Quantization Scheme of Surface Plasmon Polaritons in Two-Dimensional Helical Liquids *

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The collective modes of two-dimensional helical electron gases interacting with light have been studied in an extended random phase approximation. An inverse operator transformation that interprets electron oscillations and photons with quasi particles is developed. Because photons are initially included in the model, one can directly derive and compare the static and radiation (or vector) fields for the excited collective modes. Unlike the traditional quantization scheme that the electron oscillation's contribution is totally hidden in the dielectric function, we can directly investigate their roles when the collective modes interact with other particles. As an example, we find an additional term which plays an important role at small distance arising from electron exchanging effect when the collective modes couple to emitters.

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Recently, the quantum nature of surface plasmon poloritons (SPPs),[1−13] i.e., the coupled electromagnetic waves (or photons) and electron collective oscillations (or plasmons), has attracted great interests. It is well known that the electric fields of SPPs are quite localized and strongly enhanced,[14] so it can provide an excellent platform for investigating light matter interaction.[6] With the remarkable progresses in theories and device fabrications, researchers now can study and analyze the light matter interaction in the extreme spatial confinement,[15,16] where the quantum effect becomes significant, such as the quantum tunneling[8,13,17] and size effects.[7] Also, it can be used to study two-boson interference,[2] and it has been shown that SPPs can conserve the energy-time entanglement of a pair of photons,[18] and can be used in integrated logic devices.[19,20] Due to the strong coupling between SPPs and quantum emitters, such hybrid systems can be used as single photon sources.[21]

Due to the novel properties of Dirac electrons,[22] graphene and topological insulator (TI) become very hot topics in condensed matter physics. Most recently, plasmons and SPPs are proposed and confirmed to exist in graphene,[23−25] which provides highly tunable plasmonic metamaterials,[26] and in TI surface[27] which can be regarded as coupled electron density oscillations and spin density oscillations.[28] Researchers also find direct evidence for three-dimensional (3D) Dirac plasmon in the type-Ⅱ Dirac semimetal.[29] It has been revealed that there are rich physics in the interdisciplinary area of helical liquids and SPPs.

Generally, SPPs are regarded as quasi particles which hybrid photons and electron collective oscillations. The quantization scheme of SPPs plays a fundamental role when dealing with the interaction between them and other particles or quasi-particles,[12] also it is important to the plasmon mediated interactions.[30] When the momentum is not quite small, one can ignore photons and the quantization can be achieved by directly writing down the plasmon operator as the summation of electron density oscillating operators, then the wave function and energy are solved within random phase approximation (RPA).[28,31,32] In such a method, the interaction between electrons is the static Coulomb interaction, so the retardation effect[33] is totally ignored. However, the another scheme developed from quantizing the electromagnetic wave,[1,4,34] which is widely applied, needs to solve Maxwell's equations with the materials' dielectric functions. This means that, in this method, the roles of electrons in SPPs are hidden behind the dielectric functions. To clarify all components' roles in SPPs, we extend the first method[32,28] to include both photons and electrons.

Quantization Scheme of SPPs.—The system we have considered can be described by Fig. 1(a), where an emitter (2D exciton or molecule) is closely placed on top of a 2D electron gas (2DEG). Note that we have chosen the surface state of topological insulator (TI),[22] and it can be straightforwardly extended to other systems, such as graphene, single layer MoS_2, or quantum wells formed by semiconductor films. In the 2DEG, SPPs can generate, propagate and interact with the emitter through electromagnetic field[5,11] or through electron exchanging effect.

