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Quantum spin Hall insulator with a large bandgap, Dirac fermions, and bilayer graphene analog

Sergey S. Krishtopenko¹,² and Frédéric Teppe¹*

The search for room temperature quantum spin Hall insulators (QSHIs) based on widely available materials and a controlled manufacturing process is one of the major challenges of today’s topological physics. We propose a new class of semiconductor systems based on multilayer broken-gap quantum wells, in which the QSHI gap reaches 60 meV and remains insensitive to temperature. Depending on their layer thicknesses and geometry, these novel structures also host a graphene-like phase and a bilayer graphene analog. Our theoretical results significantly extend the application potential of topological materials based on III–V semiconductors.

INTRODUCTION
Quantum spin Hall insulators (QSHIs), also known as two-dimensional (2D) topological insulators, are characterized by a bulk bandgap and gapless helical edge channels (1, 2) protected from backscattering by time-reversal symmetry. This fascinating new quantum phase of matter suggests, among others, the design of groundbreaking electronic devices with dissipationless spin currents and the realization of Majorana bound states at the interface with superconductors (3, 4). In this context, QSHI materials with controlled growth and technological processing are in great demand. In addition, a sufficiently large bulk gap is required to overcome the constant constraint of cryogenic temperature operation and the problem of short coherence length of the edge channels. Despite numerous theoretical predictions of a QSHI state in a large number of thin films and 2D materials (5–9), its inherent characteristic—a quantum spin Hall effect (10) with its universal quantized conductance—has been experimentally established by a direct transport measurement only in inverted HgTe quantum wells (QWs) (11) and broken-gap InAs/GaSb QW bilayers (12, 13).

In HgTe QWs, the inverted band structure responsible for the QSHI state is due to the strong spin-orbit interaction in the HgTe layer. The position of the electron- and hole-like subbands therefore depends on the QW width in such a way that QWs wider than a critical value $d_c$ have inverted subbands (14), whereas QWs thinner than $d_c$ have a non-inverted band ordering and feature a trivial band insulator (BI) state. At the critical thickness, HgTe QWs experience a topological phase transition and host a single-valley spin-degenerate Dirac cone at the $\Gamma$ point of the Brillouin zone (15). The maximum bulk gap achievable in inverted HgTe QWs grown on CdTe does not exceed 16 meV, but a gap of 60 meV was recently reached (16) in compressively strained HgTe QWs. However, this large gap is only available at low temperatures, whereas increasing of temperature induces the gap closing and phase transition into trivial state (17, 18). The latter is caused by strong temperature dependence of the energy of the $\Gamma_s$ bulk band in HgCdTe materials (19). However, the high-quality HgTe-based QSHIs grown by molecular beam epitaxy (MBE) are not widely accessible. We also note that the weak bonding between Hg and Te makes these toxic species highly volatile and hazardous during processing.

Although InAs/GaSb broken-gap QWs are also made of toxic compounds, their bonds are stronger than those of HgTe, and the material is therefore stable and less dangerous. In addition, these MBE-grown III–V semiconductors having a controlled technological process are accessible worldwide. However, the bulk gap in inverted InAs/GaSb QW bilayers is much smaller (12) than that in HgTe QWs. The band inversion in these broken-gap structures embedded between AlSb barriers results from the fact that the valence band edge of GaSb is higher than the conduction band edge of the InAs layer. Because the InAs/GaSb QW bilayer does not have inversion symmetry in the growth direction, the crossing of electron- and hole-like subbands arises away from the $\Gamma$ point of the Brillouin zone (20). Because these levels are mixed at nonzero quasi-momentum $\mathbf{k}$, the band inversion opens a hybridization gap of a few millielectron volts in the QW bilayer.

Since the first experimental demonstration of the QSHI state in the InAs/GaSb QW bilayers, some attempts have been made to overcome the problem of the small inverted bandgap (13). Lately, its value has been enhanced by replacing the GaSb layer of the composite QW with a GaInSb alloy. However, the maximum value reached in this way was only on the order of 20 meV (21). Thus, the InAs/Ga(In)Sb QW bilayers seem to be less favorable for potential applications in topological electronics than the HgTe-based QWs, despite their many previously mentioned benefits and their promising physical properties, including a good interface with superconductors (22, 23) and an in situ electric tunability (24). Besides the hybridization gap opening at nonzero $\mathbf{k}$, another consequence of the structure inversion asymmetry in the InAs/GaSb QW bilayers is that unlike HgTe QWs, they do not feature a Dirac cone across topological phase transitions.

