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Split Dirac cones in HgTe/CdTe quantum wells due to symmetry-enforced level anticrossing at interfaces

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HgTe is a band-inverted compound which forms a two-dimensional topological insulator if sand-wiched between CdTe barriers for the HgTe layer thickness above the critical value. We describe the fine structure of Dirac states in the HgTe/CdTe quantum wells of critical and close-to-critical thickness and show that the necessary creation of interfaces brings in another important physical effect: The opening of a significant anticrossing gap between the tips of the Dirac cones. The level repulsion driven by the natural interface inversion asymmetry of zinc-blende heterostructures considerably modifies the electron states and dispersion but preserves the topological transition at the critical thickness. By combining symmetry analysis, atomistic calculations, and extended $k \cdot p$ theory with interface terms, we obtain a quantitative description of the energy spectrum and extract the interface mixing coefficient. We discuss how the fingerprints of the predicted zero-magnetic-field splitting of the Dirac cones could be detected experimentally by studying magnetotransport phenomena, cyclotron resonance, Raman scattering, and THz radiation absorption.

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The study of systems with gapless and lineardispersion states constituting the Dirac cone is central to the physics of topological insulators (TIs) [1, 2]. Such states are formed in the primary insulating band gap of the bulk material and studied at the surface via techniques such as angular resolved photoemission [3, 4]. While some TI compounds such as Bi₂Se₃ and Bi₂Te₃ show an insulating gap in their three-dimensional (3D) bulk band structure and so the Dirac cones can be studied at their 2D surface without modifying the material, a Mercury Telluride crystal does not have a 3D bulk band gap because the Fermi level resides within the four-fold degenerate Γ_8 band [5]. However, the topological insulation is realized by straining HgTe which opens a gap within the otherwise fourfold degenerate Γ_8 states [6, 7] or growing the material in a HgTe/CdTe quantum well (QW) geometry [8–10]. In the latter case, CdTe (or $Cd_{1-x}Hg_xTe$) barriers create quantum confinement within HgTe with the normal or inverted band structure depending on the well thickness, so that HgTe/CdTe QWs belong to the class of normal or topological insulators.

In $\mathrm{HgTe/CdTe}$ QWs of critical thickness, the heavy-hole subband HH1 switches order with the electron subband E1 [11], the band gap vanishes and elementary excitations behave as massless Dirac 2D fermions [12, 13]. The early theoretical description [8] used a model in which the symmetry of the $\mathrm{HgTe/CdTe}$ QW was implicitly assumed to contain an inversion center. Consequently, the point-group representations of the E1 and HH1 subbands are different, so these states do not mix with each other and are allowed to cross. In reality, the host zinc-blende structures lack the inversion cen-

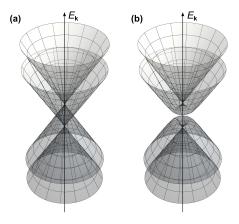


FIG. 1: Energy spectra of HgTe/CdHgTe QWs of (a) the critical and (b) close-to-critical thicknesses. The spectra are plotted after Eq. (3) for (a) $\delta = 0$ and (b) $\delta = \gamma/2$.

ter and the QW structure has a further reduced symmetry compared to the bulk materials even if the interfaces are lattice-matched and defect-free, and the well is symmetric. Indeed, each of the two (001) interfaces possesses a C_{2v} symmetry and taken together the symmetric well has the D_{2d} symmetry. It is known from analogous GaAs/AlAs QW structures that, in the D_{2d} group, the E1 and HH1 states transform according to the same spinor representations [14–16]. Therefore, the coupling matrix element between E1 and HH1 is nonzero, and these subbands must anticross rather than cross at zero in-plane wave vector \mathbf{k} . As pointed out in Refs. [17–20] in the case of HgTe/CdTe QWs a reduction in symmetry leads to the splitting of states at $\mathbf{k} = 0$. However, a theory identifying the source of the splitting (bulk vs inter-

