Subsurface impurities and vacancies in a three-dimensional topological insulator

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Using a three-dimensional microscopic lattice model of a strong topological insulator (TI) we study potential impurities and vacancies in surface and subsurface positions. For all impurity locations we find impurity-induced resonance states with energy proportional to the inverse of the impurity strength, although the impurity strength needed for a low-energy resonance state increases with the depth of the impurity. For strong impurities and vacancies as deep as 15 layers into the material, resonance peaks will appear at and around the Dirac point in the surface energy spectrum, splitting the original Dirac point into two nodes located off-center. Furthermore, we study vacancy clusters buried deep inside the bulk and find zero-energy resonance states for both single and multiple-site vacancies. Only fully symmetric multiple-site vacancy clusters show resonance states expelled from the bulk gap.

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I. INTRODUCTION

Topological insulators (TIs) are a new class of quantum matter, where strong spin-orbit coupling results in a bulk energy gap but gapless metallic surface states\(^1\). In strong TIs a topological invariant, associated with the bulk band structure, guarantees the existence of a single (or odd number) surface state with characteristic linear Dirac energy dispersion, where the electron spin is locked to the momentum\(^2\). This surface state topology also protects it against any time-reversal invariant perturbation. Taken together, these properties make TIs intriguig candidates for both spintronic devices and topological quantum computation\(^3\). The topological stability of the surface state is intimately connected with the absence of backscattering for nonmagnetic impurities, since a spin-flip is required for 180° backscattering. The lack of backscattering was established theoretically early on within a two-dimensional (2D) continuum model for the surface state\(^4\) and later also confirmed in experiments.\(^5\) The same 2D continuum model finds that, while local impurity-induced resonance states exists for a potential impurity, their weight diminish as the energy approaches the Dirac point for unitary scatterers.\(^6\) We recently showed that using a microscopic 3D lattice model for a strong TI, which naturally includes a finite bulk gap, strong impurities on the surface give rise to large resonance peaks in the local density of states (LDOS) at and around the Dirac point.\(^7\) Consequently, close to the impurity the Dirac point is destroyed in the local density of states (LDOS) and split into two nodes that move off-center. This lack of local topological protection close to strong potential scatterers is due to 3D scattering processes, which engage the bulk states.\(^8\) With a finite bulk gap, spin-flip processes, which are forbidden in a pure 2D theory, become allowed through virtual excitations to the bulk. Recent scanning tunneling spectroscopy (STS) results\(^9\) on \(\text{Bi}_2\text{Se}_3\) have demonstrated the existence of strong resonance peaks at and around the Dirac point for unitary impurities, thus confirming the importance of bulk-assisted processes for the properties of the TI surface state. Other experimental data has also shown how both localized bound states at defects\(^10\) and steps\(^11\) as well as line width broadening\(^12\) do not agree with results from a purely 2D surface continuum model.

In this article we expand our previous work on the effect of impurities in a microscopic 3D lattice model\(^12\) to also include potential impurities and vacancies located in subsurface layers. With the profound impact of strong impurities on the surface, it is very natural to ask what happens to the Dirac surface state spectrum in the presence of subsurface impurities. Since the surface state extends multiple layers into the material\(^13\) impurities within these layers might also affect the LDOS of the surface, and can thus be very important for both the functionality of the TI surface and the analysis of STS measurements. Closely connected to the properties of subsurface impurities is also that of the characteristics of bulk impurities in TIs. Both potential impurities\(^14\) and finite sized holes\(^15\) in the bulk of a 3D TI have previously been treated within a continuum theory focusing on in-gap bound states. A finite sized hole constitutes an interior surface and will thus necessarily also host a surface state in a TI. Within a continuum theory, this surface state is transformed into bound states with finitely spaced energy levels, which are eventually expelled to the bulk bands with shrinking hole radius. Here we will study how the continuum results can be related to microscopically created bulk vacancy clusters. If these clusters give rise to low-lying resonance states, a finite concentration will have a large effect on the surface conductivity as they enable edge-edge scattering.