Here, we introduce a new class of multilayer InAs/GaSb QWs, which differ from conventional QW bilayers by their band crossing at the center of the Brillouin zone. To eliminate the structure inversion asymmetry, we attach an additional InAs or GaSb layer to the InAs/GaSb QW bilayer. We now define that the novel three-layer InAs/GaSb QWs have an “InAs geometry” or a “GaSb geometry” if they contain two symmetrical layers of InAs or GaSb, respectively. First, we show that the phase transitions in the QWs with InAs geometry are similar to those of HgTe/CdTe QWs. In particular, depending on the layer thicknesses, our structures allow the realization of massless Dirac fermions, semimetal (SM), and a QSHI state with a maximum gap of 16 meV. Second, we show that the QWs with GaSb geometry partially hold the properties of two tunnel-coupled HgTe QWs (25), such as the specific band spectrum of graphene bilayer, its “unconventional” quantum Hall effect (26), and its electrically tunable bandgap. Finally, we reveal that strain engineering of the three InAs/GaInSb QWs with InAs geometry allows the realization of a QSHI with a bandgap greater than 60 meV, comparable to that of strained HgTe QWs (16), but nearly insensitive to temperature.

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RESULTS
Three-layer InAs/GaSb QWs with InAs geometry
Phase diagram
We first consider symmetrical InAs/GaSb/InAs QWs confined by outer AlSb barriers. The QW is supposed to be grown on (001) GaSb buffer. Because the electron-like subbands are localized in the InAs layers, whereas the hole-like subbands are localized in the GaSb layers, the InAs/GaSb/InAs QWs can be contingently considered as “double QW for electrons” with a GaSb middle barrier, which also plays a role of “QW for holes” (Fig. 1A). Figure 1B shows positions of electron-like (E1 and E2) and hole-like (H1) subbands at zero quasi-momentum k as a function of InAs-layer thickness d1 for GaSb-layer thickness of d2 = 4 nm. In this case, it is intuitively clear that the electron-like levels E1 and E2 are connected with even-odd state splitting, arising from the tunnel-coupled “QWs for electrons”. If InAs layers are thin (d1 < d1c, where d1c depends on d2), the E1 and E2 subbands lie above the hole-like subbands, and the three-layer QW has a trivial band ordering. For d1 > d1c, the E1 subband lies below the H1 level, and the QW is characterized by inverted band structure.

In Fig. 1C, we provide a phase diagram for the QWs of InAs geometry with different layer thicknesses. Typical band dispersions for each phase are shown in the bottom panels. In the diagram, the solid curve describing the crossing between E1 and H1 subbands divides the d1-d2 plane into a white region, corresponding to trivial BI phase with direct band ordering, and a gray region with inverted band structure. The gray region, in its turn, is split into open and striped areas, corresponding to QSHI state and SM, respectively. As shown in Fig. 1F, the SM phase is characterized by a vanishing indirect bandgap when the side maxima of the valence subband exceed in energy the conduction subband bottom. We note that the QWs grown along (001) direction possess side maxima in the valence band that are independent of d1 and d2 and always lie along (110), (110), (110), and (110) crystallographic directions. Thus, by varying the layer thicknesses in the QW with InAs geometry, one can realize BI, QSHI, or SM phases.

To describe a quantum phase transition between BI and QSHI states, we derive effective a 2D Hamiltonian valid for the states in the vicinity of the Γ point. Distinct from HgTe/CdTe QWs (2), we take into account additional E2 subband, lying close in energy to the E1 and H1 states. In the basis set {E1+, H1+, E2-, E1-, H1-}, and {E2+}, the 2D Hamiltonian for the QW of InAs geometry has the form

\[ H_{\text{InAs}}(k_x, k_y) = \begin{pmatrix} H_1(k_x, k_y) & 0 \\ 0 & H_1^*(-k_x, -k_y) \end{pmatrix} \]

where the asterisk stands for complex conjugation, kx and ky are momentum k components in the plane, k = kx ± iky, and H1(kx, ky) is written as

\[ H_1(k_x, k_y) = \begin{pmatrix} e_{E1}(k) & -Ak_x + Sk_y & 0 \\ -Ak_x - Sk_y & e_{E1}(k) + Rk_x^2 & e_{E2}(k) \\ 0 & e_{E2}(k) & e_{E1}(k) + Rk_y^2 \end{pmatrix} \]

