists an unaccounted external factor that breaks the P- and T-symmetries and leads to the effects observed. The origin of this external factor is to be determined, it is only clear that it is not a deviation from the ideal experimental geometry or any anisotropy of the sample\textsuperscript{27}. Any sort of a microscopic model for the effect origin must take into account this factor.

On the other hand, the results presented show that the interface between the topological film and the trivial buffer/cap layer is the heterostructure element where the effect appears. The topological film–vacuum interface does not possess such a feature.

Conclusions

In summary, we have demonstrated that the electron states formed at the interfaces topological insulator–trivial insulator and topological insulator–vacuum possess different features. In particular, we have shown that the PT-symmetric terahertz photoconductivity observed in heterostructures based on thick Hg\textsubscript{1−x}Cd\textsubscript{x}Te films being in the topological phase, results from processes at the interfaces film–buffer/cap layer, while the interface film–vacuum does not possess this feature. Besides, it was shown that the topological phase–trivial phase buffer and the topological phase–trivial phase cap layer form interfaces that strongly compensate each other in the effect of PT-symmetric photoconductivity. Finally, the PT-symmetric photoconductivity effect was directly linked to the non-local chiral terahertz photoconductivity. A way to extract the contribution of the chiral non-local photocurrent from measurements in the traditional Hall bar geometry has been presented.

Methods

The samples were grown by the MBE technique on semi-insulating (013) GaAs substrates. Composition of all layers of the initial heterostructure was controlled by ellipsometry in situ during growth\textsuperscript{29}. The accuracy of the composition x measurements was about 0.001, as estimated through the ellipsometric results. The procedure accuracy was confirmed by measurements of transmission spectra in samples with successively etched surface layers\textsuperscript{30}. The active layer composition x = 0.145 in the initial heterostructure corresponds to the topological phase.

In the second heterostructure studied, the cap CdTe layer was removed using the HBr + 0.01% Br\textsubscript{2} etchant. The etching rate ~ 0.05 μm/min was controlled by measuring the shift of interference peaks in the transmission spectra of the initial and the etched control sample. Deletion of the CdTe cap layer was monitored through measurements of the reflectivity spectra. The whole process was performed analogously to described in\textsuperscript{31,32}.

Electrical characterization of the samples has been done in the temperature interval 4.2–300 K. The resistivity temperature dependence of the samples studied is shown in the Fig. 5. This dependence has a minimum which is characteristic for the inverted band structure of Hg\textsubscript{1−x}Cd\textsubscript{x}Te, and hence the topological phase\textsuperscript{13,22}.

The photoconductivity was excited by a pulsed NH\textsubscript{3} gas laser with the pulse length ~ 100 ns. The laser radiation frequency was 1.1 or 3.3 THz with the power in a pulse up to 10 kW. The beam had an almost Gaussian profile with the spot size 3–5 mm depending on the laser frequency. The radiation polarization, either linear or circular, was controlled by rotation of λ/2 or λ/4 plates introduced into the laser beam. More details on the experimental setup may be found in\textsuperscript{33–36}.

The sample was directly immersed in the liquid helium to minimize possible sample heating effects. The magnetic field up to 0.5 T, if applied, was directed perpendicularly to the sample surface. The photoconductivity measurements were performed using the 4-probe method, for two opposite directions of the applied bias, as well as with the zero bias to control a possible appearance of the photovoltaic signal. In all cases, the photovoltaic contribution to the signal was negligible.

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References

1. Hasan, M. Z. & Kane, C. L. Colloquium: Topological insulators. Rev. Mod. Phys. 82, 3045–3067 (2010).
Figure 5. Resistivity temperature dependence of the samples under study.

