Proximity effect in graphene-topological insulator heterostructures

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We formulate a continuum model to study the low-energy electronic structure of heterostructures formed by graphene on a strong three-dimensional topological insulator (TI) both for the case of commensurate and incommensurate stacking. The incommensurability can be due to a twist angle between graphene and the TI surface, or a lattice mismatch between the two systems. For both the commensurate and the incommensurate case we find that the proximity to TIs induces splitting of the spin-degenerate bands of isolated graphene and the appearance of non-trivial spin textures.

The surface of strong three-dimensional (3D) topological insulators (TIs) \cite{1} and graphene \cite{2,3} have a very similar low-energy electronic structure. In both systems the conduction and the valence bands touch at isolated points, the Dirac points (DPs), and around these points the fermionic excitations are well described as massless two-dimensional (2D) chiral Dirac fermions for which the phase of a two-state quantum degree of freedom is locked with the momentum direction. However, there are also qualitative differences: (i) In graphene the chirality is associated with the sublattice degree of freedom whereas in a TI surface (TIS) it is associated with the electron spin; (ii) In graphene the number of DPs is even whereas in a TIS it is odd; (iii) In TIs the electron-phonon scattering is much stronger than in graphene. These differences imply that the transport properties of graphene \cite{4} and TIs are different in significant aspects: in graphene, without taking into account the negligible intrinsic spin-orbit (SO) coupling \cite{5-8}, no quantum spin Hall effect is expected, contrary to a TI; graphene is the material with the highest room-temperature mobility, whereas TIs have very low mobilities. These facts suggest that heterostructures formed by graphene and 3D TIs could exhibit very interesting behaviors by combining the properties of graphene and the TIS. In these systems the proximity to the TI is expected to enhance the SO coupling of graphene and create a novel 2D system with non-trivial spin textures and high, room-temperature, electron mobility. Moreover, recent works \cite{9,10}, have shown that graphene-TI heterostructure can be realized experimentally.

In this work we study the low-energy electronic structure of heterostructures formed by one sheet of graphene placed on the conducting surface of a 3D TI. In particular, we consider the TI material to be a Tetradyminite such as Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$. In these compounds the topologically protected surface states are found on the easy-cleaved 111 surface which has a three-fold rotational symmetry. The projected surface Brillouin zone (BZ) is hexagonal and a single DP is located at the zone center \cite{11}. Let $a_2$ be the effective lattice constant that corresponds to the surface BZ and $a_1 = 2.46\text{ Å}$ the graphene lattice constant. Defining $a_2/(\sqrt{3}a_1) \equiv 1 + \delta$, we have $\delta < 1\%$ for Sb$_2$Te$_3$ and $\delta \approx -3\%$ ($\delta \approx +3\%$) for Bi$_2$Se$_3$ (Bi$_2$Te$_3$). As a consequence the study of the commensurate $\sqrt{3}\times\sqrt{3}$ stacking pattern is expected to be a good approximation for heterostructures formed by graphene and Sb$_2$Te$_3$ and for developing the theory for incommensurate structures. We formulate a continuum model that is able to return the electronic, and spin, structure of graphene-TI heterostructures. Contrary to previous studies \cite{12}, our theory is able to take into account the incommensurability between the graphene layer and the TIS. As a consequence, our theory is valid also for graphene-TI heterostructures obtained via mechanical exfoliation \cite{13,20} in which the graphene lattice is normally not perfectly aligned with the lattice of the substrate, and a nonzero “twist” angle is present between the two lattices, causing graphene and the substrate to form an incommensurate structure, even when the ratio $a_2/a_1$ allows for a commensurate stacking. Twisted bilayer graphene \cite{21,37} and graphene on hexagonal boron nitride \cite{38,41} are examples of such incommensurate structures. Our results show that in graphene-TI heterostructures the SO coupling of graphene is greatly enhanced and induces non-trivial spin textures especially in the incommensurate cases. Moreover, to take into account the roughness of the interface, that we expect to be present in graphene-TI heterostructures obtained via the exfoliation technique, we present a simple case study in which tunneling processes with finite momentum transfer are not negligible and show that also in this case graphene-TI heterostructures can exhibit non-trivial spin textures.