Here, C, M, A, B_{E1}, B_{H1}, B_{E2}, Δ_{E1E2}, R, and S are structure parameters, which depend on d1 and d2. Parameter Δ_{E1E2} defines the gap between the E1 and E2 subbands at k = 0. Because we have kept the inversion symmetry and axial symmetry around the growth direction, H_{InAs}(k, k) has a block-diagonal form. The mass parameter M describes inversion between the E1 and H1 subbands: M > 0 corresponds to a trivial BI state, whereas for a QSHI state, M < 0. By applying the open boundary condition for H_{InAs}(k, k) along the y axis, one can numerically calculate dispersion of helical states, arising at the sample edge at M < 0. For instance, the edge state dispersions for the QSHI state at the layer thicknesses marked in Fig. 1C by the open blue symbol are provided in Fig. 2. The structure parameters for H_{InAs}(k, k) are given in the Supplementary Materials.

Quantum spin Hall insulator
The essential characteristic of a QSHI is the bandgap values for the bulk states. In the conventional broken-gap QW bilayers, the inverted bandgap opens away from the Γ point of the Brillouin zone because of the interaction between E1 and H1 subbands at nonzero quasi-momentum. In the case of three-layer InAs/GaSb QWs, the inverted bandgap arises in the Γ point because of quantum confinement, whereas the E1-H1 interaction vanishes at k = 0. Because the confinement effect at k = 0 is much stronger than the E1-H1 interaction at nonzero k, one can expect that the bandgap in the QSHI state is higher in the three-layer QWs than in InAs/GaSb QW bilayers.

Figure 2A shows the bandgap in the QSHI state as a function of d1 for d2 = 14 monolayers (MLs) of GaSb. Here, one ML in the given QW layer corresponds to a half of a lattice constant a0 in the bulk material. The black and red curves correspond to the structures grown on InAs and GaSb buffers, respectively. It is seen that, starting from d1 > d1c, increasing of d1 enhances the bandgap, which occurs between the H1 and E1 levels. When the bandgap approaches Δ_{E1E2} defined by

\[ Δ_{E1E2} = \max (Δ_{E1}, Δ_{E2}) \]

where Δ_{E1} and Δ_{E2} are the gaps in the conventional E1 and E2 subbands, one can realize a quantum phase transition between BI and QSHI states.
the dispersion of the edge states, obtained by numerical diagonalization of like subbands are shown in blue and red, respectively. The black curves correspond to layer thicknesses marked in Fig. 1C by blue open symbols. Electron- and heavy hole inset shows the maximum gap, which can be achieved at a given value of $d_2$. The red curves correspond to the QWs grown on InAs and GaSb buffers, respectively. The dotted vertical lines mark the values of 14 MLs, where one ML = 0.5.

**Robustness of QSHI in an external magnetic field**

A reliable fingerprint of QSHI under applied magnetic field $B$ is the characteristic behavior of a particular pair of Landau levels (LLs), the so-called zero-mode LLs (11). The origin of this pair of LLs becomes apparent when using an effective $4 \times 4$ Hamiltonian for $E1$ and $H1$ subbands (2). Below a critical field value $B_c$, the lowest zero-mode LL has an electron-like character and arises from the valence band, whereas the highest zero-mode LL has a heavy hole–like character and splits from the conduction band (see Fig. 2D). In this case, the topological edge states are still present, although they are no longer protected by time-reversal symmetry (28, 29), whereas for larger magnetic fields $B > B_c$, the band ordering becomes normal, and only a trivial state can be found. At the critical magnetic field $B_c$, corresponding to the crossing of zero-mode LLs, the quantum phase transition between the QSHI and BI states arises.

By neglecting the influence of $E2$ subbands in $H_{\text{InAs}}(k_x, k_y)$, we can roughly evaluate the critical magnetic field as (11)

$$B_c \approx \frac{\hbar c}{e} \frac{M}{B_{H1} - B_{E1}} \tag{3}$$

where $e$ is an elementary charge, $\hbar$ is a Plank constant, and $c$ is a velocity of light. For instance, by using the structure parameters for the QW with $d_1 = 38$ MLs and $d_2 = 14$ MLs given in the Supplementary Materials, $B_c \sim 10$ T.

Figure 2E provides dependence of $B_c$ on the thickness of InAs layers for the QWs grown on InAs and GaSb buffers calculated by using the eight-band Kane model (30). One can see that the critical magnetic field in three-layer InAs/GaSb QWs can reach 20 T, whereas the maximum values of $B_c$ in HgTe/CdTe QWs grown on CdZnTe buffer do not exceed 10 T (11).