2. Bernevig, B. A., Hughes, T. A. & Zhang, S. C. Quantum spin Hall effect and topological phase transition on HgTe quantum wells. *Science* **314**, 1757–1761 (2006).
3. Fu, L. & Kane, C. L. Topological insulators with inversion symmetry. *Phys. Rev. B* **76**, 045302 (2007).
4. Hsieh, D. et al. A topological Dirac insulator in a quantum spin Hall phase. *Nature* **452**, 970–975 (2008).
5. Hsieh, D. et al. A tunable topological insulator in the spin helical Dirac transport regime. *Nature* **460**, 1101–1105 (2009).
6. Volkov, B. A. & Pankratov, O. A. Two-dimensional massless electrons in an inverted contact. *JETP Lett.* **42**, 145–148 (1985).
7. Volkov, B. A., Pakhomov, S. V. & Pankratov, O. A. Supersymmetry in heterojunctions: Band-inverting contact on the basis of Pb, Sn, Te and Hg, Cd, Te. *Solid State Commun.* **61**(2), 93–96 (1987).
8. Brüne, C. et al. Quantum Hall effect from the topological surface states of strained bulk HgTe. *Phys. Rev. Lett.* **106**, 126803 (2011).
9. Liu, C. et al. Tunable spin helical Dirac quasiparticles on the surface of three-dimensional HgTe. *Phys. Rev. B* **92**, 115436 (2015).
10. Rogalski, A. HgCdTe infrared detector material: History, status and outlook. *Rep. Prog. Phys.* **68**, 2267–2336 (2005).
11. Weiler, M. In *Semiconductors and Semimetals* Vol. 16 (eds Willardson, R. & Beer, A.) 119 (Academic Press, 1981).
12. Orlita, M. et al. Observation of three-dimensional massless Kane fermions in a zinc-blende crystal. *Nat. Phys.* **10**, 233–238 (2014).
13. Tepe, F. et al. Temperature-driven massless Kane fermions in HgCdTe crystals. *Nat. Commun.* **7**, 12576 (2016).
14. Dvoretzky, S. et al. Growth of HgTe quantum wells for IR to THz detectors. *J. Electron. Mater.* **39**, 918–923 (2010).
15. Varavin, V. S., Dvoretzky, S. A., Liberman, V. I., Mikhailov, N. N. & Sidorov, V. G. Molecular beam epitaxy of high quality Hg, Cd, Te films with control of the composition distribution. *J. Cryst. Growth* **159**, 1161–1166 (1996).
16. Hubmann, S. et al. Symmetry breaking and circular photogalvanic effect in epitaxial Cd, Hg, Te films. *Phys. Rev. Mater.* **4**(4), 043607 (2020).
17. Dantscher, K.-M. et al. Photogalvanic probing of helical edge channels in two-dimensional HgTe topological insulators. *Phys. Rev. B* **95**, 201103 (2017).
18. Olbrich, P. et al. Giant photocurrents in a Dirac fermion system at cyclotron resonance. *Phys. Rev. B* **87**, 235439 (2013).
19. Otteneder, M. et al. Terahertz magnetospectroscopy of cyclotron resonances from topological surface states in thick films of Cd, Hg, Te. *Phys. Status Solidi B* **258**(1), 200023 (2021).
20. Zhohulev, M. S. et al. Cyclotron resonance in HgCdTe-based heterostructures in strong magnetic fields. *J. Phys. Conf. Ser.* **461**, 012038 (2013).
21. Ruffenach, S. et al. HgCdTe-based heterostructures for terahertz photonics. *APL Mater.* **5**(3), 035503 (2017).
22. Kvon, Z. D. et al. Cyclotron resonance photoconductivity of a two-dimensional electron gas in HgTe quantum wells. *Phys. E* **40**(6), 1885–1887 (2008).
23. Savchenko, M. L. et al. Terahertz photoresistivity of a high-mobility 3D topological insulator based on a strained HgTe film. *Appl. Phys. Lett.* **117**, 211103 (2020).
24. Yaroshchuk, A. S., Kvon, Z. D., Gusev, G. M. & Mikhailov, N. N. Microwave photoresistance of a two-dimensional topological insulator in a HgTe quantum well. *JETP Lett.* **112**(2), 121–125 (2020).
25. Galeeva, A. V. et al. Terahertz Photoconductivity in Hg, Cd, Te near the transition from the direct to inverted spectrum. *JETP Lett.* **106**, 162–166 (2017).
26. Galeeva, A. V. et al. Non-equilibrium electron transport induced by terahertz radiation in the topological and trivial phases of Hg, Cd, Te. *Beilstein J. Nanotechnol.* **9**, 1035–1039 (2018).
27. Galeeva, A. V. et al. Apparent PT-symmetric terahertz photoconductivity in the topological phase of Hg, Cd, Te-based structures. *Sci. Rep.* **10**, 2377 (2020).
28. Kazakov, A. S. et al. Non-local terahertz photoconductivity in the topological phase of Hg, Cd, Te. *Sci. Rep.* **11**, 1587 (2021).
29. Svitasev, K. K. et al. The growth of high-quality MCT films by MBE using in-situ ellipsometry. *Cryst. Res. Technol.* **29**(7), 931–937 (1994).
30. Sidorov, Yu. G. et al. Molecular-beam epitaxy of mercury–cadmium–telluride solid solutions on alternative substrates. *Semiconductors* **35**, 1045–1053 (2001).
31. Varavin, V. S. et al. HgCdTe structures for dual-band photodetectors operating in the 3–5 and 8–12 μm spectral regions. *Optoelectron. Instrum. Data Process.* **49**, 476–484 (2013).
32. Dvoretzky, S. A. et al. HgCdTe quantum wells grown by molecular beam epitaxy. *Semicond. Phys. Quantum Electron. Optoelectron.* **10**, 47–53 (2007).
33. Ganichev, S. D., Emelyanov, S. A. & Yaroshetskii, I. D. Spectral sign inversion of photon drag at far-IR wavelengths. *JETP Lett.* **35**, 368 (1982).
34. Belkov, V. V. et al. Circular photogalvanic effect at inter-band excitation in semiconductor quantum wells. Solid State. Commun. 128, 283–286 (2003).
35. Shalygin, V. A. et al. Spin photocurrents and the circular photon drag effect in (110)-grown quantum well structures. JETP Lett. 84, 570–576 (2006).
36. Plank, H. et al. Photon drag effect in (Bi1−xSbx)2Te3 three-dimensional topological insulators. Phys. Rev B 93, 125434 (2016).

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Author contributions
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Competing interests
The authors declare no competing interests.

Additional information
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