Disorder effects of vacancies on the electronic transport properties of realistic topological insulators nanoribbons: the case of bismuthene

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The robustness of topological materials against disorder and defects is presumed but has not been demonstrated explicitly in realistic systems. In this work, we use state-of-the-art density functional theory and recursive nonequilibrium Green’s functions methods to study the effect of disorder in the electronic transport of long nanoribbons, up to 157 nm, as a function of vacancy concentration. In narrow nanoribbons, even for small vacancy concentrations, defect-like localized states give rise to hybridization between the edge states erasing topological protection and enabling backscattering events. We show that the topological protection is more robust for wide nanoribbons, but surprisingly it breaks down at moderate structural disorder. Our study helps to establish some bounds on defective bismuthene nanoribbons as promising candidates for spintronic applications.

I. INTRODUCTION

Topological materials have been intensively studied in recent years [1–5] unveiling interesting new physics and opening new applications possibilities in spin-based electronic devices [6]. Of particular interest are large band gap topological insulators (TIs), that are good candidates for the realization of the quantum spin Hall (QSH) effect at room temperature [7, 8]. In two-dimensional (2D) QSH insulators, the edges of the sample carry metallic states that are protected by time-reversal symmetry (TRS) [9, 10] and decay exponentially into the bulk [11–13]. Moreover, theory predicts that in nanoribbons these edge states carry dissipationless helical spin currents. The penetration depth, that quantifies the edge states exponential decay rate, is (roughly) inversely proportional to the band gap [12, 14] and plays a key role in the nanoribbon transport properties. As it happens with 3D topological insulators [15], to fully display the features of a TI, the nanoribbon width must be much wider than the penetration depth $\xi$ of the edge states, otherwise they can easily hybridize.

Among several candidates for topological materials, bismuthene, also known as buckled or bilayer bismuthene, is of special interest due to its large electronic band gap of 0.5 eV, along with its structural stability and large spin-orbit coupling (SOC) [16–18]. The existence of charge puddles and other types of defects that could be detrimental for the formation of a topological phase do not play an important role in bismuthene, as experimentally demonstrated [7]. Indeed, since its experimental realization, it was proposed that even amorphous structures of bismuthene occurring before the annealing process [19], as well as strongly disordered systems [20], support topological states. The robustness of bismuthene non-trivial topology [19] makes it ideal for material design by tuning the lattice constant and effective spin-orbit coupling (SOC) by epitaxial constraint, and lighter elements substitutional alloying with Sb or As, without causing a topological transition [16, 21]. The presence of non-magnetic defects leaves the band topology unchanged in these materials [18–20].

However, a recent study using a schematic tight-binding model [22] has shown that vacancy induced localized states can give rise to local magnetic moments and destroy the topological protection. Contrariwise, using a different tight-binding toy model, it was numerically shown that small concentrations of vacancy defects do not eliminate the topological edge states, but can cause edge state hybridization in certain energies intervals within the topological gap [23]. To settle such kind of controversy and better understand the interplay between edge states and defects a systematic study using ab initio methods is in order.

Vacancies on bismuthene show small formation energies where $sp^2$ bismuth dangling bonds can be distributed around the vacancy leading to resonances in the band gap, modifying the electronic properties of the host material. Theoretical calculations on bismuthene show no evidence of magnetic moments induced from vacancies [17], preserving TRS and retaining its non-trivial topological band structure. Recent ab initio calculations demonstrate that transport along the edges is insensitive to vacancies created in ultra-narrow TI zigzag nanoribbons [24]. Even though these vacancies allow the development of magnetic moments, the perfect conductance for energies within the bulk gap is recovered provided the vacancies are passivated by hydrogen. Our study, in turn, addresses more realistic system sizes, namely, both much wider and much longer, unveiling a different kind

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of disordered-induced transport mechanism.