**Three-layer InAs/GaSb QWs with GaSb geometry**

**Phase diagram**

Let us now consider symmetrical GaSb/InAs/GaSb QWs sandwiched between AlSb barriers and grown on (001) GaSb buffer. In contrast to the QWs with InAs geometry, GaSb/InAs/GaSb QWs contain two tunnel-coupled “QWs for holes” with InAs layer as a “middle barrier” (Fig. 3A). We note that the InAs layer of reasonable thicknesses ($d_1 > 2$ nm) is not transparent for the particles from the hole-like subbands because of large effective mass. Therefore, the hole-like subbands localized in different GaSb layers are decoupled at $k = 0$. However, the mixing between heavy hole– and electron-like states at nonzero $k$ leads to the splitting between $H1$ and $H2$ subbands, which can also be contingently considered as odd and even combinations of the hole states in each GaSb layer.
The effective Hamiltonian is written as:

$$H_{GaSb}(k_x, k_y) = \begin{pmatrix} H_2(k_x, k_y) & 0 \\ 0 & H_2^*(-k_x, -k_y) \end{pmatrix}$$

where

$$H_2(k_x, k_y) = \begin{pmatrix} e_{H1}(k) - Ak & Rk^2 \\ -Ak & e_{H1}(k) \end{pmatrix}$$

$$H_2^*(-k_x, -k_y) = \begin{pmatrix} e_{H2}(k) & Rk^2 \\ 0 & e_{H2}(k) \end{pmatrix}$$

Here, the terms $$e_{E1}(k)$$ and $$e_{H1}(k)$$ have the same form as in Eq. 2, and $$H_{GaSb}(k_x, k_y)$$ results from keeping the inversion and axial symmetry around the growth direction. The structure parameters and comparison with band structure calculations based on the eight-band Kane model are given in the Supplementary Materials as are the structure parameters for $$H_{GaSb}(k_x, k_y)$$.

### BG phase

As mentioned above, the band inversion in the QWs with GaSb geometry induces a specific metal phase, in which the band structure is very similar to the band structure of natural BG (Fig. 4A). Here, we show that this BG phase also shares other properties of the BG.

One of the characteristics of natural BG is the unconventional quantum Hall effect (26). For natural BG, plateaus in the Hall conductivity $$\sigma_{xy}$$ occur at integer multiples of $$4e^2/h$$, where the level degeneracy $$g = 4$$ arises from the spin and valley degrees of freedom. Deviation from the conventional case occurs in the vicinity of the charge neutrality point, where there is a step in $$\sigma_{xy}$$ of $$8e^2/h$$, arising from the eightfold degeneracy of the zero-energy LL. In the BG phase under perpendicular magnetic field, a specific LL with degeneracy two times higher than other LLs also arises. This zero-mode LL, shown in Fig. 4B by the bold red curve (the h1 and h2 levels), has a pure hole-like character and is formed by the states in the vicinity of the bottom and top of the H1 and H2 subbands. Because they are touched at $$k = 0$$, the zero-mode LL has a doubled degeneracy. The latter can be analytically shown by using an effective 2D Hamiltonian $$H_{GaSb}(k_x, k_y)$$. The doubled degeneracy of the zero-mode LL requires two times more carriers to fill it, so the transition between the corresponding plateaus should be twice as wide in density as others, and the $$\sigma_{xy}$$ step between the plateaus are expected to be $$2e^2/h$$ instead of $$e^2/h$$.

In natural BG, a bandgap can be opened by breaking the inversion symmetry, for instance, by electric field applied perpendicularly to the sample plane. The QWs with GaSb geometry also holds this property. Figure 4C shows the band dispersion in a perpendicular electric field of 5 kV/cm. Even in this case, the dispersion curves are very similar to the band structure of natural BG in an external electric field (31). However, the strong spin-orbit interaction in the QWs with GaSb geometry removes the spin degeneracy at nonzero quasi-momentum due to the Rashba effect (32). Figure 4D shows the bandgap $$\Delta$$ values in the BG phase as a function of applied electric field. The bandgap is electrically tunable because it is in natural BG, although its dependence on an electric field in the InAs/GaSb QWs has nonmonotonic behavior. Note that an inversion asymmetry in the QWs with GaSb geometry also splits the zero-mode LLs, removing this doubled degeneracy order. Therefore, we anticipate the recovery of the equidistant sequence of plateaus in the Hall conductivity discussed above, such as for gate-biased natural BG (33).

