Lifetime of surface plasmons

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We calculate the lifetime of a surface plasmon on the surface of jellium metal from the process of the surface plasmon decaying into electron-hole pairs in the metal. We find that this process only occurs over a finite interval of wave vector, and at large values of wave vector. Our calculation contained two new features: (i) we used the Brillouin formula to normalize the electromagnetic field traveling along the surface and (ii) we included the spin rotation term in the electron current. The latter contribution has no effect at large wave vector of the surface plasmon.

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I. INTRODUCTION

Surface plasmons are the fundamental electronic excitation on metal surfaces [1]. Their dispersion has been measured for many common metals [2–9]. Correspondingly, theoretical calculations on many metals predict quite well the measured dispersions [10–34]. An excellent review by Plummer's group is in Ref. [35].

There has been some work [2,22,32,33,36] on the lifetime of surface plasmons. Fuchs and Kliewer [33] used the Lindhard dielectric function to evaluate the plasmon lifetime. It is the dielectric function for Coulomb interactions in the interior of the metal, but is not appropriate for the surface nor for photonic interactions. It ignores the electromagnetic component of the surface plasmon. Khurgin [32] also used the Lindhard function. A recent calculation by Gao *et al.* [36] used a semiclassical model, whereas our calculation is fully quantum mechanical. Many of the older calculations used the Feibelman d parameters [19], which is usually a small wave vector approximation. However, Liebsch [22] used the local-density approximation to calculate the real and imaginary part of the $d(\omega)$ function to high frequency: the latter is the lifetime of the surface plasmon. Our theory is valid for large wave vectors. None of these calculations included the new features of this calculation, which are using the Brillouin formula to quantize the surface plasmons, and including the spin rotation in the electron current. Our results for the surface plasmon lifetime are shown in Fig. 1. We have not found a similar figure in any previous publication. Nor have we found any prior work that found the dielectric function for the electromagnetic interactions of the surface plasmons.

Here we calculate the lifetime of surface plasmons on a jellium metal planar surface due to the plasmons exciting electron-hole pairs in the metal. Our calculation contains new features absent in earlier work: (i) the quantization of the electromagnetic modes on the surface uses the Brillouin formula [37–39] and (ii) we include the spin rotation terms in the current of the electrons [40]. Much of the previous theory was concerned with the dispersion of surface plasmons, and most theories only applied at small wave vectors. Our theory applies at small and at large wave vectors.

Plasmonics is now a huge field of research [41–46]. Our exact results should be useful for this field.

If the surface is in the (x,y) plane, then the two-dimensional wave vector of the surface is $\mathbf{q} = (q_x,q_y,0)$. A related wave vector is $\mathbf{Q} = (q_x,q_y,q_z)$. The standard formula for the surface mode is [1]

$$q^2c^2 = \omega^2\frac{\varepsilon(\omega)}{\varepsilon(\omega)+1}. \tag{1}$$

In the past, this equation was solved by ignoring the wave vector dependence and using $\varepsilon(0,\omega)$ for the dielectric function. The reason is that, although the bulk dielectric function $\varepsilon(Q,\omega)$ is known, the surface dielectric function $\varepsilon(q,\omega)$ was not known, although there have been numerous guesses. Here we derive the surface dielectric function $\varepsilon(q,\omega)$. We use it to calculate the lifetime of the surface plasmon.

We derive the transverse dielectric function for a metal surface. It is the response to a photon field. The dielectric function is a 3×3 tensor. The longitudinal component is $\hat{q}\cdot\varepsilon\cdot\hat{q}$. Earlier we derived [47] the longitudinal dielectric function of the surface. Here we derive the transverse components of the surface dielectric tensor $\hat{\xi}\cdot\varepsilon\cdot\hat{\xi}$, where $\hat{\xi}$ is the photon polarization vector.

II. THEORY

We solve for the wave vector dependence of surface plasmons on a jellium metal surface. The usual theory of surface plasmons ignores the wave vector dependence of the dielectric function: $\varepsilon(q,\omega)$ is approximated as $\varepsilon(\omega)$. Then the dispersion of surface plasmons is given by the formulas [1]

$$0 = p + \frac{\gamma}{\varepsilon}, \tag{2}$$

$$p^2 = q^2 - \frac{\omega^2}{c^2}, \tag{3}$$

$$\gamma^2 = q^2 - \varepsilon(\omega)\frac{\omega^2}{c^2}, \tag{4}$$

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2}, \quad \omega_p^2 = \frac{n_e e^2}{\varepsilon_0 m}, \tag{5}$$