We find here that: i) Subsurface impurities and vacancies as far as around 15 layers into the material create a non-dispersive resonance peak in the surface LDOS. ii) For all impurity layer positions, including the surface and the bulk, the resonance energy \(E_{\text{res}} \propto 1/U\). However, for the resonance state to enter the low-energy region, the impurity strength needs to be stronger the deeper down the impurity is buried.
In the limit of a very deep impurity inside the TI, only vacancies, which are potential impurities of infinite strength, will give a resonance peak at the Dirac point on the surface. These results show that even deep subsurface impurities will affect the low-energy region of the surface state spectrum, and will thus be important when interpreting experimental surface probe results. Single-site bulk vacancies produce a (bulk) resonance peak at $E = 0$ for mirror-symmetric conduction and valence bands. Fully symmetric 5 and 17-site vacancy clusters have no in-gap resonance peaks, as also predicted by continuum results. However, any small deviation from full symmetry produces low-lying resonance peaks. We therefore conclude that any realistic microscopically created hole in a 3D TI will have a resonance peak around $E = 0$, thus mimicking the results of a single vacancy instead of that of a finite size hole.

The rest of the article is organized as follows. In Sec. II, we introduce a general microscopic lattice model for studying defects and vacancies in a strong 3D TI. In Sec. III A, we discuss the surface LDOS and impurity-induced resonance peaks for surface and subsurface impurities and vacancies. In particular, we focus on the dependence of the resonance energy on layer position and impurity strength. Next in Sec. III B, we discuss multiple-site vacancy clusters buried deep inside the bulk of the TI. We conclude in Sec. IV by summarizing our results, listing some of their experimental consequences, and providing a comparison to recent experiments.

II. MODEL

We study a microscopic 3D lattice model similar to that of Ref. [12], but for self-containment we reiterate the details here. We create a strong TI by using a four band $s$-orbital tight-binding scheme on the diamond lattice with spin-orbit coupling.

$$
H_0 = t \sum_{(i,j)} c_i^\dagger c_j + \mu \sum_i c_i^\dagger c_i + \frac{4\lambda}{a^2} \sum_{(i,j)} c_i^\dagger s \cdot (d_{ij}^1 \times d_{ij}^2)c_j.
$$

Here $c_i$ is the annihilation operator on site $i$ where we, for simplicity, have suppressed the spin-index. Furthermore, $t$ is the nearest neighbor hopping, $\mu = 0$ the chemical potential, $\lambda = 0.3t$ the next-nearest neighbor spin-orbit coupling, $\sqrt{2a}$ the cubic cell size, $s$ the Pauli spin matrices, and $d_{ij}^1, d_{ij}^2$ the two bond vectors connecting next-nearest neighbor sites $i$ and $j$. By further distorting the hopping amplitude to $1.25t$ along one of the nearest neighbor directions not parallel to the (111) direction, this system becomes a strong TI, with a single surface Dirac cone. In order to access a surface we create a slab of Eq. (1) along the (111) direction. We are mainly studying slabs with ABBC...AABBCC stacking terminations, hereafter labeled AB termination, but will also compare these results with AABBCC...AABBCC terminated slabs, labeled AA termination, in order to generalize our results. We choose an energy scale such that the slope of the surface Dirac cone $h\nu_F \approx 1$ for an AB slab, which is achieved by setting $t = 2$ throughout this work. We find that for slabs with $r \geq 5$ lateral unit cells, where each lateral cell contains six atomic layers, there is only a minimal amount of cross-talk between the two slab surfaces, resulting in a negligible surface energy gap. We label the different layers in the slab starting with layer 1 for the surface layer. Around layer 15, the remnant DOS of the surface state is becoming negligible and also located close to the bulk gap and we are thus approaching bulk conditions at this depth.