Previously, the BG phase was predicted for two tunnel-coupled HgTe QWs (25). Here, we show the existence of this phase in the...
For the QWs with InAs geometry, the latter corresponds to the limit of large $\Delta_{B12}$. Therefore, $H_1(k_x, k_y)$ can be projected on the subspace, containing the $E1$ and $H1$ states. Because we are not interested in terms quadratic in $k$, the projection is performed by simply eliminating the third row and column of the matrix in Eq. 2 and by setting $B_{E1}$ and $B_{H1}$ to zero. In the case of the QWs with GaSb geometry, keeping linear in $k$ terms in $H_2(k_x, k_y)$ results to decoupling of the $H2$ subband from the $E1$ and $H1$ states. Therefore, the block describing the $E1$ and $H1$ subbands can be considered separately.

Without loss of generality, we can set $C = 0$ and the 2D Hamiltonian for the states in the vicinity of $k = 0$ in the crossed $E1$ and $H1$ subbands ($M = 0$), for both types of the QWs takes the form

$$H_{12}^{E1+H1}(k_x, k_y) \equiv \begin{pmatrix} 0 & \hbar v_F k_x \\ \hbar v_F k_y & 0 \end{pmatrix}$$

The similar Hamiltonian can be also derived for $H_1^*(-k_x, -k_y)$ and $H_2^*(-k_x, -k_y)$. It is seen that Eq. 6 corresponds to a massless Dirac Hamiltonian with Fermi velocity $v_F$, defined by parameter $A$ in Eqs. 2 and 5.

Figure 5 shows the band dispersion for the QWs of both geometries, when the $E1$ and $H1$ subbands are crossed. The layer thicknesses correspond to the crossing of the $E1$ and $H1$ subbands shown in Figs. 1B and 2B. Because the hybridization between $E1$ and $H1$ subbands vanishes at $k = 0$, it results in the gapless state with 2D massless Dirac fermions, just like in HgTe/CdTe QWs with critical thicknesses (15). The linear band dispersion at small $k$ is seen. However, in the QWs with GaSb geometry, the Dirac cone is accompanied by an additional decoupled “massive” branch of $H2$ subband arising at the crossing point that is similar to the case of two tunnel-coupled HgTe QWs (25).

The bottom panels in Fig. 5 provide $v_F$ for massless Dirac fermions as a function of $d_z$ in the QWs with InAs and GaSb geometries. It is seen that Fermi velocity is varied from $1 \times 10^5$ to $3 \times 10^5$ m/s, depending on the GaSb layer thicknesses. These values are several times lower than the ones in graphene ($\sim 10^6$ m/s) and HgTe/CdTe QWs (15).

Finally, we stress the effect of strain on $v_F$ of the massless particle, arising because of lattice-mismatch constant in the QW layers and buffers. It is seen that increasing tensile strain of InAs layers ($e_t > 0$) in the QWs with both geometries enhances $v_F$. As we have discussed previously, at high values of $e_t$, the band inversion drives the QWs with InAs geometry from BI into SM phase, avoiding the QSHI state. However, in this case, the Dirac cone in the $\Gamma$ point of the Brillouin zone arising at critical layer thicknesses is accompanied by the side maxima of the valence band overlapping with the conduction band bottom. We note that, in the QWs with GaSb geometry, the band inversion does not occur at high values of $e_t$. For instance, the QWs grown on AlSb buffer have trivial band ordering at all layer thicknesses.

**Large-gap QSHI in three-layer QWs with InAs geometry**

We have demonstrated that the three-layer QWs with InAs geometry and HgTe/CdTe QWs share the same possible phases and feature the comparable bandgaps in the QSHI state. As has been demonstrated recently (16), compressive strain in HgTe QWs extends the bandgap of QSHI up to 60 meV at reasonable values of the strain. Now, we address the question of whether the bandgap of the QSHI state in three-layer InAs/GaSb the QWs can be enhanced up to the same values of compressively strained HgTe QWs. However, if either compressive or tensile

