Dynamical density response and collective modes of topological insulator ultra-thin films

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We analytically calculate the intra- and inter-surface dynamical density-density linear responses of ultra-thin topological insulator films with finite tunneling between their top and bottom surfaces in both metallic and insulating regimes. Employing the random phase approximation we investigate the dispersions of in-phase and out-of-phase collective density modes of this system in the metallic regime. We find that in contrast to the bilayers of the conventional two-dimensional electron gas, where finite tunneling gapes out the out-of-phase mode, in topological insulator thin films, this mode remains linear at long wavelengths. Depending on different system parameters, the velocity of out-of-phase mode can be tuned to be larger or substantially smaller than the Fermi velocity of electrons on the isolated surfaces of the topological insulator. Finite tunneling generally reduces the energy of collective modes, making them more confined in space.

I. INTRODUCTION

Plasmons are collective charge density excitations originating from the long-range nature of the Coulomb interaction. Recently, collective modes of two dimensional Dirac materials have attracted a lot of interest [1–10]. Plasmons in these materials are tunable through gate voltage which controls the carrier concentration, and usually, have higher lifetimes due to the high mobility of host materials. Plasmons have interesting features in topological insulators, due to the strong spin-orbit coupling [11]. They have long propagation lengths and resonance frequencies in the mid-infrared and terahertz spectral regions, that can be tuned via the Fermi energy [12]. Density oscillations in topological insulators are accompanied by transverse spin oscillations (i.e., spin-plasmons) as a result of spin-orbit coupling [13]. These peculiar features suggest that the collective modes of topological materials have the potential for novel applications in plasmonics and spintronics. Due to the quantum confinement, the bulk band-gap of topological insulator thin film (TITF) is larger than the bulk topological insulator [14]. This is a useful feature as it provides a larger range of available chemical potentials for the surface states, within the gap of bulk states. Nano-scale structures of the topological insulators, such as thin films, multilayers, and nano-ribbons have large surface-to-volume ratios, so the contribution of surface states in their different physical properties are enormously enhanced. It has been experimentally [15, 16] observed that for Bi\textsubscript{2}Se\textsubscript{3} topological insulator thin films when the film thickness becomes less than 6 quintuple layers (∼ 6nm), a finite hybridization between two surfaces opens a gap in the excitation spectrum of surface states. In the following by ultra-thin films, we mean the regime where the tunneling between two surfaces is not negligible. Note that a finite gap in the spectrum of surface states of TITF could be induced by other means such as strain and external electric or magnetic fields.

Plasmons have been extensively studied in different double layer structures [17–28]. The collective density modes of topological insulator thin films in the absence of hybridization between their surface states have been investigated recently [26–28]. Electrons in uncoupled TITF behave like massless Dirac fermions, therefore, their dynamical density response function is similar to the one of graphene [29]. Within the random phase approximation (RPA), it was shown that uncoupled TITF has two collective modes, optical and acoustic modes respectively with the usual ω ∝ \sqrt{q} and ω ∝ q long-wavelength dispersions [27].

In this paper, we study the dynamical density-density response and the collective density modes of a topological insulator ultra-thin film, in the regime where the surface electronic states are hybridized due to finite inter-surface tunneling. Electrons in tunnel-coupled TITF behave like massive Dirac fermions so their total density response at low doping is similar to the density response of massive Dirac fermions [9], such as in gapped graphene [3] and other buckled honeycomb lattices [7]. However, in topological insulator thin films it is possible to probe surface-resolved density responses and therefore two distinct collective modes corresponding to the \textit{in-phase} and \textit{out-of-phase} oscillation of electrons in two surfaces is expected. Collective density modes of topological insulator films and double-layer graphene, in the absence of tunneling, have been theoretically investigated by several groups [6, 24, 26–28]. Here we look at the effects of hybridization between the surface states of topological insulators in the ultra-thin limit. The static density response and screening of this system have been explored by Liu \textit{et al.} [30].