face) and predicting its magnitude has been lacking. Experimental data on weak localization and Shubnikov-de Haas oscillations also indicate strong spin-orbit splitting of the states [21]. Here, we present a microscopic theory of the Dirac states in HgTe/CdTe QWs that predicts a very large ($\sim 15 \text{ meV}$) anticrossing gap between the tips of the Dirac cones in QWs of the critical thickness. We find that this splitting is predominantly due to the interfacial E1-HH1 repulsion mandated by the physical D_{2d} symmetry which is missed by naive continuum-medium considerations but seen when theory acquires atomic resolution. Using this picture we further provide a detailed analysis of the fine structure of Dirac states, which is a key to understanding transport phenomena. As the main result, Fig. 1 shows the energy spectra in QWs of (a) the critical thickness, $d = d_c$, and (b) close-to-critical thickness, $d \neq d_c$. Even in symmetric structures of the critical thickness, the Dirac states are split at the zone center and the spectrum consists of two cones shifted vertically with respect to each other. For $d \neq d_c$, the spectrum becomes more complex: It has a gap and extrema on a circle in the momentum space. We analyze the nature of anticrossing, obtain a quantitative description of the energy spectrum and discuss the consequences of the splitting on transport and optical properties of QWs.

Mechanisms leading to the splitting in QWs with symmetric confinement potential originate fundamentally from bulk inversion asymmetry of the host crystal and interface inversion asymmetry. The relative importance of these contributions cannot be deduced from model Hamiltonian consideration. Atomistic descriptions, on the other hand, capture accurately the relevant asymmetry via the specification of atomic types and positions.

Figures 2 and 3 show the results of atomistic calculations of the energy spectrum of HgTe/CdTe QWs obtained in the screened plane-wave pseudopotential and tight-binding theories, respectively. The pseudopotential method is described in Ref. [10]; details of the tightbinding calculations are given in Supplementary Material. The subband arrangement as a function of the QW width at k = 0 is presented in Figs. 2a, 2b and 3a. Both the pseudopotential and tight-binding calculations yield a wide anticrossing gap between the electron-like E1 and heavy-hole HH1 subbands at the Γ point. The two approaches give different values of the critical QW width because of the sensitivity of the subband structure to the model. Indeed, the pseudoptential calculation reveals for the QW structures interface-localized bands located energetically between E1 and the HH1 state, see Ref. [10]. Nevertheless, all calculations that acknowledge the atomistic symmetry – as opposed to a continuum view – do give large anticrossing at the critical QW thickness. They both predict the gap of about 15 meV far exceeding the estimate of a few meV due solely to the bulk inversion asymmetry [18, 19] and unambiguously indicating that the subband mixing is dominated by the interface contribution, in a crutial difference with the naive $\mathbf{k} \cdot \mathbf{p}$ model. Moreover, despite the existence of an additional inter-

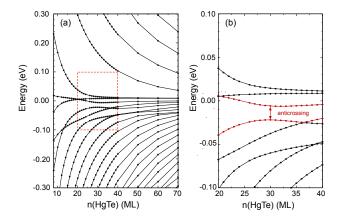


FIG. 2: (a) Arrangement of energy subbands in (001) $(\mathrm{HgTe})_n/(\mathrm{CdTe})_{40}$ QW structures as a function of the number of HgTe monolayers (ML) obtained by the pseudopotential method. (b) Zoom in of the anticrossing area.

face bands in the pseudopotential calculation, the both models predict the same dispersion of the Dirac states formed from the E1 and HH1 subbands near the anticrossing point.

Figure 3b demonstrates the energy dispersion $E(\mathbf{k})$ of the coupled E1 and HH1 states calculated by the tightbinding method for the well width of 16 monolayers which is very close to the critical thickness. The exact condition $d = d_c$ (where $\delta = 0$) cannot be fulfilled in structures with an integer number of monolayers. The spectrum consists of four almost linearly dispersed branches which are split at k=0. The slope of the branches yields the velocity 6.1×10^7 cm/s which is close to the electron velocity 7.2×10^7 cm/s [13] and 6.4×10^7 cm/s [22] determined by cyclotron resonance in HgTe/HgCdTe QWs of critical thickness. The two middle-energy branches anticross at the finite wave vector $k_0 \approx 0.17 \times 10^6 \text{ cm}^{-1}$ with the energy gap being highly sensitive to the deviation of QW width d from d_c . In QWs with $d \approx d_c$, Fig. 3b, this gap is far smaller than the splitting at k=0.