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**Fig. 4. BG phase in three-layer InAs/GaSb QWs with GaSb geometry.** (A) Band dispersions for a BG phase in the QW grown on GaSb buffer at $d_z = 10$ nm and $d_x = 6$ nm. Electron- and hole-like subbands are shown in blue and red, respectively. Solid and dotted curves correspond to different spin states. (B) LL fan chart. The zero-mode LL, which has doubled degeneracy order as compared with other levels, is marked by red bold curve (the $h1$ and $h2$ levels). This LL is formed by states of both $H1$ and $H2$ subbands. LL, containing only the states from the $E1$ subband in high magnetic fields, is given in blue. The crossing between the $e1$ and $h1$ LLs, arising at critical magnetic field $B_c = 3.7$ T, leads to the phase transition into normal (noninverted) band structure, as it is in single HgTe QW (11). (C) Band dispersions in an electric field of 5 kV/cm oriented perpendicular the QW plane. (D) Bandgap as a function of an applied electric field. (E) Band dispersion for a BG phase, calculated on the basis of $H_{\text{GaSb}}(k_x, k_y)$ with the layer thicknesses marked in Fig. 3C by blue open symbol. The black curves correspond to the dispersion of the edge states, obtained by numerical diagonalization of $H_{\text{GaSb}}(k_x, k_y)$ with open boundary conditions along the $y$ axis. Different Kramers partners are shown by solid and dotted curves.
strain is large enough, it destroys the QSHI state in three-layer InAs/GaSb QWs and induces a phase transition into the BI or SM phase, respectively (see Fig. 2B).

To increase the inverted bandgap in the InAs-designed QWs, we therefore replace the middle GaSb layer in the QW by a Ga1−xInxSb alloy. Previously, Smith and Mailhiot (34) have found that this replacement significantly enhances the inverted bandgap in strained InAs/GaInSb superlattices (SL). Recently, this idea has been applied to the InAs/GaInSb QW bilayers (36), in which the bandgap can be increased up to 20 meV.

Figure 6A shows the bandgap in the QSHI state in three-layer InAs/Ga0.6In0.4Sb QW with InAs geometry as a function of $d_1$ at $d_2 = 14$ MLs. The black, red, and blue curves correspond to the structures grown on GaSb, AlSb, and Ga0.68In0.32Sb buffers, respectively. It is seen that the inverted bandgap in the strained InAs/Ga0.6In0.4Sb QW is several times larger than the values obtained for the three-layer InAs/GaSb QWs. The inset shows the maximum gap in the strained QW grown on Ga0.68In0.32Sb buffer, which can be achieved at given value of $d_2$.

Figure 6B provides the QSHI bandgap in the InAs/Ga0.6In0.4Sb QWs with different values of $d_1$ and $d_2 = 11$ MLs as a function of strain InAs layers $e_1$ and Ga0.6In0.4Sb layer $e_2$. Note that the values of $e_1$ and $e_2$ are connected via the lattice constant of the buffer material. Figure 6B proves theoretically that, by adjusting the strain for different values of $d_1$ and $d_2$, one can obtain a QSHI with a large bandgap. It is seen that the InAs/Ga0.6In0.4Sb QW with $d_1 = 27$ MLs and $d_2 = 11$ MLs grown on Ga0.68In0.32Sb buffer features a bandgap in the QSHI state of about 60 meV. The band structure for this QW is shown in Fig. 6C. We have considered the case of symmetrical InAs/GaInSb/InAs QWs. We note that the bandgap in these QWs can be tuned by varying the strength of an applied electric field or by growing the InAs layers of different thicknesses. However, the maximum QSHI bandgap in the three-layer InAs/Ga0.6In0.4Sb QWs can be achieved only in symmetrical geometry (see the Supplementary Materials).

In addition to large-gap QSHI, the InAs/Ga0.6In0.4Sb QWs at critical thicknesses feature massless Dirac fermions with Fermi velocity (up to $7 \times 10^7$ m/s) higher than in the InAs/GaSb/InAs QWs. Moreover, a linearity of their energy dispersion persists in much higher energy ranges. The latter can be seen by comparing Figs. 6D and 5A.

**DISCUSSION**

To summarize, we have considered realization of phases in multilayer InAs/Ga(In)Sb QWs containing broken-gap band alignment at the two interfaces. We note that our results are general and valid for any type II QW heterostructures. So far, multilayer type II heterostructures based on InAs, GaSb, InSb, AlSb, and their alloys have been interesting in view of laser and detector applications in mid-infrared (IR) and terahertz range. We believe that theoretical predictions performed here make the multilayer type II QWs attractive for realistic pseudorelativistic electronics and high-temperature QSHI devices.

In particular, MBE growth technology for InAs/GaInSb heterostructures is well developed nowadays. Since the 2000s, it has been widely used for the growth of strained InAs/Ga1−xInxSb SL for mid-IR photodetectors with $x < 0.4$. Significant progress in the MBE technology of InAs/GaInSb, which has been achieved very recently (35, 36), allows pseudomorphic growth of 40-period strained InAs/Ga0.6In0.4Sb SL with good structural and surface quality. Because the InAs/Ga1−xInxSb...
QW can be contingently considered as a "single-period SL," it should not require a large critical layer thickness for the pseudomorphic growth. Therefore, the pseudomorphic growth of the InAs/Ga1−xInxSb QW structures may be possible even with $x > 0.4$. An inverted bandgap of more than 60 meV is expected in these QWs.