Firstly, let us ignore the emitter and calculate

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SPPs of the 2DEG. The radiation field can be expanded as $\frac{1}{c}\hat{\boldsymbol{A}}(\boldsymbol{x},t)=\frac{1}{\sqrt{\nu}}\sum_{\lambda\boldsymbol{p}}e^{i\boldsymbol{p}\cdot\boldsymbol{x}}\hat{\boldsymbol{A}}_{\lambda\boldsymbol{p}}(t)$ (Coulomb gauge $\nabla\cdot\hat{\boldsymbol{A}}=0$ is used throughout this article), here $\nu=SL_z$ is the space volume, and $\hat{\boldsymbol{A}}_{\lambda\boldsymbol{p}}(t)=\sqrt{2\pi\hbar/\omega_{\boldsymbol{p}}}[\boldsymbol{\xi}_\lambda(\boldsymbol{p})\hat{a}_{\lambda\boldsymbol{p}}(t)+\boldsymbol{\xi}_\lambda(-\boldsymbol{p})\hat{a}^\dagger_{\lambda,-\boldsymbol{p}}(t)]$, $\hat{a}_{\lambda\boldsymbol{p}}(t),\hat{a}^\dagger_{\lambda,-\boldsymbol{p}}(t)$ are photon annihilation and creation operators. Here $\hbar\omega_{\boldsymbol{p}}$ is the free photon energy, and $\boldsymbol{x}$ and $\boldsymbol{p}$ are the 3D position and momentum vector respectively. In this study, we use cylinder coordinates that $\boldsymbol{x}=(\boldsymbol{r},z)$ and $\boldsymbol{p}=(\boldsymbol{q},p_z)$. The photon index $\lambda=1,2$ correspond to two photon modes with the polarization vectors $\boldsymbol{\xi}_{\lambda\boldsymbol{p}}$ perpendicular to the wave vector $\boldsymbol{p}$, which have been set to $\boldsymbol{\xi}_1(\boldsymbol{p})=(-\sin\theta_{\boldsymbol{p}},\cos\theta_{\boldsymbol{p}},0)$ and $\boldsymbol{\xi}_2(\boldsymbol{p})=(-\cos\phi_{\boldsymbol{p}}\cos\theta_{\boldsymbol{p}},-\cos\phi_{\boldsymbol{p}}\sin\theta_{\boldsymbol{p}},\sin\phi_{\boldsymbol{p}})$, where $\theta_{\boldsymbol{p}}$ and $\phi_{\boldsymbol{p}}$ are angles of $\boldsymbol{p}$ in polar coordinates (please see Fig. 1(b)).

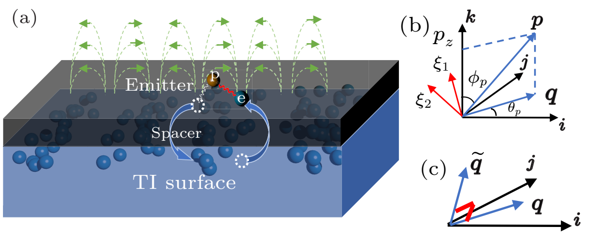

Fig. 1. (a) A molecular emitter (e means electron and p means positively charged core) is placed on top of a topological insulator with a spacer layer. When SPPs are excited in the TI surface, they can coupled to each other by the electromagnetic field and through exchanging electrons. (b) The coordinate system used in this paper. (c) The illustration of $\tilde{\boldsymbol{q}}$ which is perpendicular to $\boldsymbol{q}$ with $\theta_{\tilde{\boldsymbol{q}}}=\theta_{\boldsymbol{q}}+\pi/2$.

Generally, the energies of SPPs we've considered are quite small compared to the band gap of 2DEG. So, an effective low energy Hamiltonian, $\hat{H}_s(\hat{\boldsymbol{p}})=\hat{\boldsymbol{p}}^2/2m^*+\hbar v_{\rm f}(\hat{\sigma}_x\hat{p}_y-\hat{\sigma}_y\hat{p}_x)+h\sigma_z$, is sufficient to describing electrons for TI surface states. Note that the topological trivial term $\hat{\boldsymbol{p}}^2/2m^*$ is essential to provide a diamagnetic current, which overcomes the energy cutoff problem when dealing with the optical properties of TI surface states,[35] and also, we find that this term renormalizes the paramagnetic current-current response function to the physical one (for more details, please see the Supplementary Material).