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The rest of this paper is organized as follows. In Sec. II we introduce our effective low-energy model Hamiltonian for topological insulator thin film, obtain its intra- and inter-surface density response functions and discuss their behavior in different regimes. In Sec. III we discuss the dispersions of collective density modes of TITF, and investigate their long-wavelength behavior. Sec. IV summarizes our main findings. Finally, the full analytic forms of the dynamical density response functions in different regimes and their asymptotic behavior are presented in Appendix. A.

II. MODEL HAMILTONIAN AND LINEAR DENSITY-DENSITY RESPONSES

The effective low-energy single-particle Hamiltonian of a TITF is given by [30–34]

\[ \hat{H}_k = \hbar v_F \tau_z \otimes [\sigma \cdot (k \times \hat{z})] + t \tau_x \otimes \sigma_0, \tag{1} \]

where \( v_F \) is the Fermi velocity of the surface states, \( \tau \) and \( \sigma \) are the Pauli matrices acting in the layer and real spin spaces, respectively and their zero components are \( 2 \times 2 \) identity matrices in the corresponding space, \( t \) is the tunneling between top and bottom surfaces and \( \hat{z} \) is a unit vector in the direction perpendicular to the surfaces. Note that the first term on the right-hand-side of Eq. (1) describes the Dirac fermions on two surfaces of a topological insulator while the second term is the hybridization of two surfaces due to the tunneling of electrons in ultra-thin films. This tunneling is responsible for the gap opening in the dispersion of surface states. The minimal effective model as given by Eq. (1) captures the main low energy features of a TITF and at the same time is simple enough to make the analytical treatment of dynamical responses and collective modes feasible. Diagonalizing the effective \( 4 \times 4 \) Hamiltonian (1), one obtains the dispersions of valance and conduction bands

\[ \varepsilon_{k\lambda} = \lambda \sqrt{\hbar^2 v_F^2 k^2 + t^2}, \tag{2} \]

where \( \lambda = -1 (+1) \) refers to two spin-degenerate valance (conduction) bands. The corresponding normalized eigenstates in the \( \psi = (\psi_1^+, \psi_1^-, \psi_2^+, \psi_2^-)^T \) basis, where two indices specify the layer and spin orientation of electrons, read [30]

\[ \psi_{k\lambda}^{(1)} = \frac{1}{\sqrt{2}} \begin{pmatrix} \lambda, -i \cos \alpha_k e^{i\theta_k}, \sin \alpha_k, 0 \end{pmatrix}^T, \]
\[ \psi_{k\lambda}^{(2)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 0, \sin \alpha_k, -i \cos \alpha_k e^{-i\theta_k}, \lambda \end{pmatrix}^T, \tag{3} \]

with \( T \) referring to the transpose of a vector, and \( \alpha_k = \tan^{-1}(t/\hbar v_F k) \) and \( \theta_k = \tan^{-1}(k_y/k_x) \), are defined for notational convenience.

The density fluctuations \( \delta \rho_l(q, \omega) \) induced in the top \((l = 1)\) or bottom \((l = 2)\) surface, in the linear response regime is given by [35]

\[ \delta \rho_l(q, \omega) = \sum_{l'} \chi_{ll'}(q, \omega)V_{ll'}^{\text{ext}}(q, \omega), \tag{4} \]

where \( \chi_{ll'}(q, \omega) \) is the surface-resolved linear density-density response function and \( V_{ll'}^{\text{ext}}(q, \omega) \) is the external potential applied to surface \( l' \). Within the random phase approximation the interacting linear density-density response function could be written in the matrix form as

\[ \chi_{RPA}^{ll'}(q, \omega) = [1 - \Pi(q, \omega)V(q)]^{-1} \Pi(q, \omega), \tag{5} \]

where

\[ \Pi(q, \omega) = \begin{pmatrix} \Pi_{11}(q, \omega) & \Pi_{12}(q, \omega) \\ \Pi_{21}(q, \omega) & \Pi_{22}(q, \omega) \end{pmatrix}, \tag{6} \]

and

\[ V(q) = \begin{pmatrix} V_{11}(q) & V_{12}(q) \\ V_{21}(q) & V_{22}(q) \end{pmatrix}, \tag{7} \]