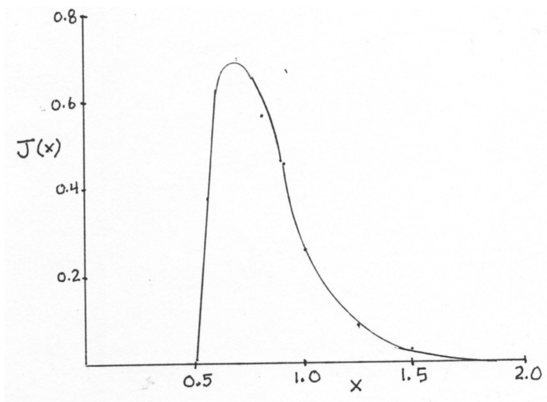

FIG. 1. The function $J(x)$ vs. x.

where $\mathbf{q}$ is the two-dimensional wave vector of the surface plasmon, and ω_p is the plasma frequency in terms of the electron density n_e and mass m. The dispersion relation can be manipulated to give

$$c^2q^2 = \omega^2\frac{\varepsilon}{\varepsilon+1}. \tag{6}$$

Solving this equation gives the surface plasmon dispersion as [1]

$$\omega_q^2 = (cq)^2 + \omega_{sp}^2 - \sqrt{(cq)^4 + \omega_{sp}^4}, \tag{7}$$

$$\omega_{sp} = \frac{\omega_p}{\sqrt{2}}. \tag{8}$$

The interactions of photons with jellium has two terms:

$$V_1 = \frac{e^2}{2m}\int d^3r\rho(\mathbf{r})A^2(\mathbf{r}), \tag{9}$$

$$V_2 = -\int d^3r\vec{j}(\mathbf{r})\cdot\vec{A}(\mathbf{r}), \tag{10}$$

where $\rho(\mathbf{r})$ is the electron charge density in the metal, $\vec{j}(\mathbf{r})$ is the electron current operator in the metal, and $\vec{A}(\mathbf{r})$ is the vector potential of the photons. The interaction V_1 has already been included in deriving the dielectric function in Eq. (5). The interaction V_2 contributes to the wave vector dependence of the surface plasmon.

III. QUANTIZATION OF THE VECTOR POTENTIAL

The first step in the derivation is to quantize the vector potential. This has been discussed in earlier publications [37–39,48], so here we summarize the result. We use the electromagnetic units called "rationalized MKSA" in Jackson [39]. The surface plasmon is a transverse magnetic mode with wave vector $\mathbf{q} = (q_x,q_y,0)$ along the surface in the (x,y) plane. We introduce an unknown function $B_0(q)$ along with some raising and lowering operators $a_\mathbf{q}^\dagger,a_\mathbf{q}$ for the surface polariton. The magnetic field generated by the surface plasmon is

$$\vec{B}(\mathbf{r},t) = \sum_\mathbf{q} B_0(q_y,-q_x,0)(\xi a_\mathbf{q} + \xi^* a_\mathbf{q}^\dagger)\phi_B(z), \tag{11}$$

$$\phi_B(z) = e^{-pz},\quad z>0, \tag{12}$$

$$= e^{\gamma z},\quad z<0, \tag{13}$$

$$\xi = \exp[i(\mathbf{q}\cdot\rho - \omega_q t)]. \tag{14}$$

Note that $\vec{\nabla}\cdot\vec{B} = 0$. From Maxwell's equation we can derive the electric field and vector potential:

$$E_{x,y}(\mathbf{r},t) = -ic^2\sum_\mathbf{q} B_0\frac{q_{x,y}p}{\omega_q}(\xi a_\mathbf{q} - \xi^* a_\mathbf{q}^\dagger)\phi_B(z), \tag{15}$$

$$E_z(\mathbf{r},t) = c^2\sum_\mathbf{q} B_0\frac{q^2}{\omega_q}(\xi a_\mathbf{q} + \xi^* a_\mathbf{q}^\dagger)\phi_z(z), \tag{16}$$

$$A_{x,y}(\mathbf{r},t) = c^2\sum_\mathbf{q} B_0\frac{q_{x,y}p}{\omega_q^2}(\xi a_\mathbf{q} + \xi^* a_\mathbf{q}^\dagger)\phi_B(z), \tag{17}$$

$$A_z(\mathbf{r},t) = ic^2\sum_\mathbf{q} B_0\frac{q^2}{\omega_q^2}(\xi a_\mathbf{q} - \xi^* a_\mathbf{q}^\dagger)\phi_z(z), \tag{18}$$