Aware that the electron excitation energy in z direction is quite higher than the energy of plane excitations we've considered, one can simplify the real space operator $\hat{\varPsi}^\dagger(\boldsymbol{x},t)$ with $\psi(z)\hat{\varPsi}^\dagger(\boldsymbol{r},t)$. Here $\psi(z)$ is the envelope function ($\int|\psi(z)|^2dz=1$), and $\hat{\varPsi}(\boldsymbol{r},t)=(\hat{c}_\uparrow(\boldsymbol{r},t),\hat{c}_\downarrow(\boldsymbol{r},t))^{\rm T}$ is a Nambu spinor. In momentum space, the single electron Hamiltonian $\hat{H}_s$ can be diagonalized by transformation $\hat{\gamma}_{s\boldsymbol{k}}=\langle s\boldsymbol{k}|\hat{\varPsi}_{\boldsymbol{k}}$, where $s=\pm1$, $\langle s\boldsymbol{k}|=e^{-\frac{i}{2}s\theta_{\boldsymbol{k}}}(u_{{\rm s}k}e^{\frac{i}{2}\theta_{\boldsymbol{k}}},-isv_{{\rm s}k}e^{-\frac{i}{2}\theta_{\boldsymbol{k}}})$ and $\hat{\varPsi}_{\boldsymbol{k}}=(\hat{c}_{\boldsymbol{k}\uparrow},\hat{c}_{\boldsymbol{k}\downarrow})^{\rm T}$ with energy $\xi_{{\rm s}k}=\mu+sE_k$, $E_k=\sqrt{v_{\rm f}^2k^2+h^2}$, and wave functions $u_{{\rm s}k},v_{{\rm s}k}=\sqrt{\frac{1}{2}(1\pm sh/E_k)}$. For shorter notations, let us omit the subscript of $\boldsymbol{k}_\parallel,\boldsymbol{q}_\parallel$ and let $\boldsymbol{k},\boldsymbol{q}$ just be the in-plane vectors, omit time t in operators for the same reason. Finally, with some considerations in the Supplementary Material, the Hamiltonian in momentum space can be expressed as

$$\begin{aligned}\hat{\boldsymbol{H}}=&\sum_{s\boldsymbol{k}}\xi_{s\boldsymbol{k}}\hat{\gamma}^\dagger_{s\boldsymbol{k}}\hat{\gamma}_{s\boldsymbol{k}}+\sum_{\lambda\boldsymbol{q}}\hbar\omega_{\boldsymbol{q}}\hat{a}^\dagger_{\lambda\boldsymbol{q}}\hat{a}_{\lambda\boldsymbol{q}}-\frac{1}{\sqrt{\nu}}\sum_{\boldsymbol{q}}\hat{\boldsymbol{j}}_{\boldsymbol{q}}\cdot\hat{\boldsymbol{A}}_{-\boldsymbol{q}}\\&+\frac{1}{2S}\sum_{ss'll'\boldsymbol{k}\boldsymbol{k}'\boldsymbol{q}}V^{ss'll'}_{\boldsymbol{k}\boldsymbol{q}}\hat{\gamma}^\dagger_{s\boldsymbol{k}+\boldsymbol{q}}\hat{\gamma}^\dagger_{l\boldsymbol{k}'-\boldsymbol{q}}\hat{\gamma}_{l'\boldsymbol{k}'}\hat{\gamma}_{s'\boldsymbol{k}},\end{aligned}\tag{1}$$

where $V^{ss'll'}_{\boldsymbol{k}\boldsymbol{q}}=V_q\langle s\boldsymbol{k}+\boldsymbol{q}|s'\boldsymbol{k}\rangle\langle l\boldsymbol{k}'-\boldsymbol{q}|l'\boldsymbol{k}'\rangle$, and $V_q=2\pi e^2/q$ is the two-dimensional Coulomb potential. The current operator $\hat{\boldsymbol{j}}(\boldsymbol{q})=\hat{\boldsymbol{j}}^p(\boldsymbol{q})+\hat{\boldsymbol{j}}^d(\boldsymbol{q})$ contains two parts, the first one is the paramagnetic current $\hat{j}^p_x(\boldsymbol{q})=e\hbar v_{\rm f}\sum_{\boldsymbol{k}}\langle s\boldsymbol{k}+\boldsymbol{q}|\sigma_y|s'\boldsymbol{k}\rangle$ and $\hat{j}^p_y(\boldsymbol{q})=-e\hbar v_{\rm f}\sum_{\boldsymbol{k}}\langle s\boldsymbol{k}+\boldsymbol{q}|\sigma_x|s'\boldsymbol{k}\rangle$ (the subscripts means components in x and y directions, and one can easily find the contribution of the z component can be ignored). The diamagnetic current arising from the topological trivial term in $\hat{H}_s$ mentioned above reads $\hat{\boldsymbol{j}}^d_y(\boldsymbol{q})=-e^2/(2m^*c^2)\int\boldsymbol{d}\boldsymbol{r}dz|\psi(z)|^2\hat{n}(\boldsymbol{r},t)\hat{\boldsymbol{A}}(\boldsymbol{x},t)e^{-i\boldsymbol{q}\cdot\boldsymbol{r}-ip_zz}\approx\sqrt{S/L_z}\varPi^d/2\sum_{p_z}\hat{A}_{\boldsymbol{q},p_z+p'_z}$ (where $\varPi^d=\bar{n}e^2/m^*$).

