Driven Electronic States at the Surface of a Topological Insulator

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Motivated by recent photoemission experiments on the surface of topological insulators we compute the spectrum of driven topological surface excitations in the presence of an external light source. We completely characterize the spectral function of these non-equilibrium electron excitations for both linear and circular polarizations of the incident light. We find that in the latter case, the circularly polarized light gaps out the surface states, whereas linear polarization gives rise to an anisotropic metal with multiple Dirac cones. We compare the sizes of the gaps with recent pump-probe photoemission measurements and find good agreement. We also identify theoretically several new features in the time-dependent spectral function, such as shadow Dirac cones.

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Introduction – Topological properties of materials manifest in the appearance of electronic states at the boundary. An especially interesting class of such topological boundary modes arises in three dimensional (3D) topological insulators (TI) and they have now been detected in several material systems. Furthermore, a new possibility for creating topological band structures in non-equilibrium was recently suggested, where an initially topologically trivial semiconductor is converted into a topological insulator via an external irradiation. The resulting state was dubbed a Floquet topological insulator and an analogue to such a state was recently realized experimentally in a photonic system. In the same vein, other theoretical works have studied the realization of a lattice quantum Hall state with time-periodic perturbations.

The focus of these previous researches has been on turning an electronic system with a topologically-trivial band structure into a topological insulator by subjecting it to a periodic-in-time perturbation. Here, on the contrary, we study the effect of an external irradiation on an initially topological state. The motivation comes from the development of new experimental probes that make it possible to access the time-resolved excitation spectrum of driven electrons at the surface of TIs using time-resolved photoemission spectroscopy. Below, we focus specifically on the properties of driven Dirac electrons on the surface of existing 3D topological materials such as Bi2Se1−x Te x alloy, Bi2Te3 and Bi2Se3 (Ref. 15). We are particularly interested in describing the modification of the spectrum of the boundary modes due to the irradiation as a function of the parameters of the incident light. The spectrum is composed of Floquet bands of the driven Dirac Hamiltonian, as discussed in the previous related works and 12 We unify and extend these previous analysis and apply our results specifically to describe an experiment, which observed an induced energy gap in driven surface states of Bi2Se3 using time- angle-resolved photoemission spectroscopy (TrARPES) on (Ref. 15). We find that our results fit the data well.

Model – We consider non-interacting electrons at the surface of a TI with incident light normal to the surface. The Hamiltonian is

\[ H(k, t) = H_0(k) + H_{ext}(t), \]

\[ H_0(k) = v(k_x \sigma_y - k_y \sigma_x), \]

\[ H_{ext}(t) = V \Theta(t - t_0)(a_x(t) \sigma_y - a_y(t) \sigma_x), \]

where \( H_{ext}(k) \) describes the perturbation. The electrons are particularly interested in describing the modification by the Peierls substitution \( k \to k - eA(t) \), where \( A(t) \) is the vector potential. We set \( \hbar = 1, c = 1 \) and the scalar potential to zero. We also ignore small magnetic effects. Two polarizations considered are \( a(t) = (\pm \cos \Omega t, \sin \Omega t) \) (circular) and \( a(t) = (\cos \Omega t, 0) \) (linear), where \( T = 2\pi/\Omega \) is the period of the external perturbation. The energy scale of the perturbation is given by \( V = evA_0 = evE_0/\Omega \) where \( E_0 \) is the amplitude of the electric field. We consider large perturbations, e.g. \( V/\Omega \approx 0.52 \), where perturbation theory is not valid and energies below the bulk gap of the TI (< 300 meV), as the Dirac Hamiltonian is only valid in this regime.