$$\phi_z(z) = e^{-pz},\quad z>0, \tag{19}$$

$$= \frac{1}{\varepsilon}e^{\gamma z},\quad z<0. \tag{20}$$

The quantization of the electromagnetic field proceeds from Brillouin's formula [37–39]:

$$\sum_\mathbf{q}\hbar\omega_q(a_\mathbf{q}^\dagger a_\mathbf{q} + 1/2) = \frac{\varepsilon_0}{2}\int d^3r[g(\omega)E^2 + c^2B^2], \tag{21}$$

$$g(\omega) = \frac{\partial}{\partial\omega}[\omega\varepsilon(\omega)] = 1 + \frac{\omega_p^2}{\omega^2}. \tag{22}$$

After some algebra, one finds

$$B_0^2 = \frac{2\gamma\hbar\omega_q}{\varepsilon_0 A(qc)^2}J(q) = \left(\frac{2\hbar}{\varepsilon_0 Ac}\right)\frac{\omega_q}{pc}J(q),$$
$$J = \frac{\omega_q^4(\omega_p^2-\omega_q^2)^2}{\omega_q^2(\omega_p^2-2\omega_q^2)(\omega_p^2-\omega_q^2)^2 + (cq)^2[\omega_q^4(\omega_p^2+\omega_q^2) + (\omega_p^2-\omega_q^2)^2(\omega_p^2+3\omega_q^2)]}, \tag{23}$$

which provides the function $B_0(q)$ needed for proper quantization. It is used to evaluate the interaction V_2. Here A is the area of the surface plane. The dimensional units of B_0 are Volts $\cdot$ sec/meter, while $J(q)$ is dimensionless.

IV. $p \cdot A$ INTERACTION

We confine the electrons in the metal slab $-L < z < 0$ so that their wave function is

$$\phi(\mathbf{r}) = \sqrt{\frac{2}{LA}} e^{i\mathbf{k}\cdot\rho} \sin(k_i z)\chi, \quad k_i = \frac{n\pi}{L}, \tag{24}$$

$$s_{i,f} = \sin(k_{i,f} z), \quad c_{i,f} = \cos(k_{i,f} z), \quad \mathbf{k} = (k_x,k_y,0), \tag{25}$$

where χ is the spin part. The derivation of the electrical current, and the interaction V_2, is detailed in the Appendix. We include in the current the spin rotation terms in the current for fermions, as described by Sakurai [40]:

$$V_2 = -\sum_{\mathbf{k},\mathbf{q},k_i,k_f} C^\dagger_{\mathbf{k}+\mathbf{q},k_f} C_{\mathbf{k},k_i}(a_\mathbf{q} - a^\dagger_{-\mathbf{q}}) M_T, \tag{26}$$

$$M_T = \frac{e\hbar(cq)^2 B_0}{2mL\omega_q^2} V_- \left\{ (\chi_j^\dagger \chi_i)\left[\mathbf{q}\cdot(2\mathbf{k}+\mathbf{q}) + \frac{1}{\varepsilon} k_+ k_-\right] - \gamma[\chi_f^\dagger(q_y\sigma_x - q_x\sigma_y)\chi_i]\left(1+\frac{1}{\varepsilon}\right)\right\}, \tag{27}$$

$$V_- = \left[\frac{1}{\gamma^2 + k_-^2} - \frac{1}{\gamma^2 + k_+^2}\right], \tag{28}$$

$$k_\pm = k_i \pm k_f. \tag{29}$$

This interaction is evaluated in the second order of perturbation theory. After squaring the matrix element M_T, we must average over spin components (s,s') in the initial and final states:

$$\frac{1}{2}\sum_{ss'} |M_T|^2 = \left(\frac{e\hbar B_0}{2mL}\right)^2 \left(\frac{cq}{\omega_q}\right)^4 V_-^2 \left\{\left[\mathbf{q}\cdot(2\mathbf{k}+\mathbf{q}) + \frac{k_- k_+}{\varepsilon}\right]^2 + \gamma^2 q^2\left(1+\frac{1}{\varepsilon}\right)^2\right\}.$$

The last term is from the spin rotation parts of the current operator. From Eq. (1), we have the identity

$$\left(1+\frac{1}{\varepsilon}\right)^2 = \left(\frac{\omega_q}{cq}\right)^4. \tag{30}$$