Similar to Nambu spinor, we can define a basis $\hat{\varPhi}^\dagger_{\boldsymbol{q}}=[\cdots\hat{\rho}^\dagger_{\boldsymbol{k}_i\boldsymbol{q}s_is'_i}\cdots\hat{a}_{\lambda_a(-\boldsymbol{q},-p_{zi})}\cdots\hat{a}^\dagger_{\lambda(\boldsymbol{q},p_{zi})}\cdots]^{\rm T}$, where $s_is'_i=++,+-,-+,--$, $\lambda=1,2$ are electron bands and photon indexes. Finally, the collective mode can be written as $\hat{Q}^\dagger_{\boldsymbol{q}}=\varPhi_{\boldsymbol{q}}\hat{\varPhi}^\dagger_{\boldsymbol{q}}$, where $\varPhi_{\boldsymbol{q}}$ is the wave function. By comparing the coefficients of different operators in the quasi particle dynamic equation $i\hbar\partial_t\hat{Q}^\dagger_{\boldsymbol{q}}=[\hat{Q}^\dagger_{\boldsymbol{q}},\hat{H}]=-\hbar\varOmega_{\boldsymbol{q}}\hat{Q}^\dagger_{\boldsymbol{q}}$, one can get the quasi particle energies ($\hbar\varOmega_{\boldsymbol{q}}$) and wave functions ($\varPhi_{\boldsymbol{q}}$). After some algebras, we find that the quasi particle energies satisfy the equation

$$V^l_{\boldsymbol{q}}V^t_{\boldsymbol{q}}|\varPi_{xy}|^2=(1-V^l_{\boldsymbol{q}}\varPi_{xx})(1-V^t_{\boldsymbol{q}}\varPi_{yy}),\tag{2}$$

where $V^l_{\boldsymbol{q}}$ and $V^t_{\boldsymbol{q}}$ are the longitudinal and transverse photon propagators, $\varPi_{xx}$ and $\varPi_{yy}$ are the diagonal current-current response functions, $\varPi_{xy}$ is the off-diagonal current-current response function.[35,36] We want to emphasize here that we can not only derive the mode energy $\hbar\varOmega_{\boldsymbol{q}}$ (Eq. (2) is just the same as the one derived by traditional method), but also the wave function $\varPhi_{\boldsymbol{q}}$. As an example, we have calculated the decoupled case when the static Zeeman field $h=0$ (in Fig. 2, we plot the wave functions $|\varPhi^{\rm a,c}_{1p_z}|^2$, the superscripts a and c means annihilation and creation, and $|\varPhi^l_{++\boldsymbol{k}\boldsymbol{q}}|^2\Delta n^{++}_{\boldsymbol{k}\boldsymbol{q}}$ in the inset figure for $q=0.1k_{\rm f}$, the superscript means longitudinal). In this case, Eq. (2) decouples to two modes, the longitudinal and transverse modes. Note that the magnitudes for photon creation and annihilation have a small difference, and only the electron density oscillations near Fermi surface contribute to SPPs.