The Hamiltonian describing the electronic degrees of freedom of the heterostructure can be written as $H = H^g + H^{TIS} + H_I$, where $H^g$ is the Hamiltonian for an isolated sheet of graphene, $H^{TIS}$ is the Hamiltonian for the TIS, and $H_I$ is the Hamiltonian describing tunneling processes between the graphene layer and the TIS. The long wavelength physics of graphene is described by a pair of 2D massless Dirac Hamiltonians: $H^g_{K} = \sum_{p} \sum_{\sigma,\tau,\tau'} c^\dagger_{K+p,\tau,\sigma} (\hbar v_{t}\tau \cdot \mathbf{p} - \mu_1)_{\tau\tau'} c_{K+p,\tau',\sigma}$ and $H^{K}_{TIS} = \sum_{p} \sum_{\sigma,\tau,\tau'} c^\dagger_{K+p,\tau,\sigma} (\hbar v_{t}^{\prime}\tau' \cdot \mathbf{p} - \mu_1)_{\tau\tau'} c_{K+p,\tau,\sigma}$, where $c^\dagger_{K+p,\tau,\sigma}$ ($c_{K+p,\tau,\sigma}$) creates (annihilates) a Dirac fermion on sublattice $\tau$ ($\tau'$) with spin $\sigma$ $(\uparrow, \downarrow)$ at a Dirac wave vector $\mathbf{p}$ measured from one of the two inequivalent BZ corners ($K$- and $K'$-valley) located at wave
vectors $\mathbf{K}$ and $\mathbf{K}'$ ($|p| \ll |\mathbf{K}|$), $\tau = (\tau^x, \tau^y)$ are Pauli matrices acting on the sublattice space, $v_1 \approx 10^6\text{m/s}$ is the Fermi velocity, and $\mu_1$ is the chemical potential. The low energy spectrum of graphene is characterized by four Dirac cones centered at two valleys with twofold spin degeneracy. The TIS states near its Dirac point can be described by an effective 2D continuum model [11]:

$$H_{\text{TIS}}^{\omega} = \sum_{\mathbf{k},\sigma} a_{\mathbf{k},\sigma}^\dagger [\hbar v_2 (\sigma \times \mathbf{k}) \cdot \hat{\mathbf{z}} - \mu_2]_{\sigma\sigma} a_{\mathbf{k},\sigma},$$

where $a_{\mathbf{k},\sigma}^\dagger$ (or $a_{\mathbf{k},\sigma}$) creates (annihilates) a surface massless Dirac fermion with spin $\sigma$ at wave vector $\mathbf{k}$ measured from the zone center ($\Gamma$-point), $\sigma = (\sigma^x, \sigma^y)$ are Pauli matrices acting on spin space, $\hat{\mathbf{z}}$ is the unit vector along the $z$ direction, and $\mu_2$ is the chemical potential. The TIS spectrum is characterized by a single Dirac cone with helical spin texture centered at the $\Gamma$-point. In Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$, the Fermi velocity $v_2$ is roughly half that of $v_1$ in graphene, in that the remainder we assume $v_2 = v_1/2$. The form of $H_t$ depends on the stacking pattern and the interface properties.

We first consider the graphene-TI heterostructure in a $\sqrt{3} \times \sqrt{3}$ commensurate stacking pattern, in which each TIS atom is directly underneath a carbon atom. The strongest tunneling is expected to occur between the directly stacked atoms, among which all the carbon atoms can be shown to belong to one sublattice (e.g., sublattice A). The BZs of decoupled graphene and TIS are shown in Fig. 1(a). As a result of the periodic tunneling potential, in the BZ of heterostructure the original graphene BZ is folded such that the two valleys are both located at the zone center overlapping with the TIS DP, as shown in Fig. 1(b). In this case the tunneling Hamiltonian can be written as $H_t = \sum_{\mathbf{k},\sigma} t_{\mathbf{A}} a_{\mathbf{k},\sigma}^\dagger e^{i\mathbf{k}_{\mathbf{A}} \cdot \mathbf{r}} + h.c.$, where $\mathbf{k} = \mathbf{K}, \mathbf{K}'$ and the tunneling matrix elements $t_A = t_A$, $t_B = 0$ are assumed to be spin and momentum independent. The Hamiltonian for such a structure takes the form

$$\hat{H}_k = \begin{pmatrix} \hat{H}_{k}^g & 0 & \hat{T}^\dagger \\ 0 & \hat{H}_{k}^T & 0 \\ \hat{T} & 0 & \hat{H}_{k}^{\text{TIS}} \end{pmatrix}, \quad \hat{T} = \begin{pmatrix} t & 0 & 0 \\ 0 & t & 0 \\ 0 & 0 & t \end{pmatrix},$$

where the graphene blocks are $4 \times 4$ matrices in sublattice and spin space whereas the TIS block is a $2 \times 2$ matrix in spin space.