In order to study the effect of local potential impurities or vacancies we create a rectangular-shaped surface supercell with $n$ sites along each direction. This gives a supercell surface area of $\sqrt{3n^2a^2}/2$ where we use $a = 1$ (the nearest neighbor distance on the surface) as the unit of length. We add impurities to our model by adding the term

$$
H_{\text{imp}} = U \sum_i c_i^\dagger c_i.
$$

to the Hamiltonian in Eq. (1). Here $U \geq 0$ is the impurity strength and the summation is over all impurity sites in the supercell. We note that by adding $H_{\text{imp}}$ we break particle-hole symmetry and thus our model, even with $\mu = 0$, corresponds to a rather general situation. We solve $H = H_0 + H_{\text{imp}}$ in the supercell using exact diagonalization. We find that a $50 \times 50$ $k$-point grid gives sufficient resolution, while at the same time using a Gaussian broadening of $\sigma = 0.005$ when calculating the LDOS.

III. RESULTS

A. Subsurface impurities

We start by studying single-site, isolated, potential impurities with varying impurity strength $U$ including the case of a single vacancy where $U \rightarrow \infty$, which represents the unitary scattering limit. Earlier results treated only impurities on the surface of the TI, but here we extend the study to the situation where the impurity is located in subsurface layers, all the way down to the bulk. Figure I shows layer resolved LDOS on in-plane nearest neighbor sites for both a vacancy (left column) and a $U = 40$ impurity (right column), for different impurity layer positions. Starting with a surface layer vacancy (topmost left), there is a wide, double-peak resonance roughly centered at the Dirac point at $E = 0$. As carefully analyzed in previous work [12] a surface vacancy creates a resonance peak firmly situated on top of the original Dirac point, which splits into two Dirac points situated on either side of the resonance peak. These two Dirac points are the termination points of the valence and conduction band Dirac surface state spectra, respectively. The local destruction of the topologically protected low-energy Dirac surface state spectrum, and its Dirac point, is due to surface-bulk interaction always present in TIs with a finite bulk band gap. In the presence of a strong impurity, the finite bulk band gap enables virtual spin-flip bulk scattering events, which then allows $180^\circ$...
FIG. 1: (Color online) Layer resolved LDOS averaged over in-plane nearest neighbor sites to a vacancy (a-f) and an $U = 40$ impurity (g-l) positioned in layer 1, 2, 3, 4, 7, 20 (counted from the top) plotted for each layer across a $r = 7$ lateral unit cell wide slab with AB termination with a supercell size of $n = 10$. Zero (white), 0.1 (black) states per energy and area unit. Red/Grey dashed vertical lines mark impurity layer, whereas horizontal dotted lines mark $E = 0$.

The double-peak structure is also less visible as $n$ increases, but the center of the peak remains fixed. In fact, the resonance peak is non-dispersive throughout the whole slab for all impurity concentrations.

Since the surface state penetrate relatively deep into the material, by reciprocity argument impurities positioned in subsurface layers might have a profound effect on the surface LDOS. Figures 1(b-f) show single vacancies positioned in layer 2, 3, 4, 7, and 20 respectively. There is some oscillation in the energy of the peak as function of vacancy layer position, but for all subsurface layer positions $< 15$, there is still a finite sized resonance peak located at or around $E = 0$ in the surface LDOS. Thus, the original single Dirac point on the surface is destroyed and split into two nodes that move off-center even for vacancies positioned relatively deep into the TI. There is again no energy dispersion of the resonance peak throughout the material. We also find a single-double peak oscillation where double peaks only appear for vacancies in every other layer. However, this layer position difference diminishes with increasing impurity-impurity distance. When approaching the bulk layers, the resonance peak centers firmly at $E = 0$, and its impact on the surface state diminishes when the distance to the surface increases. In Fig. 1(f), the vacancy is positioned deep within the bulk, and there is a narrow, but tall, impurity resonance peak at $E = 0$, but which do not penetrate to the surface. This result can be understood rather straightforwardly by applying the $T$-matrix formalism to an idealized, but normal, insulator. In the presence of a scattering potential $\hat{V}$, the Green’s function $\hat{G}$ is determined by

$$\hat{G} = \hat{G}^0 + \hat{G}^0 \hat{T} \hat{G}^0,$$  \hspace{1cm} (3)