Inspired by the atomistic results we now present an effective model Hamiltonian – the atomically inspired $k \cdot p$ model (AIKP) – which takes into account the correct D_{2d} symmetry of the quantum well. We limit the AIKP model to four basis Bloch states functions forming the Dirac states. Such an approach is valid as long as the anticrossing gap at k = 0 is smaller than the energy separation from the coupled E1-HH1 subbands to other excited subbands. The basis functions have the form [8]

$$|E1, +1/2\rangle = f_1(z)|\Gamma_6, +1/2\rangle + f_4(z)|\Gamma_8, +1/2\rangle ,$$

$$|HH1, +3/2\rangle = f_3(z)|\Gamma_8, +3/2\rangle ,$$

$$|E1, -1/2\rangle = f_1(z)|\Gamma_6, -1/2\rangle + f_4(z)|\Gamma_8, -1/2\rangle ,$$

$$|HH1, -3/2\rangle = f_3(z)|\Gamma_8, -3/2\rangle ,$$
(1)

where $f_1(z)$, $f_3(z)$, and $f_4(z)$ are the envelope functions, z is the growth direction, $|\Gamma_6, \pm 1/2\rangle$, $|\Gamma_8, \pm 1/2\rangle$, and $|\Gamma_8, \pm 3/2\rangle$ are the basis functions of the Γ_6 and Γ_8 states

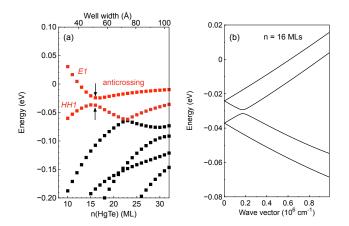


FIG. 3: (a) Arrangement of energy subbands in (001) $(\text{HgTe})_n/\text{CdTe}$ QW structures as a function the number of HgTe monolayers obtained by the sp^3 tight-binding method. (b) Electron dispersion $E(\mathbf{k})$ in the 16-ML-wide QW.

at the Γ point of the Brillouin zone, respectively. The 4×4 effective Hamiltonian describing the coupling at a finite in-plane wave vector \mathbf{k} can be constructed by using the theory of group representations. Taking into account that, in the D_{2d} point group, the states $|E1, \pm 1/2\rangle$ and $|HH1, \mp 3/2\rangle$ transform according to the spinor representation Γ_6 while the wave vector components k_x, k_y belong to the irreducible representation Γ_5 , one derives the effective Hamiltonian to first order in the wave vector as

$$H = \begin{pmatrix} \delta & iAk_{+} & 0 & i\gamma \\ -iAk_{-} & -\delta & i\gamma & 0 \\ 0 & -i\gamma & \delta & -iAk_{-} \\ -i\gamma & 0 & iAk_{+} & -\delta \end{pmatrix} , \qquad (2)$$

where A, δ , and γ are linearly independent parameters, $k_{\pm} = k_x \pm \mathrm{i} k_y$, $x \parallel [100]$ and $y \parallel [010]$ are the in-plane axes. The value 2δ stands for the energy spacing between the E1 and HH1 subbands in the absence of mixing; δ can be tuned from positive to negative value by varying the QW thickness d. In particular, $\delta = 0$ for the critical thickness $d = d_c$ [11]. The parameter A determines the velocity of Dirac fermions. In the $\mathbf{k}\cdot\mathbf{p}$ model, it is given by $A = (P/\sqrt{2})\int f_1(z)f_3(z)dz + \delta A$, where P is the Kane matrix element and δA stands for the contributions from remote bands. Finally, γ describes the coupling of E1 and HH1 states at $\mathbf{k} = 0$ in zinc-blende-lattice QWs [18, 19], $2|\gamma| \approx 15$ meV as it follows from Figs. 2 and 3 neglecting the influence of additional interface states.