The wave functions and energies for all quasi particles described by our extended RPA (not only the collective excitations) can be proved to satisfy an eigenvalue equation $\bar{H}\Phi_{n\boldsymbol{q}} = \hbar\Omega_{n\boldsymbol{q}}\Phi_{n\boldsymbol{q}}$. In principle, one can always calculate and find all the solutions, then get $(\hat{Q}^\dagger_{\boldsymbol{q}}, \hat{Q}_{-\boldsymbol{q}}, \hat{Q}^\dagger_{1\boldsymbol{q}}, \hat{Q}_{1,-\boldsymbol{q}}, \hat{Q}^\dagger_{2\boldsymbol{q}}, \hat{Q}_{2,-\boldsymbol{q}}, \cdot, \cdot, \cdot)^{\rm T} = U\hat{\Phi}^\dagger_{\boldsymbol{q}}$. Here $U = [\cdots \Phi^{\rm T}_{n\boldsymbol{q}}, \Phi^{\rm T}_{-n\boldsymbol{q}} \cdots]^{\rm T}$, the annihilator operators appear because $\bar{H}_{\boldsymbol{q}}$ has particle-hole symmetry that $R^\dagger \bar{H}_{\boldsymbol{q}} R = -\bar{H}^*_{-\boldsymbol{q}}$, $\hat{Q}_{-n,-\boldsymbol{q}} = \hat{Q}^\dagger_{n,\boldsymbol{q}}$, where $-n$ indicates negative energy. Note that $\bar{H}$ is not Hermitian, instead, it satisfies $\bar{H}^\dagger J = J\bar{H}$ with $J = {\rm diag}[\Delta n^{s_1 s_1'}_{\boldsymbol{k}_1\boldsymbol{q}_1}, \Delta n^{s_2 s_2'}_{\boldsymbol{k}_2\boldsymbol{q}_2} \cdots, 1, 1, \cdots, -1, -1, \cdots]$, one can immediately find $\langle \Phi_{n\boldsymbol{q}}|J|\Phi_{m\boldsymbol{q}}\rangle = 0, \pm 1$ ($+1$ for $n = m$ and positive energy, -1 for $n = m$ and negative energy, 0 for $n \neq m$ or $\Delta n^{ss'}_{\boldsymbol{k}\boldsymbol{q}} = 0$) and get the inverse transformation

$$U^{-1} = \begin{pmatrix} I_{2M\times 2M} & 0 \\ -J_{\rm nz}U^\dagger_{\rm nz}\sigma_3\otimes I_N U_z & -J_{\rm nz}U^\dagger_{\rm nz}\sigma_3\otimes I_N \end{pmatrix}, \tag{3}$$

where $J_{\rm nz}$ is the nonzero sub-matrix of J, and $U_{\rm nz}$ is the corresponding sub-matrix of U,

$$U = \begin{pmatrix} I_{2M\times 2M} & 0 \\ U_z & U_{\rm nz} \end{pmatrix}.$$

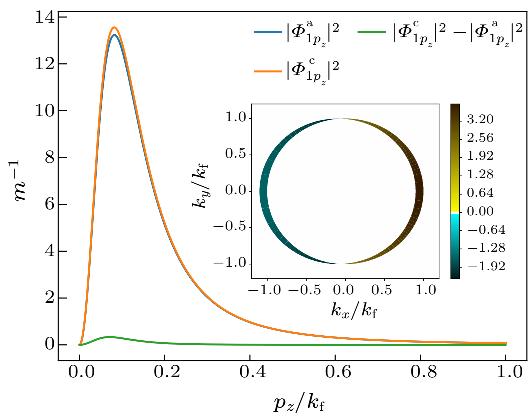

Fig. 2. $|\Phi^{\rm a,c}_{1p_z}|^2$ and $|\Phi^l_{++\boldsymbol{k}\boldsymbol{q}}|^2\Delta n^{++}_{\boldsymbol{k}\boldsymbol{q}}$ (inset figure) as functions of p_z and $\boldsymbol{k}$ at $q = 0.1k_{\rm f}$. The chemical potential for electron gas is 0.1 eV and the Fermi velocity of TI $v_{\rm F} = 5\times 10^5$ m/s.

Emitter SPP coupling.–Once getting U^{-1}, we can calculate the interaction between quasi particles with other systems. As an example, suppose we have an emitter (a hydrogen-like molecule) localized at $z = z_0, \boldsymbol{r} = 0$ (Fig. 1(a)). The plane wave functions $e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$ and the discrete localized states $\phi_i(\boldsymbol{x})$ of the emitter form a complete set, so we can expand $\hat{\Psi}(\boldsymbol{x}) = \hat{\Psi}_{\rm 2D}(\boldsymbol{x}) + \hat{\Psi}_{\rm et}(\boldsymbol{x}) = \frac{1}{\sqrt{S}}\sum_{\boldsymbol{k}} e^{i\boldsymbol{k}\cdot\boldsymbol{r}}\psi(z)\,\Psi^\dagger_{\boldsymbol{k}} + \sum_i \phi_i(\boldsymbol{x})\hat{\phi}^\dagger_i$, where $\hat{\phi}_{i^\dagger}$ is the ith creation operator of emitter. Note that the emitter's wave function is well localized, one reasonable assumption is that its total electron number is conserved when interacting with SPPs. So, we only keep the emitter electron conserved terms in the total Hamiltonian which has three parts, i.e., $\hat{H} = \hat{H}_{\rm et} + \hat{H}_{\rm ee} + \hat{H}_{\rm 2D}$, where the last term $\hat{H}_{\rm 2D}$ is the Hamiltonian of 2DEG interacting with light defined above, $\hat{H}_{\rm et}$ is the emitter Hamiltonian under vector field $\hat{\boldsymbol{A}}(\boldsymbol{x})$ and emitter potential $V_{\rm et}(\boldsymbol{x})$. The second term $\hat{H}_{\rm ee}$ is the electron-electron interaction Hamiltonian, which can be expressed as the emitter electron interacting with fields generated by the 2DEG. Now