where $\hat{G}^0$ is the bare Green’s function and the $T$-matrix is given by

$$\hat{T} = (1 - \hat{V} \hat{G}^0)^{-1} \hat{V}. \hspace{1cm} (4)$$

Since the poles of the Green’s function give the energy spectrum for single-particle excitations, we can find the energy $E_{\text{res}}$ of any impurity-induced resonance state by searching for poles in the $T$-matrix. For an atomically sharp impurity, described by the $\delta$-function potential $\langle x | \hat{V} | x \rangle = V \delta(x)$, the resonance energy is given by

$$\frac{1}{V} = \text{Re}[G^0(E_{\text{res}})], \hspace{1cm} (5)$$

given that $\text{Im}[G^0(E_{\text{res}})]$ is sufficiently small. Using an idealized insulator with $k$-independent valence and conduction bands separated by a band gap $E_g$, the bare Green’s function is $G^0(\omega, k) = (\omega - E_g/2 + i\eta)^{-1} + (\omega + E_g/2 - i\eta)^{-1}$, with $\eta$ infinitesimal small, and thus Eq. (5) gives $E_{\text{res}} \to 0$ as $V \to \infty$. If, on the other hand, the TI has a finite doping such that the Fermi energy $E_F = E_D + x$, where $E_D$ is the energy of the Dirac point and $|x| < E_g/2$ for $E_F$ to still be in the surface state, the same argument gives $E_{\text{res}} = -x$. That is, the resonance will always be situated at the Dirac point for a unitary impurity, independent of the doping of the system. We have confirmed this result numerically by including a finite chemical potential in Eq. (1). The above derivation is dependent on the valence and conduction bands being mirror-symmetric with respect to $E_D$ for all $k$-values. While this is
true in our model TI, it is in general not true in a real material. However, as long as valence and conduction bands are approximately mirror-symmetric in $E_D$ in the part of the Brillouin zone where the band gap is as smallest, we expect our results to still be qualitatively correct. If on the other hand, the energy difference $E_c$ between conduction band and Dirac point, and $E_v$ between valence band and Dirac point are different, the resonance energy is instead $E_{res} = -x + (E_c - E_v)/2$, resulting in a resonance peak away from the Dirac point for a bulk vacancy.

The LDOS for a finite $U$-impurity are very similar to those of a single vacancy. The main difference is that the resonance peak in general do not appear at or around $E = 0$, unless $U$ is large, and thus do not destroy the low-energy features of the Dirac surface state. There is also a clear trend that the deeper the impurity, the larger the $U$ needed for a low-energy resonance peak. This is clearly seen in in Figs. [g-l] where a $U = 40$ surface impurity is seen to destroy the original Dirac point, but where the same impurity in subsurface layers produces an impurity-induced resonance away from $E = 0$, alongside some layer dependent oscillation in peak energy position. Figure [I(l)] shows how a bulk $U = 40$ impurity clearly produces an in-gap resonance peak, which is associated with a state tightly bound to the impurity site. It was recently argued, based on results from a continuum model, that a non-magnetic \( \delta \)-function impurity cannot produce in-gap bound states in a 3D TI[13]. Our results, however, show that the closest lattice equivalent of a \( \delta \)-function, i.e. the single-site impurity, clearly produces in-gap bound states. This result is true as long as the impurity strength is large enough to put the resonance peak within the bulk gap. In our model system that means $U \gtrsim 20$. For smaller $U$ there is still a resonance state but it is located at energies above the bulk gap.