The electronic structure can be obtained by directly diagonalizing the Hamiltonian matrix. On the other hand, key insights into the proximity effect of the TIS on graphene can be achieved using a perturbative approach [12], which has the advantage to be easily extended to cases in which the exact diagonalization is not feasible. In the perturbative approach the effect of tunneling processes on the graphene spectrum is captured by the self-energy $\Sigma_k(i\omega_n) = \hat{V}^\dagger G_k^0(i\omega_n) \hat{V}$, where

$$G_k^0(i\omega_n) = \left[ i\omega_n - \hat{H}_k^{\text{TIS}} \right]^{-1}$$

is the Green’s function of the TIS ($\omega_n$ are the fermionic Matsubara frequencies) and $\hat{V}$ is the tunneling vertex. The form of $\Sigma$ becomes transparent in the basis formed by the eigenstates of the Hamiltonian of isolated graphene, $\Phi_{\lambda,\mathbf{k},\alpha,\sigma}$ where $\alpha = \pm$ refer to the upper and lower bands that are fourfold degenerate:

$$\Sigma_k(i\omega_n) = \begin{pmatrix} \Sigma_k^S(i\omega_n) & e^{-i\mathbf{k}_{\mathbf{A}} \cdot \hat{\mathbf{z}}} \Sigma_k^A(i\omega_n) \\ e^{i\mathbf{k}_{\mathbf{A}} \cdot \hat{\mathbf{z}}} \Sigma_k^A(i\omega_n) & \Sigma_k^S(i\omega_n) \end{pmatrix} \otimes (I_\alpha + \sigma_\alpha^z) \otimes (I_\lambda + \sigma_\lambda^z),$$

where $\Sigma_k^{S/A}(i\omega_n) = (\bar{t}^2/2) G_k^{S/A}(i\omega_n)$ with $G_k^{S/A}(i\omega_n) = [1/(i\omega_n - \hbar v_2 k + \mu_2) + 1/(i\omega_n + \hbar v_2 k + \mu_2)]/2$. The first $2 \times 2$ matrix in Eq. [2] acts in the spin space, whereas $(I_\alpha + \sigma_\alpha^z)$ acts in the band space and $(I_\lambda + \sigma_\lambda^z)$ in the valley space, where $I$ is the $2 \times 2$ identity matrix. $\theta_{\mathbf{k}} = \arctan(k_y/k_x)$ represents the momentum direction. The appearance of non-zero off-diagonal spin components with phase factor $(\theta_{\mathbf{k}} - \pi/2)$ in the self-energy indicates an induced helical spin texture on some of the graphene bands. The renormalized graphene bands in the perturbative approach coincide with those obtained by direct diagonalization of [1]. Figure 1(c) shows the band structure for the case when $\mu_1 = \mu_2 = 0$ (the results for $\mu_1 \neq \mu_2$ are qualitatively similar) and $t = 45\text{meV}$. We see that the tunnel degeneracy is partially lifted with two bands gapped and split. The band structure and spin texture can be understood by looking into the band of the renormalized eigenvectors: $\Psi_{r,k}$ ($r =$...
Interestingly, four of the eigenvectors are simply antisymmetric combinations of the states of isolated graphene at opposite valleys: $\frac{1}{\sqrt{2}} (\Phi_{K,\alpha,1} - \Phi_{K',\alpha,1})$ and $\frac{1}{\sqrt{2}} (\Phi_{K,\alpha,2} - \Phi_{K',\alpha,2})$ for $\alpha = \pm$. The mixing of different valley states, mediated by the TIS states, gives rise to a reconstructed Dirac cone with twofold spin degeneracy in the low energy spectrum. The other four bands acquire a non-trivial spin texture and become gapped and split. By calculating the expectation values $S^{\nu}_{\tau} = \frac{1}{2} \sum_{\lambda, \sigma} \frac{1}{\Omega} \Psi^{\nu}_{\tau, \lambda, \alpha, \sigma} \Psi^{\nu}_{\tau, \lambda, \alpha, \sigma}$ with $\nu = x, y, z$, we obtain the spin configuration on the renormalized bands. As expected from the form of the self-energy, the results, in Fig. 1(d), show that for the gapped bands the in-plane spin is locked perpendicular to the momentum direction and winds around the $\Gamma$ point either clockwise or counterclockwise, analogous to a system with Rashba-type SO coupling. We have also considered the system with bilayer graphene (BLG) instead of the single layer graphene (SLG). Figure 1(e) shows the renormalized band structure for BLG-TIS. We find that the effective Rashba-type splitting induced by the proximity to the TIS is strongly enhanced in BLG compared to SLG. Figure 1(f) plots the band splitting $\Delta_{R}$ in the two systems as a function of $t$.