Solution of the secular equation for the matrix Hamiltonian (2) yields the energy spectrum

$$E_{1,4} = \mp \sqrt{\delta^2 + (Ak + \gamma)^2}, \ E_{2,3} = \mp \sqrt{\delta^2 + (Ak - \gamma)^2}.$$
(3)

The corresponding wave functions are given by

$$\Psi_{1,4} = \frac{1}{2} \begin{pmatrix} a_{1,4} \\ b_{1,4} e^{-i\varphi} \\ -a_{1,4} e^{-i\varphi} \\ b_{1,4} \end{pmatrix}, \ \Psi_{2,3} = \frac{1}{2} \begin{pmatrix} a_{2,3} \\ b_{2,3} e^{-i\varphi} \\ a_{2,3} e^{-i\varphi} \\ -b_{2,3} \end{pmatrix}, \ (4)$$

where $a_{1,2} = -i\sqrt{(E_{1,2} + |\delta|)/E_{1,2}} \operatorname{sign}(Ak \pm \gamma)$, $b_{1,2} = \sqrt{(E_{1,2} - |\delta|)/E_{1,2}}$, $a_{3,4} = i\sqrt{(E_{3,4} + |\delta|)/E_{3,4}}$, $b_{3,4} = \sqrt{(E_{3,4} - |\delta|)/E_{3,4}} \operatorname{sign}(Ak \mp \gamma)$ for $\delta > 0$ and $a_{1,2} = -ib_{1,2}$, $b_{1,2} = ia_{1,2}$, $a_{3,4} = ib_{3,4}$, and $b_{3,4} = -ia_{3,4}$ for $\delta < 0$; $k = |\mathbf{k}|$, and φ is the polar angle of the wave vector \mathbf{k} , $\mathrm{e}^{\mathrm{i}\varphi} = k_+/k$. The coefficients a_j and b_j are defined in such a way that the wave functions (4) are continuous in \mathbf{k} -space.

The electron dispersion (3) is shown in Fig. 1. In the structure of the critical thickness the spectrum consists of two Dirac cones shifted vertically with respect to each other by $2|\gamma|$. At $d \neq d_c$, the gap of $2|\delta|$ opens in the spectrum at the wave vector $k_0 = |\gamma/A|$ and the system behaves as an insulator. The topological class of the insulator is determined by the sign of δ , similarly to the model where interface inversion asymmetry is neglected [8]. To confirm this, we calculate the \mathbb{Z}_2 topological index ν following the procedure described in Ref. [1]. The index ν is determined by the quantities $\zeta(\Lambda_{\alpha})$ ($\alpha=1...4$) calculated at four certain points of the Brillouin zone Λ_{α} which are invariant with respect to the time inversion, $(-1)^{\nu} = \prod_{\alpha} \zeta(\Lambda_{\alpha})$. We find that

$$\zeta(\mathbf{k} = 0) = \lim_{\mathbf{k} \to 0} \frac{\Pr\left[\langle \Psi_i(\mathbf{k}) | \Theta | \Psi_j(\mathbf{k}) \rangle \right]}{\sqrt{\det\left[\langle \Psi_i(\mathbf{k}) | \Theta | \Psi_j(\mathbf{k}) \rangle \right]}} = \operatorname{sign} \delta, (5)$$

where the indices i and j run over the occupied branches 1 and 2, Θ is the operator of time inversion, and $\operatorname{Pf}[A]$ is the Pfaffian of the matrix A. All other $\zeta(\Lambda_{\alpha})$ are determined by the wave functions at the Brillouin zone edge and are unlikely to be affected by a small change of δ . Thus, we conclude that the topological transition occurs at $\delta=0$: The QW structures with $d< d_c$ and $d> d_c$ belong to trivial and topological two-dimensional insulators, respectively. In the phase of topological insulator, one expects the formation of helical edge states leading to the quantum spin Hall effect and conductivity quantization [9] although the electron structure of the edge states is modified by strong interface-induced coupling.