The evolution operator, which is a \( 2 \times 2 \) matrix, obeys the time-dependent Schrödinger equation \( i\partial_t U_k(t, t') = H(k, t)U_k(t, t'), \) with initial condition \( U_k(t, t) = 1. \) Once the evolution operator is known, all other correlators can be calculated in terms of the initial state of the system. We are interested in computing the retarded Green function which is given by \( g^{<}_{\alpha\beta}(k, t, t') = -i\Theta(t - t')\langle \{c_{k\alpha}(t), c_{k\beta}^\dagger(t')\} \rangle \equiv -i\Theta(t - t')U_{k\alpha\beta}(t, t'), \) where \( U_{k\alpha\beta} \) is the \( (\alpha, \beta) \) matrix element of \( U_k. \) Similarly the lesser Green function is \( g^{\langle}_{\alpha\beta}(k, t, t') = i\langle c_{k\beta}^\dagger(t')c_{k\alpha}(t) \rangle \approx iU_{k\alpha\beta}^\dagger(t', t_0)\langle c_{k\beta}^\dagger(t_0)c_{k\alpha}(t_0) \rangle U_{k\alpha\beta}(t, t_0) \)

We use the convention of summation over repeated indices.

We set \( t_0 = -\infty \) so that there is no memory of the initial conditions. Then the evolution operator satisfies \( U_k(t + T, t' + T) = U_k(t, t') \), and the retarded Green function is periodic in \( t = (t + t')/2 \) with period \( T. \) This structure allows for a formal (Wigner) representation similar to the Lehmann representation of equilibrium correlators. To see how this comes about, we use concepts
from Floquet theory reviewed in Ref. 19. Indeed, the wave function can be written as \( \psi(t) = \phi(t)e^{-i\epsilon t} \), where \( \phi(t) = \phi(t + T) \) and the phase \( \epsilon \) is the quasienergy, defined modulo \( \Omega \). Substituting into the Schrödinger equation gives the eigenvalue problem \( H_F\phi(t) = (H(t) - i\partial_t)\phi(t) = \epsilon\phi(t) \), where we suppressed the momentum label for brevity. The same equation in frequency space reads \( \sum_{\alpha\beta}(\alpha|H_F|\beta)m\langle \beta|\phi_{\gamma}^n\rangle = \epsilon_{\gamma}m(\alpha|\phi_{\gamma}^n) \), where the so-called Floquet Hamiltonian is defined as \( (\alpha|H_F|\beta)m = H_{n-m} + i\Omega\delta_{\alpha\beta}\delta_{nm} \) and the quasienergies as \( \epsilon_{\gamma}n = \epsilon_{\gamma} + n\Omega \). \( H^n \) is the \( n \)th Fourier mode of \( H(t) \). By direct substitution one can show that the evolution operator can be written as \( U_{\alpha\beta}(t,t') = \sum_{\gamma\eta} \langle \alpha|\phi_{\gamma}^n\rangle \langle \phi_{\gamma}^n|\beta\rangle e^{-i\epsilon_{\eta}m(t-t')+in\Omega t} \). \( \langle \alpha|\phi_{\gamma}^n\rangle \) represents the \( \alpha \) component of the eigenvector \( |\phi_{\gamma}^n\rangle \) of the Floquet Hamiltonian. Putting the momentum label back, substituting into the equation for the retarded Green function above and Fourier transforming in the relative time, \( t_s = t-t' \), and average time, \( t = (t+t')/2 \), we obtain the Wigner representation\(^\text{20}\) of the retarded Green function,

\[
g_{\alpha\beta}(k,n,\omega) = \sum_{\gamma\eta} \frac{\langle \alpha|\phi_{\gamma}^n\rangle \langle \phi_{\gamma}^n|\beta\rangle}{\omega - \epsilon_{k\gamma\eta} + n\Omega/2 + i0^+}. \tag{4}
\]