We now consider the incommensurate structures. A general expression for the tunneling matrix elements between graphene and TIS can be written as:

$$T_{\tau}(k_{2}, k_{1}) = \sum_{G_{1}, G_{2}} \frac{t(k_{1} + G_{1})}{\sqrt{3} \Omega_{1}} e^{i G_{1} \cdot d_{r}} \delta_{k_{2} + G_{2}, k_{1} + G_{1}}$$

where the crystal momentum is conserved by the tunneling process in which a graphene quasiparticle of wave vector $k_{1}$ residing on sublattice $\tau$ hops to a TIS state with wave vector $k_{2}$ (both $k_{1}$ and $k_{2}$ are measured from the first BZ center). Here $\Omega_{1}$ is the unit cell area of graphene and $d_{A} = 0$, $d_{B} = (-a_{0}, 0)$ are the positions of the two carbon atoms in a unit cell at carbon-carbon distance $a_{0}$. $\{G_{1}\}, \{G_{2}\}$ are the reciprocal lattice vectors of graphene and the TIS, respectively. $t(k)$ are the Fourier amplitudes of the tunneling potential $t(r)$ assumed to be a smooth function of $r$, the separation between graphene and TIS atoms projected onto the interface plane. Analogous to twisted bilayer graphene [29], given that the graphene-TIS separation distance exceeds the inter-atomic distance in each material, the dominant tunneling amplitudes of $t(k)$ near the graphene Dirac point are the ones with $|k| = K_{D}$, where $K_{D} \equiv |K|$. This allows to restrict the sum over $\{G_{1}\}$ to three vectors: $g_{1} = (0), g_{2}, g_{3}$, where the latter two connect a valley with its equivalent first BZ corners. For small wave vectors measured from the respective Dirac points, we have the tunneling Hamiltonian $H_{t} = \sum_{p, \tau, \sigma} \sum_{j, l, \ldots} \{T_{\tau,j} a_{p+q_{j}, \sigma}^{\dagger} c_{p, \tau, \sigma}^{\dagger} + T_{\tau,j} c_{p, \sigma} a_{p+q_{j}, \sigma} + \ldots\}$, where $T_{\tau,j} = t' e^{i g_{j} \cdot d_{r}}$ with $t' \equiv t(K_{D}) / (\sqrt{3} \Omega_{1})$, $\{q_{j}\}$ are the offset vectors between the graphene DP and the three “nearest-neighbouring” TIS DPs, and $q_{j} = |q_{j}| = 2 K_{D} \sin(\theta/2)$.

For very small twist angles or lattice mismatches such that the dimensionless parameter $\gamma \equiv \frac{t'}{\alpha_{DP}} > 1$, graphene and TIS will be strongly coupled. However, when $\gamma < 1$, a weak coupling theory is valid [21][29][31]. In this case, to investigate the low-energy spectrum of graphene, we truncate the $k$-space lattice and obtain the Hamiltonian:

$$\hat{H}_{p} = \begin{pmatrix} H_{p}^{K} & \hat{T}_{1}^{\dagger} & \hat{T}_{2}^{\dagger} & \hat{T}_{3}^{\dagger} \\ \hat{T}_{1} & 0 & 0 & \hat{T}_{3}^{\dagger} \\ \hat{T}_{2} & \hat{T}_{3}^{\dagger} & 0 & \hat{T}_{2}^{\dagger} \\ \hat{T}_{3}^{\dagger} & \hat{T}_{2}^{\dagger} & \hat{T}_{3}^{\dagger} & 0 \end{pmatrix},$$

$$\hat{T}_{1} = \begin{pmatrix} t' & t' & 0 & 0 \\ 0 & 0 & 0 & t' \end{pmatrix},$$

$$\hat{T}_{2} = \begin{pmatrix} t' e^{-i \frac{2\pi}{2}} & 0 & 0 \\ 0 & 0 & 0 & t' e^{-i \frac{2\pi}{2}} \end{pmatrix},$$