$$\hat{H}_{\rm et} = \int \hat{\Psi}^\dagger_{\rm et}(\boldsymbol{x})\Big[\frac{1}{2m^*}(\hat{p} + \frac{e}{c}\hat{A}(\boldsymbol{x}))^2 + V_{\rm et}(\boldsymbol{x})\Big]\hat{\Psi}(\boldsymbol{x})d\boldsymbol{x},$$

$$\hat{H}_{\rm ee} = \int\int \hat{\Psi}^\dagger_{\rm et}(\boldsymbol{x})\hat{V}_{\rm eff}(\boldsymbol{x},\boldsymbol{x}')\hat{\Psi}_{\rm et}(\boldsymbol{x}')d\boldsymbol{x}d\boldsymbol{x}'. \tag{4}$$

The effective potential $\hat{V}_{\rm eff}(\boldsymbol{x},\boldsymbol{x}')$ contains two parts, one originates from the direct Coulomb interaction $\delta(\boldsymbol{x}-\boldsymbol{x}')\hat{V}_{\rm ec}(\boldsymbol{x})$, and the other comes from electron exchanging effect $\hat{V}_{\rm ex}(\boldsymbol{x},\boldsymbol{x}')$, where

$$\hat{V}_{\rm ec}(\boldsymbol{x}) = \int V(|\boldsymbol{x}-\boldsymbol{x}''|)\,\hat{\Psi}^\dagger_{\rm 2D}(\boldsymbol{x}'')\,\hat{\Psi}_{\rm 2D}(\boldsymbol{x}'')d\boldsymbol{x}'',$$

$$\hat{V}_{\rm ex}(\boldsymbol{x},\boldsymbol{x}') = -V(|\boldsymbol{x}-\boldsymbol{x}'|)\,\hat{\Psi}^\dagger_{\rm 2D}(\boldsymbol{x}')\,\hat{\Psi}_{\rm 2D}(\boldsymbol{x}). \tag{5}$$

It should be emphasized here that we want to derive the interaction between emitter and SPPs, but not the interaction between emitter with the total 2DEG. To do so, we can utilize the inverse transformation and only keep the contribution from SPPs in $\hat{V}_{\rm eff}(\boldsymbol{x},\boldsymbol{x}')$. For the direct Coulomb potential, we have $\hat{V}^{\rm spp}_{\rm ec}(\boldsymbol{x}) = \sum e^{-i\boldsymbol{q}\cdot\boldsymbol{r}-q|z|}L^{\rho *}_{\boldsymbol{q}}\hat{Q}^\dagger_{\boldsymbol{q}} + {\rm H.c.}$, where $L^\rho_{\boldsymbol{q}}$ is the normalization factor of wave function $\Phi_{\boldsymbol{q}}$, the superscript spp means only considering the contribution of SPPs.

It is convenient to study the interaction through electric field in dipole approximation, one can find that the total electric field reads

$$\begin{aligned}\hat{E}(\boldsymbol{x}) &= \hat{E}^{\rm s}(\boldsymbol{x}) + \hat{E}^{\rm r}(\boldsymbol{x}) \\ &= -\frac{1}{e}\sum_{\boldsymbol{q}} e^{-i\boldsymbol{q}\cdot\boldsymbol{r}-q'z}[(i\boldsymbol{q}', q)L^{\rho *}_{\boldsymbol{q}} + \Omega_{\boldsymbol{q}}\frac{\tilde{\boldsymbol{q}}}{q}L^{y*}_{\boldsymbol{q}}]\hat{Q}^\dagger_{\boldsymbol{q}} + {\rm H.c.}\end{aligned} \tag{6}$$