In Fig. [2] we analyze in more detail the resonance energy peak position $E_{res}$ in the surface spectra as function of both impurity layer position (a) and impurity strength (b). Since the resonance peak is non-dispersive, the peak energy position is the same in all layers. As seen clearly in Fig. [2(a)], the resonance peak appear at larger (negative) energies, i.e. farther from the low-energy region, for subsurface impurity positions. Thus for an impurity to influence the low-energy region of the surface Dirac spectrum it needs to be stronger the farther it is from the surface. It is also clear that the resonance peak move toward the low-energy region from larger (negative) energies as $U$ increases. This is equally true for both surface and subsurface positions. Apart from these trends, there is also a layer oscillation in the peak position but it quickly dies out as the impurity position approaches the bulk. We have here also included results for an AA terminated surface (thin lines, \( \circ \)) alongside the AB surface results (thick lines, \( \times \)). We note that the specifics of the layer oscillations are somewhat surface dependent as the AA surface termination produces slightly different results, but, in general, both surface terminations display remarkably similar results. In Fig. [2(b)] we plot the peak position as function of the inverse impurity strength $1/U$. For all impurity layer positions, including both the surface and the bulk, the peak position is proportional to $1/U$, with $E_{res} = k/U + m$. For AB surface termination, the slope $k$ is approximately constant between different impurity layer positions but the off-set $m$ varies for impurities close to the surface. For AA surface termination there is also some variation of the slope $k$ between layer positions. However, already for impurities in layer 4, the peak position is largely set by the bulk behavior (thickest line). The $1/U$-dependence for the resonance peak position in the bulk follow directly from the same $T$-matrix argument given above, whereas the $1/U$-dependence for surface impurities has also been established using a 2D continuum model for the surface states[14]. However, the resonance peak was in the latter case found to disappear at unitary scattering, something we most notably do not see in our microscopic lattice model. To summarize this section, we conclude that subsurface, and even bulk impurities, behave surprisingly similar to surface impurities, although a stronger impurity is needed in the subsurface situation in order to observe in-gap resonances. The non-dispersiveness of the resonance peak means that for any finite impurity-surface coupling, a resonance peak will also be present in the surface energy spectrum. We find that resonance peak traces are clearly present in the surface LDOS for impurities as far down as \( \approx 15 \) layers below the surface.

### B. Bulk vacancy clusters

The $E = 0$ resonance peak present for a single-site bulk vacancy is associated with a very tightly bound state around the vacancy site. Above we also showed that the $E_{res} = 0$
result is the same as that of a vacancy in an idealized normal insulator. On the other hand, Shan et al.\textsuperscript{13} recently used a continuum model to demonstrate the existence of bound states for a finite sized hole in a TI. In that case the bound states were simply a manifestation of the fact that a finite sized hole creates an interior surface in the TI. Holes with a very large radius $R$ possess a surface state very similar to that of a planar surface, although, technically, the surface state will have to obey periodic boundary conditions around the hole. As the radius $R$ becomes finite, the surface state turns into bound states with an energy separation which gets larger with decreasing $R$. Finally, for small enough holes all surface/bound states will be expelled to the bulk bands. Most notably, this continuum model do not produce $E = 0$ bound states for any size holes, unless $R \to \infty$. Clearly this result is at odds with our microscopic result for a single vacancy. To further shed light on this discrepancy we have studied highly-symmetric bulk vacancy clusters, involving as many as 17 sites, in order to increase the effective radius of our microscopically created hole. Figure 3(a) shows the LDOS on nearest neighbor sites to both a single-site vacancy (dashed line) and three different 5-site vacancy clusters. The diamond lattice has four nearest neighbors situated at the corners of a tetrahedron, a distance $\sqrt{3}a/4 \approx 0.43a$ from the center site. For such a 5-site nearest neighbor vacancy cluster, the resonance peak move up close to the bulk band gap at $E \approx 0.6$ (thick line). However, if we replace one of the nearest neighbors with a next-nearest neighbor, the impurity-bound state reappears close to $E = 0$ (black line). Further distortion by replacing two nearest neighbors with next-nearest neighbors creates a resonance state at $E = 0$ (red/grey line), the same result as for the single-site vacancy. We see in Fig. 3(c) how these peaks also show up as extremely small impurity resonances in the surface LDOS at the same energies when these vacancies are centered around layer 17. In Fig. 3(b, d) we show the same result for even larger vacancy clusters. The diamond (111) slab has six in-plane next-nearest neighbors and an additional six next-nearest neighbors out-of-plane, situated a distance $a/\sqrt{3} \approx 0.58a$ from the center site. An 11-site cluster including the four nearest neighbors and the six in-plane next-nearest neighbors creates a resonance around $E = 0$ (dashed line). When including all next-nearest neighbors into a fully-symmetric 17-site cluster, the resonance peaks move up to around $E = 0.4$ (thick line). However, distorting this 17-site cluster by exchanging only one next-nearest neighbor for a next-next-nearest neighbor again produces peaks in the very low-energy part of the spectrum (black and red/grey lines). We thus find that fully-symmetric vacancy clusters involving all nearest and next-nearest neighbor sites expels the impurity-bound states to high energies, in accordance with earlier continuum model results. Also, when increasing the radius from 0.43a for the nearest neighbor cluster to 0.58a for the next-nearest neighbor cluster, the resonance peak moves to slightly lower energies, in agreement with the continuum results. However, even the smallest possible distortion of either of these two clusters produces results more resembling those of a single-site vacancy, where the resonance peak sits firmly at $E = 0$. Thus, despite the topological origin of the surface state in a TI, there is a surprisingly large sensitivity to small cluster shape deviations. Since any microscopically sized hole in a TI will likely have some asymmetry, we conclude that even for fairly large such holes, the continuum limit will not be reached but a resonance peak will be present at or around $E = 0$.