Another way to increase a bandgap in QSHI state is based on utilizing InAsSb alloys. Having the lowest bandgap among all III–V semiconductors, InAsSb alloys attract attention both for fundamental research as a potential alternative to HgCdTe for long-wavelength IR optoelectronics. The recent development of the MBE technique made it possible to grow high-quality InAsSb bulk films (37) and type II strained InAs1−ySb/yInAs1−ySb, y SL (38). Preliminary estimations of band structure calculations performed for a modeled three-layer InAsSb/GaInSb and InAs1−xSbx/InAs1−xSbx QWs also show theoretical possibility for the large gap in the QSHI state of more than 60 meV but at lower strain in the QW layers.

Finally, we stress an important advantage of QSHI based on the three-layer InAs/GaInSb QWs as compared with the one based on HgTe/CdTe QWs. Because temperature does not affect the ordering of the $\Gamma_8$ and $\Gamma_9$ bands in bulk InSb, InAs, GaSb, and AlSb semiconductors, one can expect a substantially weaker dependence of the bandgap on temperature in the multilayer broken-gap QWs than in HgTe/CdHgTe QWs.

Very recently, the quantum spin Hall effect in 1T'-WTe2 MLs at temperatures up to 100 K has been reported (39). We note that this is the highest temperature at which the main hallmark of the QSHI state has ever been observed. This high temperature is consistent with a relatively large inverted bandgap (~45 meV) in the 1T'-WTe2 extracted from recent spectroscopy (40, 41). Moreover, it is suggested that the 100-K temperature may not be an intrinsic limit of 1T'-WTe2 MLs.

In view of these novel experimental results and mature fabrication technology, the multilayer broken-gap QWs based on III–V semiconductors become extremely attractive for high-temperature topological electronics.

MATERIALS AND METHODS

Band structure calculations were performed by using the eight-band Kane model (30), which directly takes into account the interactions between $\Gamma_8$, $\Gamma_9$, and $\Gamma_7$ bands in bulk materials. This model well describes the electronic states in a wide range of narrow-gap semiconductor QWs, particularly in the broken-gap InAs/GaSb QWs (12). In the eight-band Kane Hamiltonian, we also took into account the terms, describing the strain effect arising because of the mismatch of lattice constants in the buffer, QW layers, and AlSb barriers. To calculate LLs, we used the so-called axial approximation. Within this approximation, one keeps in-plane rotation symmetry by omitting the warping terms in the Hamiltonian. The calculations had been performed by expanding the eight-component envelope wave functions in the basis set of plane waves and by numerical solution of the eigenvalue problem. Details of calculations, notations of the LLs, and the form of the Hamiltonian can be found in the study of Krishtopenko et al. (30). Parameters for the bulk materials and valence band offsets used in the eight-band Kane model are taken from Vurgaftman et al. (27). Here, we considered the structures grown along (001) crystallographic orientation.

To derive effective six-band 2D Hamiltonians $H_{InAs}(k_x, k_y)$ and $H_{GaSb}(k_x, k_y)$, valid in the vicinity of the $\Gamma$ point from the eight-band Kane model, we followed the procedure proposed by Bernevig et al. (2) and described in detail in the supplementary materials of Krishtopenko et al. (25). First, we split the Kane Hamiltonian $H_{Kane}$ into two parts, $H_{Kane} = H^{(0)}(k_x) + H^{(1)}(k_y, k_x, k_y)$, where $H^{(0)}(k_x)$ is the Kane Hamiltonian at $k_y = k_z = 0$. In our case, the z axis is oriented along (001) direction, whereas the $x$ and $y$ axes correspond to the (100) and (010) directions, respectively. Then, we numerically diagonalized $H^{(0)}(k_x)$ to obtain the energies and envelope functions and to classify electronic levels as electron-like $E_n$, heavy hole-like $H_n$, light hole-like $L_nH_n$, or spin-off–like $SO_n$ levels ($n = 1, 2, \ldots$), as it is performed by König et al. (11).