The correction to the photon Green's function is

$$\mathcal{D}_{\mu\nu}(\mathbf{q},\omega) = \frac{2\omega_q}{\omega^2 - \omega_q^2 - \Pi}, \tag{31}$$

$$\Pi = \frac{\omega_q}{\hbar} \sum_{\mathbf{k},k_i,k_f,s,s'} |M_T|^2 \frac{n_F(\mathbf{k},k_i) - n_F(\mathbf{k}+\mathbf{q},k_f)}{E(\mathbf{k},k_i) - E(\mathbf{k}+\mathbf{q},k_f) - \hbar\omega}, \tag{32}$$

$$= C_0 I, \tag{33}$$

$$C_0 = \frac{2e^2\gamma}{\varepsilon_0 m}\left(\frac{cq}{\omega_q}\right)^2 J(q), \tag{34}$$

$$I = \int \frac{dk_i}{\pi} \int \frac{dk_f}{\pi} \int \frac{d^2k}{(2\pi)^2} n_F(\mathbf{k},k_i) V_-^2 T \times \left[\frac{1}{q\cdot(2\mathbf{k}+\mathbf{q}) + \xi_+} + \frac{1}{q\cdot(2\mathbf{k}+\mathbf{q}) + \xi_-}\right], \tag{35}$$

$$T = \left[\left[\mathbf{q}\cdot(2\mathbf{k}+\mathbf{q}) + \frac{k_- k_+}{\varepsilon}\right]^2 + q^2\gamma^2\left(1+\frac{1}{\varepsilon}\right)^2\right], \tag{36}$$

$$\xi_\pm = k_f^2 - k_i^2 \pm q_0^2, \quad q_0^2 = 2m\omega/\hbar, \tag{37}$$

where C_0 has the units of meter^2/sec^2.

V. LIFETIME

$\Pi(q,\omega)$ has an imaginary part, which is determined by

$$\Im\{\Pi\} = L(\omega) - L(-\omega), \tag{38}$$

$$L(\omega) = \pi \int \frac{dk_i}{\pi} \int \frac{dk_f}{\pi} \int \frac{d^2k}{(2\pi)^2} n_F(\mathbf{k},k_i) T V_-^2 \times \delta[q\cdot(2\mathbf{k}+\mathbf{q}) + \xi_+]. \tag{39}$$

The integral over $\int d^2k$ gives

$$\int d^2k\,\delta[q\cdot(2\mathbf{k}+\mathbf{q}) + \xi_+] = \frac{1}{2q^2}\sqrt{4q^2(k_F^2 - k_i^2) - (q^2 + \xi_+)^2}. \tag{40}$$

Denote $s = \sqrt{k_F^2 - k_i^2}$ and get for the right-hand-side

$$= \frac{1}{2q^2}\sqrt{(k_f^2 - k_l^2)(k_u^2 - k_f^2)}, \tag{41}$$

$$k_l^2 = k_F^2 - q_0^2 - (q+s)^2, \tag{42}$$

$$k_u^2 = k_F^2 - q_0^2 - (q-s)^2. \tag{43}$$

The integral is nonzero only when $k_u^2 > 0$, which constrains

$$2q\sqrt{k_F^2 - q_0^2} + q_0^2 - q^2 > k_i^2 > q_0^2 - q^2 - 2q\sqrt{k_F^2 - q_0^2}. \tag{44}$$

When $\omega = \omega_{sp}$, the ratio $(q_0/k_F)^2 = \hbar\omega_{sp}/E_F = 0.665\sqrt{r_s}$, where r_s is the density parameter of the electron gas [49]. So $q_0^2 > k_F^2$ for all metals with $r_s > 2.26$, which includes all but a dozen metals. In a metal with $r_s > 2.66$, when $\omega = \omega_{sp}$, then $k_u^2 < 0$ and the imaginary part of $\Pi(q,\omega)$ is zero. Next examine k_l^2. It is negative for many values of (q,ω,k_i). In those cases, the integral $\int dk_f$ has its lower limit of zero.