In this work, we investigate the electronic transport properties of buckled bismuthene nanoribbons of realistic sizes using the full Hamiltonian in the orbital representation obtained from density functional theory (DFT) [25–27] calculations combined with recursive nonequilibrium Green's functions (NEGFs) [28–31]. First, we explore the backscattering mechanism due to inter-edge hopping mediated by vacancy localized states in different nanoribbon widths and discuss the detrimental effects in transport properties caused by single vacancies in narrow nanoribbons. We proceed to investigate the robustness of the edge states for different ribbon widths, reaching extended lengths (157 nm), as a function of vacancy concentration, demonstrating distinct behaviors in the transport properties for bulk and edge states. We show that narrow ribbons, whose widths \( w \) are comparable with the edge states penetration depth \( \xi \), present the onset of Anderson localization effects already at low vacancy concentrations. In distinction, for wide nanoribbons, where \( w \gg \xi \), the edge states maintain a quantized conductance in the topological gap at low disorder concentrations. Surprisingly, we find that topological protection is destroyed already at modest vacancy concentrations.

II. COMPUTATIONAL METHODS

Our calculations combine the flexibility of a plane-wave basis set to obtain the optimized structures with a localized basis for electronic transport. In both cases, we use the Perdew-Burke-Ernzerhof (PBE) [32, 33] exchange-correlation functional. We perform the geometry optimizations with a plane-wave basis as implemented in the VASP package [34, 35]. In these calculations, we employ 400 eV for the plane-wave expansion cutoff. Vacancies are modeled by removing an atom and performing the geometry optimization with force criterion of \( 5 \times 10^{-3} \text{eVÅ}^{-1} \).

The transport calculations use full \textit{ab initio} DFT Hamiltonian matrices obtained directly from the SIESTA code [37], employing atom-centered single-\( \zeta \) plus polarization (SZP) basis sets. We use the energy cut-off for real-space mesh of 350 Ry, sampling the reciprocal space with 10 \textit{k}-points along the periodic direction of the presented nanoribbons, which edges were hydrogen-passivated. We add 20 Å of vacuum in both non-periodic directions to avoid spurious interactions between periodic images. The self-consistent SOC is introduced via an on-site approximation [38] using fully-relativistic norm-conserving pseudopotentials [39]. The system Hamiltonian and overlap matrices are obtained after performing a full self-consistent cycle.

The electronic transport calculations are implemented following the standard NEGF approach [28–30]. We consider a two-probe terminal setting, as illustrated in Fig. 1(a). The left (L) and right (R) electrodes are modeled by semi-infinite pristine zigzag bismuthene leads.

Our approach employs a decimation technique [40–45] that allows us to address large system sizes. The scattering region is partitioned into building blocks connected by first neighbour interactions. Each building block is computed through DFT. The procedure takes into account all degrees of freedom of the system comprised of all the building blocks. The partition scheme is depicted in Fig. 1(b). Accordingly, the system Hamiltonian is expressed in the localized basis by the block matrix

\[
H = \begin{pmatrix}
H_L & H_C & 0 \\
H_C^\dagger & H_S & H_C \\
0 & H_C^\dagger & H_R
\end{pmatrix}
\]  

(1)

where \( H_L \) and \( H_R \) are the Hamiltonian matrices describing the left and right electrodes, while \( H_C \) is the coupling between the leads and the central region, and \( H_S \) is the tridiagonal block matrix

\[
H_S = \begin{pmatrix}
H_1 & H_C & 0 & \ldots & 0 \\
H_C^\dagger & H_2 & \ldots & \ldots & \vdots \\
0 & \vdots & \ddots & \vdots & 0 \\
\vdots & \ldots & \ldots & H_{N-1} & H_C \\
0 & \ldots & 0 & H_C^\dagger & H_N
\end{pmatrix}
\]  