It contains two parts, the superscript s means the contribution from $\hat{V}^{\rm spp}_{\rm ec}$, and r means the contribution from vector field. Here $\tilde{\boldsymbol{q}}$ is an in-plane vector perpendicular to $\boldsymbol{q}$ ($\theta_{\tilde{\boldsymbol{q}}} = \theta_{\boldsymbol{q}} + \pi/2$, illustrated in Fig. 1(c)), and $\boldsymbol{q}'$ means the magnitude of $\boldsymbol{q}$ reduced from q to $q' = \sqrt{q^2 - \Omega^2_{\boldsymbol{q}}/c^2}$, and $L^y_{\boldsymbol{q}}$ is another normalization factor existing in transverse or longitudinal transverse hybrid modes and $L^y_{\boldsymbol{q}} = (1 - V_{\rm ql}\Pi_{xx})/(V_q\Pi_{\rho y})L^\rho_{\boldsymbol{q}}$.

In Fig. 3, we plot the relative ratio of radiation to static electric field (the parallel constituent, and because their vertical parts have the same magnitude but opposite sign, we do not put it in the figure) as a function of q at $z = 0$ for the pure longitudinal mode. It is clear that when $q/k_{\rm f}$ is less than 4×10^{-5} (for a typical chemical potential $\mu = 100$ meV with Fermi velocity $v_{\rm F} = 5\times 10^5$ m/s, the critical value of q is $\approx$100 cm^{-1}), the radiation component is large enough and cannot be ignored. It should be noted that the energy of SPP is about 1 meV at the critical point, which means the radiation part is important for TI when considering physical processes with energy of meV.

Now, the interaction Hamiltonian through electric field $\hat{H}^{f}_{\rm int} = -\int \hat{\mu}(\boldsymbol{x}) \cdot \hat{\boldsymbol{E}}(\boldsymbol{x}) d\boldsymbol{x}$ reads

$$\begin{aligned}\hat{H}^{f}_{\rm int} =& e \int \hat{\Psi}^{\dagger}_{\rm et}(\boldsymbol{x})[\boldsymbol{x} \cdot \hat{\boldsymbol{E}}(\boldsymbol{x})]\hat{\Psi}_{\rm et}(\boldsymbol{x}) d\boldsymbol{x} \\ &\approx \sum_{ij\boldsymbol{q}} g^{ij}_{\rm f}(\boldsymbol{q}) \hat{\phi}^{\dagger}_{i} \hat{\phi}_{j} \hat{Q}^{\dagger}_{\boldsymbol{q}} + \text{H.c.},\end{aligned} \tag{7}$$

with the interaction strength

$$g^{ij}_{\rm f}(\boldsymbol{q}) = -i(\boldsymbol{q}' \cdot \boldsymbol{d}^{||}_{ij} + q d^{\perp}_{ij}) L^{\rho *}_{\boldsymbol{q}} - \frac{\Omega_{\boldsymbol{q}}}{q} \tilde{\boldsymbol{q}} \cdot \boldsymbol{d}^{||}_{ij} L^{y *}_{\boldsymbol{q}}, \tag{8}$$

where $\boldsymbol{d}_{ij} = \int \phi^{*}_{i}(\boldsymbol{x}) \boldsymbol{x} \phi_{j}(\boldsymbol{x}) d\boldsymbol{x}$ is the transition dipole moment, the superscripts $||$ and $\perp$ mean the parallel and vertical parts, respectively.

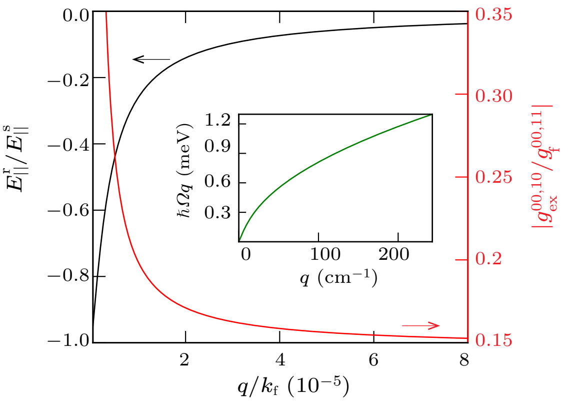

Fig. 3. The relative ratios $E^{\rm r}_{||}/E^{\rm s}_{||}$ at $z = 0$ (black line) and $g^{\rm ex}_{00,10}/g_{f00,11}$ (red line). Here the superscripts r and s mean radiation and static respectively, the subscript $_{||}$ means the in-plane component. The insert figure is the energy of the pure longitudinal mode for $\mu = 100$ meV.