Then, we grouped the eigenstates of $H^{(0)}(k_x)$ into $A$ and $B$ subsets. The $A$ subset includes the basis sets of $[|E_1, \pm z\rangle, |J_1, \pm z\rangle, |E_2, \pm z\rangle]$ levels for the InAs-designed QWs and $|E_1, \pm z\rangle, |J_1, \pm z\rangle, |J_2, \pm z\rangle$ for the GaSb-designed structures. In the $B$ subset, we considered eight above-lying, electron-like states and eight subbands for light hole– and heavy hole–like states. All the other subbands were neglected because they were well separated in energy. The states in both classes were not coupled because they are eigenstates of $H^{(0)}(k_x)$. The presence of $H^{(1)}(k_x, k_y, k_z)$ introduces the mixing between the states from two subsets. To derive effective 2D Hamiltonians $H_{InAs}(k_x, k_y)$ and $H_{GaSb}(k_x, k_y)$, we treated $H^{(1)}(k_x, k_y, k_z)$ as a small perturbation and applied the second-order Löwdin perturbation method to eliminate the coupling between the states from different subsets.

$$H_{eff}(k_x, k_y)_{m,m'} = H^{(1)} + \frac{1}{2} \left( H^{(1)} \right)_{m,m'} \left( \frac{1}{E_m - E_l} + \frac{1}{E_{m'} - E_l} \right)$$

Here, the indices $m, m'$ correspond to states in set $A$, the indices $l$ correspond to the states in set $B$, and $H^{(1)}_{m,m'}$ is the matrix element of $H^{(1)}(k_x, k_y, k_z)$, calculated by using the envelope function of $H^{(0)}(k_x)$. Parameters for both effective Hamiltonians for different thicknesses of InAs $d_1$ and GaSb $d_2$ layers are provided in the Supplementary Materials.

To calculate dispersion of the edge states on a single edge, we dealt with a system on a half-plane of $y < 0$ and replaced $k_y \rightarrow -i\partial_y$ in $H_{InAs}(k_x, k_y)$ and $H_{GaSb}(k_x, k_y)$. If $M < 0$, both types of three-layer InAs/GaSb QWs support the edge states, which exponentially decay at $y \rightarrow -\infty$. The 2D effective Hamiltonians $H_{InAs}(k_x, -i\partial_y)$ and $H_{GaSb}(k_x, -i\partial_y)$ are block-diagonal, and the eigenvalue problem of the upper and lower blocks was solved separately. To find the energy spectrum of the edge states for each block, we numerically solved the Schrödinger equation with the boundary condition for the wave function to vanish at $y = 0$. Taking into account the translation invariance along the $x$ axis, the wave function of the edge states has the form

$$\Psi_{edge}(x, y) = e^{ik_xx} \sum_{n=1}^{3} \alpha_n e^{i\lambda_n y} C_n$$

where $k_x$ is the wave vector along the edge, $L_x$ is the sample size along the $x$ axis, $\alpha_n$ is the coefficient determined by the boundary conditions, $\lambda_n$ is the complex-valued reciprocal lengths, and $C_n$ are the position-independent normalized three-component columns.

For a given wave vector $k_x$, relation $\lambda_n(E_{edge})$ and columns $C_n$ are found from the matrix equation

$$H_{1,2}(k_x, -i\lambda) C_{n,edge} = E_{edge} C_{n,edge}$$

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Note that $\lambda_{\text{in}}$, in general, can be complex, corresponding to a mixture of the edge and bulk states. The energy spectrum of the edge states is found from the condition of wave function decay at $y \to -\infty$ [by implying real part of complex number $\Re(e)$ (a real part of complex number) $\lambda_{\text{in}} > 0$] and from the boundary condition at $y = 0$:

$$\sum_{n=-\infty}^{\infty} a_n C_n^{(\text{trap})} = 0$$

The energy spectrum of the edge states for the lower blocks in $H_{\text{InAs}}(k_{\text{in}})$ and $H_{\text{GaSb}}(k_{\text{in}})$ is found in a similar manner.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/4/eaap5729/DC1

**Supplementary Text**

fig. S1. Comparison between calculations within the eight-band Kane model and by using the effective Hamiltonians for three-layer InAs/GaSb QWs.

fig. S2. Bandgap in QSHI state in asymmetrical three-layer InAs/GaInSb QWs.

table S1. Parameters involved in the effective Hamiltonian $H_{\text{InAs}}(k_{\text{in}})$ for three-layer InAs/GaSb QWs with InAs geometry grown on GaSb buffer.

table S2. Parameters of the effective 2D Hamiltonian $H_{\text{GaSb}}(k_{\text{in}})$ for three-layer InAs/GaSb QWs with GaSb geometry grown on GaSb buffer.

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