(2)

representing the scattering region (S).
The scattering region spin resolved retarded Green’s function \[30, 46\] reads

\[
G^r_S(E) = \left( E^+ S_S - H_S - \Sigma^r_L - \Sigma^r_R \right)^{-1} \tag{3}
\]

where \( E^+ = \lim_{\gamma \to 0^+} E + i \delta \), \( S_S \) is the overlap matrix, and \( \Sigma^r_{R/L}(E) = (E^+ S_C - H_C)G^r_{0,R/L}(E^+ S_C^\dagger - H_C^\dagger) \) are the embedding self-energies that account for the system decay width due to the coupling with the leads. Here \( G^r_{0,R/L} \) is the retarded surface Green’s function of the \( R/L \) electrode \[40, 47\]. The block structure of \( H_S \) allows for a very efficient computation of \( G^r_S(E) \) using the recursive Green’s function method (see, for instance, ref. \[44\], for a review).

In the linear response, at small bias, the zero-temperature conductance is given by the Landauer formula \( G(E_F) = (e^2/h) T(E_F) \), where the transmission \( T \) reads \[28–30\]

\[
T(E) = \text{Tr} [\Gamma_L(E) G^r_S(E) \Gamma_R(E) G^r_S(E)] \tag{4}
\]

where \( G^r_S = [G^r_S]^\dagger \) and the decay width matrices \( \Gamma_{L/R} \) are given by \( \Gamma_{L/R} = i(\Sigma_{L/R} - \Sigma^a_{L/R}) \).

### III. RESULTS AND DISCUSSION

In this section we analyze the effect of vacancies on the transport properties of bismuthene zigzag nanoribbons. We begin by discussing the topological properties of pristine nanoribbons of different representative widths. Next, we investigate the effect of a single-vacancy on the electronic and transport properties of these systems. Finally, we study the conductance of these systems for different vacancy concentrations and nanoribbon widths.

Let us first investigate the effect of the ribbon width on pristine systems. In Fig. 2 we show the electronic band structure and the conductance for bismuthene nanoribbons with 3 different representative widths. To help the discussion, the bulk-topological gap \( \Delta_{TG} \), corresponding to \(-0.1 \text{ eV} < E < 0.4 \text{ eV}\), is indicated in grey. Figure 2(a) shows the results for a narrow nanoribbon, \( w_{20} \) of width 20 Å, comparable to those of ref. [24]. The electronic states bridging the bulk-topological gap \( \Delta_{TG} \) are split and do not display the Kramer’s degeneracy. Hence, there is no manifestation of the topological edge states. Figure 2(b) shows an intermediate width nanoribbon, namely, \( w_{65} \) of width 65 Å. Here, there is still a small gap at the \( \Gamma \)-point, though the edge states are degenerate as expected for a topological insulator. In addition, the top of the valence and bottom of the conduction trivial state bands correspond the corresponding bulk energies, indicating that finite width quantization effects are much smaller than in the previous case. Finally, Fig. 2(c) shows a wide nanoribbon, \( w_{110} \) of width 110 Å, with no gap and degenerate edge states with helical texture, as expected for a TI. Here, the bulk-topological gap corresponds very closely to the energy interval where one finds only edge states. In all cases, the conductance \( G(E) \) is quantized and the conductance steps are observed as expected for pristine systems \[30\].

The lack of topological protection in narrow ribbons is a result of strong overlap between the states at the opposite system edges. This can be understood in terms of the spatial localization of edge states or, more precisely, their so-called penetration length \( \xi \). The latter can be roughly estimated from the mass term in the \( k \cdot p \) Dirac Hamiltonian describing the inverted bulk band gap as \[14, 48–50\]

\[
\xi \approx \hbar v_F / \Delta_{TG}, \tag{5}
\]