are the matrices of non-interacting density-density response function and Coulomb interaction [25], respectively. If we assume that a thin slab of topological insulator with thickness \( d \) and the dielectric constant of \( \varepsilon_{TI} \) is embedded in an environment with the dielectric constant \( \varepsilon_{en} \) (see, Fig. 1), the intra-surface and inter-surface components of the Coulomb interaction matrix are respectively given by [26]

\[ V_{11}(q) = V_{22}(q) = \frac{2(\epsilon + 1)e^{qd} + (\epsilon - 1)e^{-qd}}{D(q)} v_q, \tag{8} \]

and

\[ V_{12}(q) = V_{21}(q) = \frac{4 \epsilon}{D(q)} v_q. \tag{9} \]

Here, \( v_q = 2\pi e^2/\varepsilon_{en} q \), \( \epsilon = \varepsilon_{TI}/\varepsilon_{en} \), and \( D(q) = (\epsilon + 1)^2 e^{qd} - (\epsilon - 1)^2 e^{-qd} \). Note that in the uniform i.e., \( \epsilon = 1 \) limit, we recover the familiar expressions \( V_{11}(q) = v_q \), and \( V_{12}(q) = v_q e^{-qd} \), for the intra-surface and inter-surface interactions, respectively.

FIG. 1. Schematic picture of an ultra-thin film of topological insulator with thickness \( d \) and dielectric constant \( \varepsilon_{TI} \), surrounded by a medium with the dielectric constant \( \varepsilon_{en} \).
The components of the surface-resolved non-interacting polarization functions are given by

$$
\Pi_l'(q, \omega) = \frac{1}{S} \sum_{k,\lambda,\lambda'} F_{l\lambda\lambda'}(k, k') \left[ f(\varepsilon_{k\lambda}) - f(\varepsilon_{k'\lambda'}) \right],
$$

where $S$ is the surface area, $k' \equiv k + q$, $f(\varepsilon)$ is the Fermi distribution function, and the form-factors are

$$
F_{l\lambda\lambda'}^{(l)}(k, k') = \sum_{i,j} \langle \Psi_{k\lambda}^{(i)} | \rho_l^{(i)} | \Psi_{k'\lambda'}^{(j)} \rangle \langle \Psi_{k'\lambda'}^{(j)} | \rho_l^{(i)} | \Psi_{k\lambda}^{(i)} \rangle,
$$

with the surface-resolved density operators of TITF defined as $\rho_l^{(i)} = [\tau^0 - (-1)^l \tau^z] \otimes \sigma^0/2$. It is straightforward to show that

$$
F_{l\lambda\lambda'}^{(l)}(k, k') = \frac{1}{2} \left( 1 + \lambda \varepsilon_{k\lambda} \frac{h^2 v_F^2 \mathbf{k} \cdot \mathbf{k}'}{\varepsilon_{k'\lambda'}} \right),
$$

and

$$
F_{l\lambda\lambda'}^{(l)}(k, k') = \frac{1}{2} \lambda \varepsilon_{k\lambda} \frac{h^2 v_F^2 \mathbf{k} \cdot \mathbf{k}'}{\varepsilon_{k'\lambda'}}.
$$

Here, $\varepsilon_k \equiv |\varepsilon_{k\lambda}|$ is introduced for notational convenience. We should note that the total density response function

$$
\Pi(q, \omega) = \sum_{l,l'} \Pi_{l'}(q, \omega)
$$

$$=
\frac{1}{S} \sum_{k,\lambda,\lambda'} \left[ f(\varepsilon_{k\lambda}) - f(\varepsilon_{k'\lambda'}) \right]
\frac{h\omega + \varepsilon_{k\lambda} - \varepsilon_{k'\lambda'} + \imath 0^+}{h\omega + \varepsilon_{k\lambda} - \varepsilon_{k'\lambda'} + \imath 0^+}
\left( 1 + \lambda \varepsilon_{k\lambda} \frac{h^2 v_F^2 \mathbf{k} \cdot \mathbf{k}'}{\varepsilon_{k'\lambda'}} \right),
$$