One can similarly obtain simple expressions for the inverse of the retarded Green function by noting that \( U_k(t,t')^{-1} = U_{k}(t',t) \). Eq. \( 4 \) is similar to Eq. 46 of Ref. \(^\text{21}\) and \(^\text{22}\) for a two-band model. The important point is that single-particle excitations, which occur at the poles of the spectral function, are given by the quasienergies of the driven Dirac Hamiltonian, Eq. \( 1 \). The index \( n \) represents the number of photons contributing (also the order of perturbation theory in \( V \)). For example, a one-photon resonant transition creates exited states shifted by \( \pm \Omega/2 \) with respect to the original Dirac bands. To see this, expand the Green function in Pauli matrices: \( g = g_0 + g_\sigma \). After taking the trace, only \( g_0 \) remains. The zero-th and first order contributions, for circular polarization, are

\[
g^{(0)}_0(k_x,\tilde{t},t_r) = -i\Theta(t_r)\cos(vk_x t_r),
\]

\[
g^{(1)}_0(k_x,\tilde{t},t_r) = -\frac{2iV}{\Omega}\Theta(t_r)\sin(vk_x t_r)\sin\left(\frac{t_r\Omega}{2}\right)\cos(\tilde{t}\Omega),
\]

where \( k_y = 0 \). See that to zeroth order, we obtain the usual Dirac-like dispersion \( \pm ik_x \) corresponding to the eigenvalues of \( H_0 \). To first order, the Green function is explicitly time-periodic in \( t_r \), with sharply defined excitation bands at \( \pm ik_x + \Omega/2 \). In general, the spectral weight of odd-\( n \) bands is time-dependent, as expected from linear response theory. This dependence on \( n \) was not considered in Ref. \(^\text{11}\).

We are interested in the single-particle excited states which are given by the singularities of the non-equilibrium spectral function. In the Wigner representation, this function is given by

\[
A(k,n,\omega) = -2\text{Im}[\text{Tr}[g(k,n,\omega)]] = -2\text{Im}\left[\sum_{\gamma\eta} \frac{\langle \alpha|\phi_{\gamma}^n\rangle \langle \phi_{\gamma}^n|\alpha\rangle}{\omega - \epsilon_{k\gamma\eta} + n\Omega/2 + i0^+}\right]. \tag{5}
\]

Of particular interest is the average over a period \( T \) of the driving force, which is just the \( n = 0 \) term:

\[
A(k,0,\omega) = 2\pi \sum_{\gamma\eta} |\langle \alpha|\phi_{\gamma}^n\rangle|^2\delta(\omega - \epsilon_{k\gamma\eta}). \tag{6}
\]

It satisfies the sum rule \( \int (d\omega/2\pi)A(k,0,\omega) = 2 \). We note that this property is not shared by any other moment of the non-equilibrium spectral function. To obtain it we used the completeness of the \( |\phi_{\gamma}^n\rangle \) states. Note that in equilibrium systems, the spectral function does satisfy this sum rule which, in that case, derives from fermion conservation and the factor of two comes from the spin. In this sense, the average calculated above is more physical than the non-equilibrium spectral function, by itself.

Let us consider the general structure of \( \epsilon_{k\alpha} \). See the similar spectra of graphene\(^\text{12,16,17}\). Usually, one can accurately obtain the quasienergies by diagonalizing a truncated version of the Floquet Hamiltonian, \( (\alpha|H_F|\beta)m = H_{n-m} + i\Omega\delta_{\alpha\beta}\delta_{nm} \). For example, in Fig. \( 1 \) we consider circular and linear polarizations with six modes as a function of momentum along \( k_x \) and \( k_y \).

We verified higher modes do not change the spectrum significantly in the range of energies we consider. One can understand the structure of the spectrum as composed of copies of the original Dirac bands shifted by...
Hence the gap at the Dirac point is $\frac{1}{2}$ magnetic field. An explicit computation gives $U$ formally equivalent to a spin-1/2.

The few analytically solvable driven two-level models are associated with symmetries of the Hamiltonian $\mathcal{H}_F$. If there are no symmetries, any crossings/degeneracies are accidental. An early result of Von Neumann and Wigner for time-independent Hamiltonians establishes that two (three) parameters are necessary to produce an accidental degeneracy for real (complex) Hamiltonians. Hence by varying only one parameter, such as $k_x$ or $k_y$, we expect to produce only avoided crossings.