where \( v_F \) is the Fermi velocity corresponding to the topological bands, namely, \( \hbar v_F = d\xi / dk \). By estimating the Fermi velocities from Figs. 2(b) and (c) we find: For the \( w_{65} \) nanoribbon \( v_F = 4.58 \times 10^5 \text{ m s}^{-1} \), that renders \( \xi = 0.6 \text{ nm} \), while for \( w_{110} \) the Fermi velocity is \( 5.79 \times 10^5 \text{ m s}^{-1} \) and \( \xi = 0.8 \text{ nm} \). These estimates of \( \xi \)
for buckled bismuthene are slightly larger than the values reported in experiments \cite{7, 51} that obtain $\xi \approx 0.4 \text{ nm}$ in SiC(0001) supported flat bismuthene. We stress that these are different material systems. The discrepancy can be explained by recalling that $\Delta_{\text{TG}}$ of the buckled bismuthene is smaller than the band gap of the flat one.

A more quantitative estimate of $\xi$ is taken from the local density of states (LDOS) averaged over the $x, z$ directions as a function nanoribbon transversal axis $y$, see Fig. 3. Fitting an exponential function, we estimate the penetration length as $\xi = 0.9 \text{ nm}$ for the $w_{110}$ ribbon, in good agreement with $\xi$ obtained using Eq. (5). Using $\xi$, we can also estimate a threshold length for the LDOS decay for which the interedge states hybridization becomes negligible. For instance, at distances $\Delta y \simeq 2.8 \text{ nm}$ from the edges, the LDOS decays by $\sim 95\%$. Hence, we expect that narrow nanoribbons with $w \lesssim 2\Delta y$ lack topological protection due to the strong overlap of states localized at opposite system edges. The $w_{65}$ nanoribbons are at the crossover between non-protected and topologically protected phases.

Next, we investigate ribbons containing a single-vacancy. For a slab geometry, we calculate the formation energy $E_{V}$ of these single vacancies systems using the following expression \cite{52}

$$E_{V} = E_{\text{tot}} - (E_{\text{pristine}} + \mu_{V} N_{V})$$ \hspace{1cm} (6)

where $E_{\text{tot}}$ is the total energy of the single-vacancy system given by a fully relaxed DFT calculation, $E_{\text{pristine}}$ is the energy for the pristine ribbon, $N_{V}$ is the number of vacancies (in our case $N_{V} = 1$), and $\mu_{V}$ is the chemical potential to remove a bismuth atom. Here, we take $\mu_{V}$ as the energy per atom of the pristine bismuthene monolayer. For purposes of comparison, we simulate a $5 \times 5$ supercell containing a vacancy. By doing so, we avoid that theses vacancies interact with their periodic images obtaining a formation energy of 1.04 eV, which is in agreement with previous reports \cite{17}.

Figure 4 shows the conductance of the $w_{20}$ and $w_{110}$ ribbons in the presence of a single vacancy placed close to one of the system edges. For this calculation, the scattering region corresponds to a single building block $N = 1$ containing the vacancy. For the $w_{20}$ ribbon the quantized conductance is destroyed, showing that the edge states are not robust against disorder. In turn, the $w_{110}$ ribbon shows no deviation from perfect conductance within the topological gap.

Several studies on a variety of 2D materials indicate that single-vacancies give rise to localized states \cite{22, 53}. It has been further shown that such disorder-induced localized states cause the formation of local magnetic moments \cite{53–57} that, if close to the system edges, can be detrimental for the topological protection, differently from dual topological insulators \cite{58}. Let us study the relevance of these findings to bismuthene.

Figure 5 shows the local density of states (LDOS) for the $w_{20}$, $w_{65}$ and $w_{110}$ nanoribbons calculated for the energy window $0.2 \text{ eV} < E < 0.3 \text{ eV}$. The strong enhancement of the LDOS centered around the vacancy corresponds to exponentially decaying orbitals paired after the atom relaxation. Figure 5(a) shows that the vacancy states increase the overlap between inter-edge states, whereas for the $w_{65}$ nanoribbon the overlap is small even in the presence of a vacancy. For the $w_{110}$ ribbon shown in Fig. 5(c) the overlap between the edge
states and the vacancy localized states is negligible.