When the emitter and 2DEG get close to each other and their wave functions overlap in z direction (e.g. the distance is smaller than 1 nm), $\hat{V}_{\rm ex}(\boldsymbol{x}, \boldsymbol{x}')$ arising from the electron exchanging becomes important, after some similar calculations, we can find that the exchange Hamiltonian $\hat{H}^{\rm ex}_{\rm int}$ has the same form of Eq. (7) with the interaction strength

$$g^{ij}_{\rm ex}(\boldsymbol{q}) = \frac{q \lambda_{\rm f} L^{\rho *}_{\boldsymbol{q}}}{8\pi^3} \eta^{ij}_{\boldsymbol{q}}, \tag{9}$$

where $\lambda_{\rm f}$ is the Fermi wavelength of 2DEG, and $\eta^{ij}_{\boldsymbol{q}}$ is defined in the Supplementary Material. The exchange strength or $\eta^{ij}_{\boldsymbol{q}}$ is quite complicated and should be numerically calculated, but for a simple situation that a longitudinal SPP interacts with a 2D hydrogen-like molecule, it has analytical results.

Note that the field interaction strength $g^{ij}_{\rm f}(\boldsymbol{q})$ now equals $-i\boldsymbol{q}' \cdot \boldsymbol{d}_{ij} L^{\rho *}_{\boldsymbol{q}}$, and we can compare it with the exchange strength by calculating the ratio $|g^{ij}_{\rm ex}(\boldsymbol{q})/g^{ij}_{\rm f}(\boldsymbol{q})| = \eta_{ij}(\lambda_{\rm f}/a_0)/(8\pi^3) q \lambda_{\rm f}/(\boldsymbol{q}' \cdot \boldsymbol{d}_{ij})$. When q is not very small, $q' \approx q$ and these two interaction strengths are comparable. When $q \to 0$, because $q/q' \to \infty$, we can conclude that the exchange effect plays the most important role at small q. To provide a perspicuous and direct impression, we plot $|g^{00,10}_{\rm ex}(\boldsymbol{q})/g^{00,11}_{\rm f}(\boldsymbol{q})|$ for the transitions from the ground state (00) to the first (10) and second (11) excited states in Fig. 3 (red line). Because the two strengths have different selection rules (for example, $g^{00,10}_{\rm ex}(\boldsymbol{q})$ is none zero, while the transition dipole moment $\boldsymbol{d}_{00,10} = 0$), and $g^{00,11}_{\rm ex}(\boldsymbol{q})$ is quite complicated, we have calculated $|g^{00,10}_{\rm ex}(\boldsymbol{q})/g^{00,11}_{\rm f}(\boldsymbol{q})|$ instead of $|g^{00,11}_{\rm ex}(\boldsymbol{q})/g^{00,11}_{\rm f}(\boldsymbol{q})|$. It is clear that it diverges at $q = 0$, and $\approx$15% at large q. Also, it shares the same critical value $q_{\rm c}$ and the same energy scale meV with $E^{\rm r_{||}}/E^{\rm s_{||}}$.

In conclusion, we have developed a quantization scheme for the collective excitations of 2DEGs when considering photon fields. The quasi particle energy and wave function have been derived, and we have calculated the inverse operator transformation which interprets photons and electron density oscillations with quasi particles. With the help of such transformation, the interaction between SPPs and molecule emitters are studied. We find that when they are closely placed, a new type of interaction originating from exchanging electron emerges, which has different selection rules compared to the traditional interaction and has comparable strength at large $\boldsymbol{q}$, and plays the most important role at small momentum ($\boldsymbol{q} \lesssim 100\,{\rm cm}^{-1}$). Finally, we emphasize here that this method can also be applied to other type emitters such as excitons and quantum dots, and other 2DEGs beyond TI surface states, or just investigate the interaction between SPPs from different layers. Also, one can utilize it to quantize other excitations under RPA.

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