Consider the case of circularly polarized photons. They break time reversal symmetry, and since no other symmetries remain we expect only avoided crossings (Fig. 1b, b). If the perturbation is small, $V/\Omega \ll 1$, we can restrict the analysis to the two crossing bands in question. For concreteness, consider $v k_x \approx \Omega /2$ and $k_y = 0$. The effective Hamiltonian is a $4 \times 4$ matrix,

$$H_{2v k_x=\Omega} = \begin{pmatrix} H_0 + \Omega & i V \sigma^- & 0 \\ -i V \sigma^+ & H_0 & i V \sigma^- \\ 0 & -i V \sigma^+ & H_0 - \Omega \end{pmatrix},$$

(7)

where we used $H_{ext}(t) = i V \sigma^- e^{i \Omega t} + h.c.$ and $\sigma^\pm = (\sigma_x \pm i \sigma_y)/2$. In the absence of the external perturbation, the eigenvalues of the above matrix are $vk_x, -vk_x, vk_x + \Omega,$ and $-vk_x + \Omega$. Two bands cross at $vk_x = \Omega /2$. Hence, for small $V/\Omega$ the gap is $O(1)$, $\Delta^{circ}/\Omega \approx V/\sqrt{V^2 + \Omega^2} = V/\Omega + \cdots$. These results agree with previous perturbative calculations using the rotating wave approximation.

Note that for large $V/\Omega$, the resonance occurs at lower momentum, $vk_x < \Omega /2$ (Fig. 1b, b). This Bloch-Siegert shift is an effect beyond the scope of the rotating wave approximation.

Next we consider the Dirac point where a gap is photoinduced due to the absorption and subsequent emission of a photon by an electron near the Dirac point. The effective Hamiltonian is

$$H_{k=0} = \begin{pmatrix} H_0 + \Omega & 0 & i V \sigma^- \\ 0 & H_0 & 0 \\ -i V \sigma^+ & 0 & H_0 - \Omega \end{pmatrix}. \quad (8)$$

In the limit of $V/\Omega \ll 1$ the gap is $\Delta^{circ} \approx 2(V^2/\Omega)$. Here we provide an alternative derivation by noting that at the Dirac point, the Hamiltonian in Eq. (8) is one of the few analytically solvable driven two-level models, formally equivalent to a spin-1/2 in a circularly polarized magnetic field. An explicit computation gives $U_0(t,t') = e^{-i \Omega t/2} e^{-i H_F(t-t')} e^{i \Omega t'/2}$, where $H_F = V \sigma_y - \Omega \sigma_z$. Hence the gap at the Dirac point is

$$\Delta_0^{circ} = \sqrt{\Omega^2 + 4 V^2} \mod \Omega. \quad (9)$$

Shadow Dirac points – In Fig. 1a, d, we see level crossings at the Dirac point and finite momentum arising from a dynamical symmetry of $\mathcal{H}_F$. To see this, consider the Floquet operator for linear polarization along $x$, $v_k \sigma_z + V \cos \phi_k \cos(\Omega t) \sigma_z - V \sin \phi_k \cos(\Omega t) \sigma_y - i \phi_k$, in the basis of eigenvectors of $H_0$. $\phi_k$ is the angle of $k$ and the $x$-axis. If the momentum is along $k_x$, the perturbation commutes with $H_0$ and produces the trivial spectrum in Fig. 1a with Dirac points at $vk_x = \alpha \Omega$. If the momentum is along $k_y$, the perturbation commutes with $H_0$ and produces the trivial spectrum in Fig. 1a with Dirac points at $vk_y = \alpha \Omega$. Hence states with well defined parity, $\phi_{\alpha n}$, can be defined according to whether $\alpha + n$ is even or odd. The perturbation does not have matrix elements between states of different symmetries. This can be explicitly shown by writing $\mathcal{H}_F$ in frequency space in the basis of eigenvectors of $H_0$ and noting that it splits into disjoint blocks $\mathcal{H}_F = \mathcal{H}_{even} \oplus \mathcal{H}_{odd}$. One implication is that the $k = 0$ point remains gapless,