apart from a trivial degeneracy factor, is identical to the dynamical density response function of two-dimensional massive Dirac fermions [3, 9, 36]. It is possible to find analytic expressions for the real and imaginary parts of the layer-resolved density-density response functions for arbitrary frequency and wave-vector (c.f. Appendix A). As the total density response function $\Pi(q, \omega)$ is analytically known [3], we have simply presented the analytic expressions for the inter-surface component of the density response function $\Pi_{12}(q, \omega)$ in Appendix A. The intra-surface component of the density response function could be readily obtained from $\Pi_{11}(q, \omega) = \Pi(q, \omega)/2 - \Pi_{12}(q, \omega)$. The behavior of dynamical density response function versus frequency at two representative wave-vectors is illustrated in Fig. 2 in both insulating and metallic regimes.

A. The static limit

The static density-density response functions are obtained from the $\omega \to 0$ limit of the dynamical responses.

In the electron doped metallic regime i.e., $\mu > t$, where
\( \mu \) is the chemical potential of the system, we find
\[
\Pi_{11}(q) = -\frac{\mu}{2\pi \hbar^2 v_F^2} \left\{ 1 - \Theta(q - 2k_F) \right\} \times \left[ \frac{\sqrt{q^2 - 4k_F^2}}{2q} - \frac{\hbar v_F q}{4\mu} \arctan \left( \frac{\hbar v_F \sqrt{q^2 - 4k_F^2}}{2\mu} \right) \right],
\]
and
\[
\Pi_{12}(q) = \frac{t^2 \Theta(q - 2k_F)}{2\pi \hbar^2 v_F^3 q} \arctan \left( \frac{\hbar v_F \sqrt{q^2 - 4k_F^2}}{2\mu} \right),
\]
where \( k_F = \sqrt{\mu^2 - t^2/\hbar v_F} \) is the Fermi wave vector in the conduction band and \( \Theta(x) \) is the Heaviside step function.

For the inter-band transitions contribute to the dynamical density-density response of the static non-interacting density-density correlation. The behavior of the inter-surface and intra-surface components of the static non-interacting dynamical density-density response function vanishes in Eq. (19) and (20) with the tunneling \( t \), system behaves at zero temperature like an insulator and we get
\[
\Pi_{11}(q) = -\frac{t}{4\pi \hbar^2 v_F^2} - \frac{q}{16\pi \hbar v_F} \arccos \left( \frac{4t^2 - \hbar^2 v_F^2 q^2}{4t^2 + \hbar^2 v_F^2 q^2} \right),
\]
and
\[
\Pi_{12}(q) = \frac{t^2}{4\pi \hbar^2 v_F^3 q} \arccos \left( \frac{4t^2 - \hbar^2 v_F^2 q^2}{4t^2 + \hbar^2 v_F^2 q^2} \right),
\]
which are clearly independent of the chemical potential. The behavior of the inter-surface and intra-surface components of the static non-interacting density-density response function of a TITF in the static regime is illustrated in Fig. 3.

**III. COLLECTIVE MODES**

Collective density oscillations could be obtained from the poles of the interacting polarization function (5), which is equivalent to the solutions of the following equation
\[
[1 - V_+(q)\Pi_+(q, \omega)] [1 - V_-(q)\Pi_-(q, \omega)] = 0,
\]
where \( V_\pm(q) = V_{11}(q) \pm V_{12}(q) \) and \( \Pi_{\pm}(q, \omega) = \Pi_{11}(q, \omega) \pm \Pi_{12}(q, \omega) \). The solutions of Eq. (21) results in two collective density mode branches \( \omega_+(q) \) and \( \omega_-(q) \), corresponding respectively to the symmetric and asymmetric oscillations of density in two surfaces. Un-damped collective modes occur in the regions of \((q, \omega)\)-plane where the imaginary parts of the density response functions are zero, i.e., outside the electron-hole continuum (EHC). The analytic expressions for the non-interacting density response functions \( \Pi_{\ell}\Pi(q, \omega) \) are provided in Appendix A. The imaginary parts of the density response functions are non-zero only in regions 3A, 4A, 1B and 5B [see, Eq. (A5) and Fig. 8 for the definitions]. Note that at zero temperature no collective density oscillation could be excited in the insulating regime (i.e., \(|\mu| < t\)). Therefore, in the following, we discuss the dispersions of collective modes in the metallic regime.