The large anticrossing of the E1 and HH1 subbands at $\mathbf{k} = 0$ revealed in the atomistic calculations indicates that the subband mixing mainly originates from the interface inversion asymmetry related to the anisotropy of chemical bonds. Since the E1 subband is formed from the electron and light-hole basis functions and the HH1 subband is of the heavy-hole type, see Eq. (1), they are efficiently coupled due to heavy-hole-light-hole mixing at the interfaces, the effect known for zinc-blende-lattice QW structures [14–16, 23, 24]. This short-range mixing can be modeled by introducing interface terms to the effective Luttinger Hamiltonian which take into account low spatial symmetry of individual interfaces. The additional terms related to for the left (L) and right (R) interfaces have the form $V_{L,R} = \pm \hbar^2 t_{l-h} / (\sqrt{3}a_0 m_0) \{J_x J_y\}_s \delta(z - t_0)$ $z_{L,R}$), where t_{l-h} is the (real) mixing constant, a_0 is the lattice constant, m_0 is the free electron mass, $\{J_xJ_y\}_s =$ $(J_xJ_y+J_yJ_x)/2$, J_x and J_y are the matrices of the angular momentum 3/2, and $z_{L,R}$ are the interface positions.

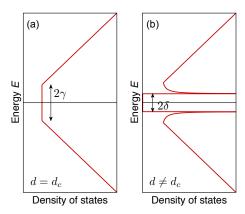


FIG. 4: Sketch of the density of states in HgTe/CdTe QWs of the (a) critical and (b) close-to-critical thinknesses.

The terms lead to the coupling of the E1 and HH1 subband with the parameter

$$\gamma = \frac{\hbar^2 t_{l-h}}{2a_0 m_0} \left[f_3(z_R) f_4(z_R) - f_3(z_L) f_4(z_L) \right] . \tag{6}$$

Since the envelope functions $f_3(z)$ and $f_4(z)$ have opposite parities, the parameter γ is non-zero. Comparing the results of atomistic calculation with the $k \cdot p$ theory we obtain $t_{l-h} \approx 1.5$. Extrapolation to HgTe/Cd_{0.7}Hg_{0.3}Te QWs, the structures commonly used in experiment [9, 12, 13], gives $t_{l-h} \approx 1.1$ and $2|\gamma| = 10$ meV.

The splitting of Dirac states may affect many phenomena, including weak localization, Shubnikov-de Haas oscillations and quantum Hall effect, spectra of THz radiation absorption, spin-flip Raman scattering, etc. The basic characteristic of electron systems is the single-particle density of states

$$\rho(E) = \sum_{l,k} \delta[E - E_l(k)]. \tag{7}$$

For the QW system under study, it has the form

$$\rho(E) = \frac{|E|}{\pi A^2} \times \begin{cases} 0, & \text{if } |E| \le |\delta|, \\ |\gamma|/\sqrt{E^2 - \delta^2}, & \text{if } |\delta| < |E| \le \sqrt{\gamma^2 + \delta^2}, \\ 1, & \text{if } |E| > \sqrt{\gamma^2 + \delta^2}. \end{cases}$$

Figure 4 illustrates the density of states vs. electron energy for QWs of the critical and close-to-critical thicknesses. The gapless structure behaves as a two-dimensional semimetal with the finite density of states in the whole energy range, Fig. 4a. The density of states linearly scales with the energy $\rho(E) = |E|/(\pi A^2)$ at $|E| > |\gamma|$ and is energy independent, $\rho(E) = |\gamma|/(\pi A^2)$, at $|E| < |\gamma|$. In the latter region, electrons and holes coexist and the conductivity is bipolar. In the QWs of close-to-critical thicknesses, Fig. 4b, the density of states has a gap of $2|\delta|$ and van Hove singularities at $E = \pm \delta$.

The splitting of Dirac cones leads to a beating pattern in the Shubnikov-de Haas oscillations as well as splitting of the absorbance peak in experiments on cyclotron

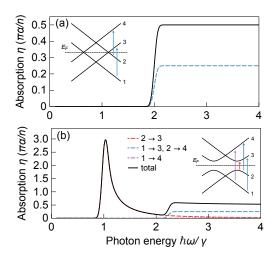


FIG. 5: Photon energy dependence of the QW absorbance for the structures of (a) the critical thickness, $\delta=0$, and (b) the close-to-critical thickness, $\delta=0.5\gamma$. Dashed curves show partial contributions to the absorbance. Inset sketches the energy spectrum and allowed optical transitions. The steep spectral edges are smoothed by convoluting the spectra with the Gauss function with the standard deviation of 0.05γ .

resonance. The peak positions are determined by the effective cyclotron masses $m_l = \hbar^2 k/(dE_l/dk)$. In n-doped structure with the Fermi energy E_F , one can expect the splitting of resonant line into two components, their positions at $E_F \gg |\gamma|$ being determined by the effective masses $m_{3,4} = (\hbar/A)^2 (E_F \pm \gamma)$.