\( G/G \) for single vacancy

Our \textit{ab initio} fully relativistic calculations show no magnetic moments generated by the defects. Nonetheless, when SOC is turned off, spin polarized calculations show a magnetic moment of 0.8 \( \mu_B \) around the vacancy. This result rules out vacancy-induced local moments [22] as a method to hinder the topological protection.

We now study the conductance as a function of vacancy concentration. For that purpose, we put together building blocks (as described in Sec. II) of length \( N \) ranging from 0.025 \( \text{Å}^{-3} \text{eV}^{-1} \), corresponding to an energy range 0.2 eV \( \leq E \leq 0.3 \) eV.

\( G/G \) for\( \xi \).

FIG. 5. Top (x-y axis) and lateral (y-z axis) projections of the local density of states (LDOS) for single vacancy in (a) \( w_{20} \), (b) \( w_{65} \) and (c) \( w_{110} \) nanoribbons. The isosurface value for the LDOS is of 0.025 \( \text{Å}^{-3} \text{eV}^{-1} \), corresponding to an energy range 0.2 eV \( \leq E \leq 0.3 \) eV.

Let us now address the case of wide nanoribbons, namely, \( w \gg \xi \). Figures 6(i) to 6(l) correspond to \( w_{110} \) nanoribbons with \( n_V \) ranging from 0.13 \( \% \) to 0.48 \( \% \). As in the previous cases, \( G \) decreases with increasing \( n_V \) for energies outside \( \Delta_{TG} \). In distinction, the edge states are topologically protected by time-reversal symmetry and \( G = G_0 \) over most of the bulk band gap energies. Although topological protection is more robust than in the previous cases, when \( n_V \geq 0.30 \% \), \( G/G_0 \) is strongly suppressed for certain energy intervals within \( \Delta_{TG} \), see Figs. 6(k) and (l), indicating the presence of vacancy-induced inter-edge backscattering processes.

These results suggest that the number of vacancy-induced states necessary for an effective inter-edge hybridization the system edge states increases with the nanoribbon width \( w \).

IV. CONCLUSIONS AND OUTLOOK

In this paper we have studied the robustness of the conductance quantization against vacancy disorder in large scale nanoscopic buckled bismuthene nanoribbons at the QSH phase.

We have found that vacancies give rise to mid-gap localized states and are non-magnetic, ruling out local
magnetic moments [22] as a mechanism to destroy topological protection. We have shown that the vacancy-induced mid-gap states can give rise to inter-edge scattering processes. These depend on the edge state penetration depth, vacancy concentration, and nanoribbon width. The interplay of these quantities has been qualitatively discussed in QSH tight-binding models [23] and within DFT for ultra-narrow systems [24]. Here, we have established the presence of vacancy-induced inter-edge backscattering processes in bismuthene nanoribbons of realistic sizes using ab initio techniques.

Our calculations show different transport behavior for bulk and edge states, the first demonstrating localization effects and the latter showing robust topological response for low vacancy concentrations. At moderate $n_V$ values, topological protection is destroyed even for wide ribbons $w \gg \xi$.

Our findings are also applicable to other materials in the QSH regime. Since the penetration depth is material dependent, we conclude that it is possible to engineer different samples with QSH electronic transport behavior in the presence of disorder. Our findings suggest an interesting application for a spintronic device, the level of doping or the width of the device modules the edge states degeneracy, therefore providing the ON/OFF states for a transistor switch. This might be obtained by changing the chemical potentials on different leads or by applying a gate voltage. In particular, for the topological $w_{65}$ and $w_{110}$ nanoribbons, characterized by a pronounced drop in the conductance becoming broader for energies around the valence band as the level of vacancy concentration increases, allowing the possibility of reaching high ON/OFF ratios. The mechanism to modify the conductance in these nanoribbons does not rely on changing the topology of its band structure as it would happen by applying an electric or magnetic field.

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