**A. Analytic results in the long-wavelength limit**

We begin with the presentation of our analytic results for the dispersions of collective modes in the long wavelength, i.e., \( q \rightarrow 0 \) limit. For the symmetric mode in the long wavelength limit, upon substituting the vanishing wave vector behavior of \( \Pi_+(q \rightarrow 0, \omega) \) from Eq. (A8) into the first square bracket on the left-hand-side of Eq. (21), we obtain
\[
\frac{\hbar \omega_+(q \rightarrow 0)}{\mu} \approx \sqrt{1 - t^2} \sqrt{\frac{\alpha_{ee} \hbar v_F q}{\mu}} + \mathcal{O}(q^{3/2}),
\]
which has the expected \( \sqrt{q} \) dependance of the plasmon dispersion in two dimensions [35]. Here, \( t = t/\mu < 1 \) is the dimensionless tunneling parameter, and \( \alpha_{ee} = e^2/(\epsilon\epsilon_r \hbar v_F) \) is the dimensionless coupling constant of massless Dirac electrons. This expression is valid for small wave vectors i.e., \( qd \ll 2/\epsilon \), and it is interesting to note that the long-wavelength behavior of the symmetric mode is not sensitive to the dielectric constant of the topological insulator. For intermediate values of \( q \), it is possible to find the leading order quantum correction to the mode dispersion [21, 27]
\[
\frac{\hbar \omega_+(q \rightarrow 0)}{\mu} \approx \sqrt{1 - t^2} \sqrt{\frac{\alpha_{ee} \hbar v_F q}{\mu}} (1 + \epsilon q d/2)^{-1},
\]
which is valid for \( qd \ll 1 \) and the dependance on \( \epsilon_{TI} \) enters through \( \epsilon \). Note that the dispersion of optical mode
from Eq. (23) shows an unphysical plateau-like future at large wave vectors (c.f., Fig. 5).

To obtain the acoustic mode in the long wavelength limit, we substitute the vanishing $q$ limit of $\Pi_{-}(q, \omega)$ from Eq. (A9), in the second square bracket on the right-hand-side of Eq. (21) to find

$$\omega_{-}(q \to 0) \simeq v_{s}q = \frac{\sqrt{1 - t^{2}}}{g(\gamma, \bar{t})} v_{F}q,$$

(24)

with

$$g(\gamma, \bar{t}) = \sqrt{1 - \left( \frac{1 - t^{2}}{1 + \sqrt{1 - t^{2}/\gamma}} \right)^{2}},$$

(25)

and $\gamma = \alpha_{ee} k_{F} d / \epsilon$. As expected, the out-of-phase oscillation of electrons in two surfaces is influenced by the dielectric constant of the TITF through $\gamma$. Note that the border between 4A and 2A regions (c.f., Fig. 8) at the long wavelength limit is given by $\omega = \sqrt{1 - t^{2}} v_{F}q$. As $g(\gamma, \bar{t})$ is always smaller than one, we have $v_{s} > \sqrt{1 - t^{2}} v_{F}$, and the acoustic mode is always undamped at long wavelengths. For $v_{s} > v_{F}$ this mode lies in region 5B, whereas for $\sqrt{1 - t^{2}} v_{F} < v_{s} < v_{F}$ it resides in region 4A. As we have illustrated in Fig. 4, depending on different system parameters, the velocity of acoustic mode $v_{s}$ can be larger or substantially smaller than the Fermi velocity of isolated surface states $v_{F}$.

In the absence of inter-surface hybridization i.e., $t = 0$, we have $\mu = \hbar v_{F} k_{F}$, and Eqs. (22), (23) and (24) all reduce to the standard results obtained for double layer graphene and topological insulator thin films in the absence of tunneling [26, 27].