$$\Delta^{lin}_{\alpha 0} = 0, \quad (10)$$

to all orders in perturbation theory, as the bands $\epsilon_{k_1,0}, \epsilon_{k_2,0}$ have different parities, see also Ref. 17. For the same reason, there are symmetry-protected band crossings at finite momentum between $\epsilon_{k_1,odd}$ and $\epsilon_{k_2,odd}$, e.g., $\epsilon_{k_1,-1}$ and $\epsilon_{k_2,1}$ (Fig. 1a). Note that for arbitrary direction in momentum space, other than $k_x$ and $k_y$, the states have no well defined parity, degeneracies are not symmetry-protected, and hence gaps develop.
quasienergies cross, the dynamics of the system can significantly slow down, similar to the coherent destruction of tunneling
effect. In Fig. 1, we see the crossing of quasienergies (shadow Dirac points) at $\pm k_y^*$. By requiring zero energy solutions of the eigenvalues of $H_F$, it can be shown that $v k_y^*/\Omega = (1/2)(10 - 2(V/\Omega)^2 - 36 + 8(V/\Omega)^2 + (V/\Omega)^6)^{1/2}$, where $V = 0.5292$ as in Fig. 1, $k_y^* \approx 0.9192$. As momentum increases, with fixed $V/\Omega < 1$, the crossings asymptote to $v k_y^* = n\Omega$ (see Ref. 23). However the crossing of $\epsilon_{k_1,0}$ and $\epsilon_{k_2,1}$ bands can be gapped as these bands belong to the same symmetry class. Indeed, there is a gap of $O(V)$ at momentum $v k_y \approx \Omega/2$. By using similar arguments as for circular polarization, we find that this gap is $\Delta_{lin}^c = V/\Omega < 1$. From this we see that the number of Dirac points can be \textit{engineered} with a properly chosen frequency.

Discussion – In a TrARPES experiment, the measured photo-current is proportional to the lesser Green function, which in turn is proportional to the distribution function (generally unknown) and the spectral function. Under the assumption of a smooth distribution function, we expect the features we found in this work such as the crossings and anti-crossings be generic. Also, for a system were there is no phase coherence between the pump and probe pulses, we expect the photocurrent to time-average over the period of the driving force. Then the measured spectrum would be characterized qualitatively by the average of the spectral function, Eq. [2] calculated numerically in Fig. 2. For concreteness, if the electric field is $E_0 \approx 2.2 \times 10^5$ V/m and the photon energies are $120$ meV then using $v = 5 \times 10^{5}$ m/s as the speed of Dirac electrons on the surface of Bi$_2$Se$_3$, we obtain a coupling $V/\Omega = 0.52$. Using Eq. [9] we obtain a gap $\Delta_{lin}^c = 51$ meV, in agreement with our simulation in Fig. 2 and the experiment. $\Delta_{lin}^c = 53 \pm 4$ meV. At finite momentum and circular polarization, we obtain $\Delta_{circ}^c = 37$ meV along $k_x$ and $k_y$, and so the spectrum is isotropic (see Fig. 2a, b). For linear light, we obtain $\Delta_{lin}^c = 62$ meV at $k_y$, in agreement with the experiment. $\Delta_{lin}^c = 62 \pm 5$ meV. The position of the first shadow Dirac point is $v k_y^* = 109$ meV.

In conclusion, we have calculated the non-equilibrium spectral function of electrons at the surface of TIs in the presence of an incident light with circular and linear polarization. Depending on polarization we observed crossings and anti-crossings in the spectrum which can be explained with symmetry arguments as discussed above. This theory along with the experimental technique would allows for optical engineering of non-equilibrium spectra in topological materials.

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