**IV. CONCLUDING REMARKS**

Using a 3D microscopic lattice model of a strong TI we have shown that strong potential impurities and vacancies create low-lying impurity-bound resonance peaks in the surface LDOS for impurities as far as around 15 layers below the surface. This is also approximately the penetration depth of the surface state into the interior of the TI. This result shows how not only surface but also subsurface impurities will be important when analyzing STS measurements on a TI. There is an $1/U$-dependence for the resonance peak energy for impurities in any layer, including the bulk. However, subsurface impurities need to be stronger than surface impurities in order for the resonance peak to enter the low-energy region. On the other hand, any vacancy, within the penetration depth of the TI sur-
face state, always produces a peak in the LDOS at or very near the Dirac point, which is subsequently destroyed and split into two nodes that move off-center.

Recent STS data\textsuperscript{13} on non-magnetic unitary impurities in Bi$_2$Se$_3$ has shown very sharp energy resonance peaks at the Dirac point, with diverging strength as the Fermi level approaches the Dirac point. The experimental presence of such strong resonance states at the Dirac point confirms the need for a 3D model, which explicitly includes bulk states, since 2D continuum results do not find any strong resonance peaks near the Dirac point.\textsuperscript{14} Our results further show that the impurities do not necessarily have to be located on the surface, but can reside in subsurface layers as well. For surface impurities the resonance peak decay quickly, approximately as $1/R^3$ with surface distance and we find a similar dependence for subsurface impurities in the surface LDOS. This fast decay should be contrasted with a rather extended spread perpendicular to the surface. Experimentally, the resonance peaks were found to decay within as little as 2Å, which is in qualitative agreement with our results. Such fast decay signals a quick healing of the single Dirac point spectrum on an experimentally detectable level, which is expected for a topologically protected surface.

We have also studied bulk impurities and vacancies and find $E_{\text{res}} = 0$ peaks for single-site vacancies in the bulk. This result is consistent with a simple $T$-matrix calculation in a normal insulator, but does not agree with continuum model results for finite holes in a TI. To expand on this discrepancy we have also studied extended bulk vacancy clusters. We find that, while fully-symmetric 5 and 17-site clusters do not have any low-energy resonance states in agreement with continuum results, any asymmetry in the clusters produces $E_{\text{res}} \approx 0$ resonance peaks. Since any vacancy cluster of microscopic origin is likely to not be fully symmetric, we conclude that a microscopic approach is required for such holes. The $E_{\text{res}} \approx 0$ resonance peak for deep subsurface and bulk vacancies can have a profound effect on the surface conductivity. In the presence of a finite overlap between surface and vacancy states, the surface electrons will be scattered by these near zero-energy gap resonance states. In the limit of dense vacancy concentration, vacancy-band formation will allow edge-edge transitions, thus opening a gap in the topologically protected surface.

Acknowledgments

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