B. Numerical results on the dispersions of collective modes

The dispersions of the collective modes for arbitrary wave vectors could be obtained from the numerical solution of Eq. (21). The system parameters we have used in our numerical calculations are $t \approx 126$ meV and $v_{F} \approx 4.71 \times 10^{5}$ ms$^{-1}$, which results in $\hbar v_{F} \approx 3.1$ eV A and $\alpha_{ee} \approx 4.65$. These values correspond to an ultra-thin film of Bi$_2$Se$_3$ with the thickness of two quintuple layers $d \approx 20\AA$, whose electronic structure has been explored experimentally by means of the angel resolved photoemission spectroscopy [15]. Unless otherwise stated, we are going to use $\epsilon_{TI} = 30$ for the dielectric constant of the Bi$_2$Se$_3$ thin film, which is smaller than its bulk value (i.e., $\epsilon_{TI} \approx 100$) [37], and we also take $\epsilon_{en} = 1$ for the environment.

In Fig. 5 the dispersion of in-phase or symmetric collective mode $\omega_{+}(q)$ has been illustrated for two different values of the chemical potential. The full numerical results are compared with the analytical expressions in Eqs. (22) and (23). It is evident that the finite tunneling reduces the energy of symmetric mode.

The full dispersion of asymmetric (acoustic) mode has been plotted in Fig. 6. Here, for better visibility, we have used a much smaller value for the dielectric constant of the topological insulator thin film (i.e., $\epsilon_{TI} = 2$) as the mode dispersion for $\epsilon_{TI} = 30$ lies too close to the boundary of EHC. Again, it is evident that finite tunneling lowers the energy of collective mode. Notice that in the presence of a finite tunneling between two surfaces, undamped collective mode can also propagate in region 4A, where $\omega_{\pm}(q) < v_{F}q$.

A better insight into the dispersions of collective modes and their Landau damping inside the EHC could be gained from the imaginary parts of the inverse dielectric functions

$$\Im \left[ \frac{1}{\epsilon_{\pm}^{RPA}(q, \omega)} \right] = \Im \left[ \frac{1}{1 - V_{\pm}(q)\Pi_{\pm}^{0}(q, \omega)} \right],$$

(26)

which represent the spectral weights of the collective modes. The imaginary parts of inverse dielectric functions have Dirac delta form at $\omega = \omega_{\pm}(q)$ outside the EHC where the imaginary parts of the non-interacting responses are zero and acquire finite width inside the continuum due to the Landau damping of the collective modes [35].

Imaginary parts of the symmetric and asymmetric components of the inverse dielectric functions are illustrated in Fig. 7. The dispersions of collective modes as well as their broadening due to Landau damping inside the EHC are easily noticeable.

IV. SUMMARY

We have obtained analytic expressions for the surface resolved dynamical linear density-density response func-
t/µ = 0.5
- t/µ = 0

\( t/\mu = 0.5 \)
- \( t/\mu = 0 \)

\( t/\mu = 0.7 \)
- \( t/\mu = 0 \)

\( t/\mu = 0.5 \)
- \( t/\mu = 0 \)

\( t/\mu = 0.7 \)
- \( t/\mu = 0 \)

FIG. 5. Full dispersion for the in-phase plasmon mode (solid red) of a topological insulator ultra-thin film is compared to its analytic long wavelength solutions obtained from Eqs. (22) (dotted black) and (23) (dash-dotted black). The chemical potential \( \mu = 252 \) meV (top) and \( \mu = 180 \) meV (bottom), and \( t = 126 \) meV has been used here. The dispersions in the absence of tunneling (dashed black) are also plotted for comparison. Different regions in the \((q, \omega)\)-plane are as introduced in Appendix A.