The energy spectrum can be also studied by means of optical spectroscopy. For direct optical transitions between occupied and empty states, the QW absorbance at the normal incidence of radiation is given by

$$\eta = \frac{4\pi^2 \alpha \,\hbar}{\omega n_{\omega}} \sum_{l,m,k} |\mathbf{e} \cdot \mathbf{v}_{ml}|^2 \,\delta(E_m - E_l - \hbar\omega) \,, \quad (8)$$

where α is the fine-structure constant, ω is the light frequency, n_{ω} is the refractive index of the medium, e is the (complex) unit vector of the light polarization, and $\mathbf{v}_{ml} = \hbar^{-1}(\partial H/\partial \mathbf{k})_{ml}$ is the matrix element of the velocity operator. We consider undoped QW structures where the optical transitions can occur between the occupied branches 1 and 2 and the empty branches 3 and 4 of the energy spectrum, see insets in Figs. 5a and 5b. For normally-incident radiation, the QW absorbance is independent of the light polarization and has the form

$$\eta = \frac{\pi \alpha A^2}{\hbar \omega n_{\omega}} \sum_{\substack{l=1,2\\m=3,4}} \sum_{k_t} \frac{k_t f_{ml}(k_t)}{|dE_m/dk - dE_l/dk|_{k=k_t}} , \qquad (9)$$

where k_t is the wave vector at which the optical transitions between the branches l and m occur, it is found from the equation $E_m(k_t) - E_l(k_t) = \hbar \omega$, and $f_{ml}(k)$ are the dimensionless functions defined by $f_{ml}(k)$

 $(\hbar/A)^2 |\boldsymbol{e} \cdot \boldsymbol{v}_{ml}|^2$. Straightforward calculations yield

$$f_{41} = \frac{\delta^2}{E_4^2} , \quad f_{32} = \frac{\delta^2}{E_3^2} , \qquad (10)$$

$$f_{31} = f_{42} = \frac{1}{2} \left(1 - \frac{A^2 k^2 - \gamma^2 + \delta^2}{E_1 E_3} \right) .$$

Figures 5a and 5b demonstrate the photon energy dependence of the QW absorbance for structures of the critical and close-to-critical thicknesses. In the structure with two split Dirac cones, Fig. 5a, the absorption spectrum has a step-like shape with the edge at $\hbar\omega = 2|\gamma|$ although there is no band gap in the energy spectrum. Such a behavior is dictated by the selection rules and energy conservation law: Direct optical transitions in QWs of the critical thickness are allowed only between the branches $1 \rightarrow 3$ and $2 \rightarrow 4$ and these transitions can occur at $\hbar\omega \geq 2|\gamma|$. In QWs of non-critical thickness, Fig. 5b, the branches 2 and 3 anticross at finite wave vector, and the direct optical transitions between them get allowed. It leads to the emergence of an additional sharp band in the absorption spectrum at $\hbar\omega = 2|\delta|$. The spectral shape of this absorption band is determined by the van Hove singularities in the density of states.

To summarize, we have described the splitting of Dirac states in HgTe/CdTe quantum wells of critical and close-to-critical thicknesses. In structures of the critical thickness, the splitting between the Dirac cones reaches the value of 15 meV and is dominated by symmetry-enforced light-hole–heavy-hole mixing at the quantum well interfaces. These structures behave as a two-dimensional semimetal with non-vanishing density of states in the whole energy range. In quantum wells of close-to-critical thicknesses, a gap opens at a finite in-plane wave vector, which leads to the emergence of extremum circles in the electron dispersion and corresponding van Hove singularities in the density of states. We have also discussed the consequences of the level mixing on the optical and transport properties that await testing.

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