$$\hat{T}_{3} = \begin{pmatrix} t' e^{i \frac{2\pi}{2}} & 0 & 0 \\ 0 & 0 & t' e^{i \frac{2\pi}{2}} \end{pmatrix}.$$
original twofold spin degeneracy of the graphene Dirac cone is completely lifted; (ii) of the two original degenerate linear bands one is now fully gapped and the other is no longer linear at the DP; (iii) the bands acquire in-plane spin textures. Figure 3(b)-(d) reveal the evolution of the spin texture from high to low energy cuts on the low-energy bands indicated in Fig. 3(a). The induced spin texture around the graphene DP (at the origin in Fig. 3(b)-(d)) on the gapless band can be interpreted as a smooth interpolation between the spin helices of the neighbouring TIS cones on the high-energy connected band (Fig. 3(b)). Correspondingly, the gapped band acquires an in-plane chiral spin texture in which the spin direction is almost antiparallel to the momentum direction. At lower energy (Fig. 3(c)), the bands become disconnected, composed of five pockets, two centered at the graphene DP and the other three associated with the three offset TIS DPs. The spin configuration on the graphene pockets can be understood as a continuous evolution from that at higher energy, exhibiting chiral structure with spin approximately parallel or antiparallel to the momentum. At very low energy (Fig. 3(d)), the inner graphene pocket disappears and only the outer chiral spin pocket is left.

In the case of large twist angles or lattice mismatches, the graphene and the TIS Dirac cones are well separated. The tunneling processes that conserve the crystal momentum are strongly suppressed due to the large momentum mismatch between the low-energy states of graphene and the TIS. However, the presence of surface roughness and/or phonons may allow for tunneling processes with finite momentum transfer. We expect that in general the effect of such processes might be weak, however, to gain some insight on such effect, we consider the case in which the tunneling amplitude has a Gaussian profile with respect to the momentum transfer \( q \): \( t_0 = t_0 \exp(-|q|^2/(2\sigma^2)) \), where \( t_0 \) characterizes the tunneling strength and \( \sigma \) is the variance. To qualitatively understand the effect of such processes, we study the case of an isolated graphene Dirac cone separated by a large wave vector \( Q \) from the closest TIS Dirac cone. Using the perturbative approach outlined above, the proximity effect on the graphene is taken into account by the self-energy

\[
\hat{\Sigma}_{Q+p}(i\omega_n) =
\begin{pmatrix}
\Sigma_{Q+p}^S(i\omega_n) \\
\Sigma_{Q+p}^A(i\omega_n)
\end{pmatrix}
= 
\begin{pmatrix}
\Sigma_{Q+p}^S(i\omega_n) \\
\Sigma_{Q+p}^A(i\omega_n)
\end{pmatrix}
\begin{pmatrix}
e^{-i(\theta_Q + p - \frac{\pi}{2})} & \Sigma_{Q+p}^A(i\omega_n) \\
\Sigma_{Q+p}^A(i\omega_n) & \Sigma_{Q+p}^S(i\omega_n)
\end{pmatrix}
\otimes (I_n + \sigma^x_n)
\] (5)

with

\[
\Sigma_{Q+p}^S(i\omega_n) = \frac{\Omega \Omega_S}{2\pi} \exp \left[-\frac{|Q+p|^2}{\sigma^2} \right] \int_0^\infty k \exp \left[-\frac{k^2}{\sigma^2} \right] \times
\int_0^{2\pi} \frac{2Q+p}{\sigma^2} k G_{Q+p}^S(i\omega_n) dk,
\]

where \( I_n(x), n = 0, 1 \) are the modified Bessel functions of the first kind. The form of the phase factors in the off-diagonal spin components of \( \hat{\Sigma} \) implies an induced spin texture on graphene with the spin direction perpendicular to the wave vector \( Q + p \), Fig. 4(a). We find, Fig. 4(b), that also in this case the spin degenerate bands are split and that the remaining gapless bands are no longer linear. The calculated spin configuration is demonstrated in Fig. 4(a). Figures 4(c)-(d) show the size of the gap between spin-split bands as a function of \( t_0 \) and \( \sigma \), respectively. We see that for small values of \( \sigma \) the gap grows rapidly with increasing \( \sigma \).

In conclusion, we have studied the proximity effect of a strong 3D TI on the low-energy spectrum of graphene.
in commensurate and incommensurate structures as well as in a case with surface roughness. Our results show that in these systems the proximity effect induces a spin-splitting of the graphene bands with the appearance of non-trivial in-plane spin textures. Our results suggest that graphene-TI heterostructures have interesting properties both from a fundamental perspective and for technological applications, due to the unique coupling between the different degrees of freedom of their electronic states.

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[47] Note that the Dirac Hamiltonian of graphene in the sub-lattice basis has a phase factor on the off-diagonal terms to take into account a rotational transformation of coordinates due to the choice of the current BZ orientation.