FIG. 6. Full dispersion for the acoustic collective mode (solid red) of a topological insulator ultra-thin film is compared to its analytic long wavelength solution Eq. (24) (dotted black). The chemical potential \( \mu = 252 \) meV (top) and \( \mu = 180 \) meV (bottom) has been used here together with \( t = 126 \) meV and \( \epsilon_{TI} = 2 \). The results in the absence of tunneling (dashed black) are also plotted for comparison. Different regions in the \((q, \omega)\)-plane are as introduced in Appendix A.

tions of a topological insulator ultra-thin film with finite tunneling between its surfaces. The full dispersions of the collective density modes and their analytic form at the long-wavelength limit are also investigated. The velocity of acoustic mode and the Drude weight of the optical mode is tunable through different system parameters. The energy of both symmetric and asymmetric collective modes are suppressed due to the inter-surface tunneling. This means that for a given energy, the collective mode is shifted to large wave-vectors, making them more confined in space. Moreover, the tunneling induced finite gap in the spectrum of surface states makes it possible to switch on or off the plasmon modes through the gate voltage which tunes the chemical potential. Similar behavior has been observed in tunnel coupled double-layer graphene [38]. These futures are expected to be useful for practical implementation in future plasmonic circuitry.

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FIG. 7. Top: density plot of the symmetric component of the inverse dielectric function $\Pi(q, \omega)$ Eq. (10). As the total density-density response function of TITF is identical for the real and imaginary parts of the inter-surface density-density response function, respectively. Here, we have defined according to the arguments of the Dirac delta-functions in the literature [3, 9], at the zero temperature we find

$$\Re \Pi_{12}(q, \omega) = f(q, \omega) x_1^2$$

$$\Im \Pi_{12}(q, \omega) = f(q, \omega) x_1^2$$

for the real and imaginary parts of the inter-surface density-density response function, respectively. Here, we have defined $x_0 = \sqrt{1 + 4t^2/(h^2 v_F q^2 - h^2 \omega^2)}$, $x_1 = 2t/(h v_F q)$, and

$$f(q, \omega) = \frac{q^2}{16\pi \sqrt{|h^2 v_F q^2 - h^2 \omega^2|}}$$

Different regions in the $(q, \omega)$-plane are introduced according to the arguments of the Dirac delta-functions in

**Appendix A: Analytic results for the non-interacting density-density response function of TITF**

In this appendix we calculate the surface resolved non-interacting density-density response functions of a topological insulator ultra-thin film as defined through Eq. (10). As the total density-density response function $\Pi(q, \omega) = \sum_{\mu, \nu} \Pi_{\mu \nu}(q, \omega)$ of a TITF is identical to the density-density response function of 2D massive Dirac fermions, whose full analytic expressions are available in the literature [3, 9], here we will simply present the analytic expressions for the inter-surface density-density response function $\Pi_{12}(q, \omega)$. The intra-surface components could be readily obtained using $\Pi_{11}(q, \omega) = \Pi(q, \omega)/2 - \Pi_{12}(q, \omega)$. From Eqs. (10) and (13) we find

$$\Pi_{12}(q, \omega) = \frac{1}{2S} \sum_{k, \lambda, \lambda'} \left( \lambda' \epsilon_{k}^2 \right) \frac{f(\epsilon_{k} \epsilon_{k'}) - f(\epsilon_{k'} \epsilon_{k})}{\epsilon_{k} + \epsilon_{k} - \epsilon_{k'} - \epsilon_{k'} + i0^+},$$

where $k' = k + q$. Assuming $\mu > t$ and $\omega > 0$, and following similar procedures as Refs. [3, 9], at the zero temperature we find

$$\Re \Pi_{12}(q, \omega) = f(q, \omega) x_1^2$$

$$\Im \Pi_{12}(q, \omega) = f(q, \omega) x_1^2$$

for the real and imaginary parts of the inter-surface density-density response function, respectively. Here, we have defined $x_0 = \sqrt{1 + 4t^2/(h^2 v_F q^2 - h^2 \omega^2)}$, $x_1 = 2t/(h v_F q)$, and

$$f(q, \omega) = \frac{q^2}{16\pi \sqrt{|h^2 v_F q^2 - h^2 \omega^2|}}$$

Different regions in the $(q, \omega)$-plane are introduced according to the arguments of the Dirac delta-functions in

$$\Re \Pi_{12}(q, \omega) = f(q, \omega) x_1^2$$

$$\Im \Pi_{12}(q, \omega) = f(q, \omega) x_1^2$$

for the real and imaginary parts of the inter-surface density-density response function, respectively. Here, we have defined $x_0 = \sqrt{1 + 4t^2/(h^2 v_F q^2 - h^2 \omega^2)}$, $x_1 = 2t/(h v_F q)$, and

$$f(q, \omega) = \frac{q^2}{16\pi \sqrt{|h^2 v_F q^2 - h^2 \omega^2|}}$$

Different regions in the $(q, \omega)$-plane are introduced according to the arguments of the Dirac delta-functions in
the imaginary part of the density-density response function [3, 9]

\[
\begin{align*}
1A & : \bar{\hbar} \omega < - \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} \\
2A & : \pm \mu \mp \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} < \bar{\hbar} \omega < - \mu + \sqrt{\hbar^2 v_F^2 (q+k_F)^2 + t^2} \\
3A & : - \mu - \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} < \bar{\hbar} \omega < - \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} \\
4A & : \bar{\hbar} \omega < \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} \\
1B & : q < 2k_F, \sqrt{\hbar^2 v_F^2 q^2 + 4t^2} < \bar{\hbar} \omega < \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} \\
2B & : \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} < \bar{\hbar} \omega < \mu + \sqrt{\hbar^2 v_F^2 (q+k_F)^2 + t^2} \\
3B & : \bar{\hbar} \omega > \mu + \sqrt{\hbar^2 v_F^2 (q+k_F)^2 + t^2} \\
4B & : q > 2k_F, \sqrt{\hbar^2 v_F^2 q^2 + 4t^2} < \bar{\hbar} \omega < \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2} \\
5B & : \bar{\hbar} \omega < \mu + \sqrt{\hbar^2 v_F^2 (q-k_F)^2 + t^2}
\end{align*}
\]

(A5)

and are also illustrated in Fig. 8.

1. Results in $|\mu| < t$ limit

When the chemical potential lies inside the band gap, only the inter-band transitions contribute to the density-density response function, and we find

\[
\Re \Pi_1^0(q, \omega) = \frac{t^2}{4\pi \hbar^3 v_F^2} \left( \frac{v_F^2 q^2 - \omega^2}{v_F^2 q^2 - \omega^2/\hbar^2} \right) \times \left\{ \Theta(v_F q - \omega) \arccos \left( \frac{\omega^2 - 4t^2/\hbar^2}{\omega^2 - v_F^2 q^2} \right) \right\},
\]

and

\[
\Im \Pi_1^0(q, \omega) = \frac{t^2}{4\pi \hbar^3 v_F^2} \sqrt{\omega^2 - v_F^2 q^2 - 4t^2},
\]

respectively for the real and imaginary parts of the inter-surface density-density response functions.

2. The long-wavelength behaviors

To obtain the long-wavelength behavior of the collective modes, we investigate the vanishing $q$ limit of the symmetric and asymmetric components of the density-density response functions, respectively in the dynamical and acoustic limits [35], from the full analytic results we have just presented for the $\mu > t$ regime.

The long-wavelength behavior of the symmetric (i.e., optical) plasmon mode is obtained from the long-wavelength behavior of $\Pi_+ (q, \omega)$. For $\mu > t$, taking the $q \to 0$ and $\omega \to 0$ limits such that $\omega^2/q = c = \text{constant}$, one finds

\[
\Pi_+ (q, \omega) \approx \frac{\mu^2 - t^2 q}{4\pi \hbar^3 \mu c}.
\]

(A8)

To obtain the long wavelength behavior of the asymmetric mode, the long wavelength behavior of $\Pi_- (q, \omega)$ in the acoustic (i.e., $\omega/q = \gamma = \text{constant}$) limit is required

\[
\Pi_- (q \to 0, y) \approx \frac{\mu}{2\pi \hbar^2 v_F} \left[ \frac{y(1 - \bar{t}^2)}{\sqrt{y^2 - v_F^2 (1 - \bar{t}^2)}} - 1 \right],
\]

(A9)

where $\bar{t} = t/\mu$. Substituting Eqs. (A8) and (A9) into Eq. (21), we obtain Eqs. (22) and (24) respectively for the long wavelength dispersions of the symmetric and asymmetric collective modes.

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