There are two different regions of wave vector q. At small values $q \sim \omega_{sp}/c$, the surface plasmon dispersion rises from zero to its full value $\omega_q \approx \omega_{sp}$. However, all of the experiments on surface plasmons are performed at much larger wave vectors, of the order $q \sim 1/\text{Å}$. We confine our calculation to this latter region, which simplifies the integrals. Then $\gamma \approx p \approx q$, $\omega_q = \omega_{sp}$, and $\varepsilon = -1$. Then $T = q_0^4 + \omega_{sp}^4/c^4$ and $J \to \omega_{sp}^2/8(qc)^2$. The second term in T is negligible. Collecting the remaining terms gives

$$L(\omega) = \frac{q_0^4}{q^2(2\pi)^3}\int_0^{k_F} dk_i \int dk_f \sqrt{(k_f^2 - k_l^2)(k_u^2 - k_f^2)}\, V_-^2.$$

Next consider the term $L(-\omega)$, change the sign of q_0^2. The constraint that $k_F^2 > q_0^2$ is now relaxed, and this $L(-\omega)$ is nonzero for all metals. Now the constraint $k_u^2 > 0$ is

$$2q\sqrt{k_F^2 + q_0^2} - q_0^2 - q^2 > k_i^2 > 0, \tag{45}$$

which requires a nonzero value of q to be satisfied:

$$k_F + \sqrt{k_F^2 + q_0^2} > q > \sqrt{k_F^2 + q_0^2} - k_F. \tag{46}$$

At zero frequency, the electron-hole excitations in a metal are constrained to be $0 < q < 2k_F$. However, for nonzero frequency, the minimum and maximum values of q increase, in this case to the above values. The integral for $L(-\omega)$ is evaluated numerically in dimensionless units: $x = q/k_F, y = k_i/k_F, z = k_f/k_F, W = q_0^2/k_F^2 = 0.665\sqrt{r_s}$.

$$\Im\{\Pi\} = G(q)\mathcal{J}(x), \tag{47}$$

$$G(q) = \frac{\omega_{sp}^2}{2\pi^2(k_F a_0)}, \tag{48}$$

$$\mathcal{J}(x) = \frac{1}{x}\int_0^1 dy \int_{z_l}^{z_u} dz \sqrt{\left(z_u^2 - z^2\right)\left(z^2 - z_l^2\right)} \times \left[\frac{1}{(y-z)^2 + x^2} - \frac{1}{(y+z)^2 + x^2}\right]^2, \tag{49}$$

$$z_{u,l}^2 = W + y^2 - x^2 \pm 2x\sqrt{1 - y^2}, \tag{50}$$

where a_0 is the Bohr radius.

(1) For $z_u > 0$, then

$$1 - (x - \sqrt{1+W})^2 > y^2 > 1 - (x + \sqrt{1+W})^2. \tag{51}$$

(2) For $z_l > 0$ then

$$y^2 > 1 - (x - \sqrt{1+W})^2. \tag{52}$$

(3) Therefore, $z_l^2 < 0$ where $z_u^2 > 0$, so that the integral $\int dz$ goes from $z_u > z > 0$.

Figure 1 shows an evaluation of the double integral for $\mathcal{J}(x)$ as a function of $x = q/k_F$. For numerical convenience, we chose $W = 1.25, \sqrt{1+W} = 1.5$, which is a metal with $r_s = 3.53$. Then the limits of the wave vector are $0.5 < x < 2.5$. The values of $\mathcal{J}(x)$ reach a peak right above the threshold, and then decline in value. It is negligible at the upper end $x = 2.5$. The decline at large values of q comes from the asymptotic behavior $V_-^2 \to q^{-8}$.

The imaginary part of $\Pi(\omega)$ has the units of frequency squared. At these large wave vectors, we can write the photon Green's function as

$$\mathcal{D}(q,\omega) = \frac{2\omega_{sp}}{\omega^2 - \omega_{sp}^2[1 + i\phi(q/k_F)]}, \tag{53}$$

$$\phi(x) = 0.093\mathcal{J}(x), \tag{54}$$

where the prefactor of $\phi(x)$ equals $1/[2\pi^2(k_F a_0)]$, where $1/(k_F a_0) = r_s/1.9192$ is evaluated at $r_s = 3.53$. What is the lifetime $\tau(q)$ of the surface plasmon? We answer this question by taking a Fourier transform of the Green's function from frequency to time, which results in an exponent

$$\exp[-i\omega t] \to \exp[-it\omega_{sp}\sqrt{1 + i\phi}], \tag{55}$$

$$\approx \exp\left[-it\omega_{sp}\left(1 + \frac{i\phi}{2}\right)\right], \tag{56}$$

$$\frac{1}{\tau} = \frac{\omega_{sp}\phi(x)}{2}. \tag{57}$$

At its peak value, $1/\tau$ is about 2% of the surface plasmon frequency.

VI. DISCUSSION

We have calculated the lifetime of a surface plasmon on the surface of jellium metal from the process of the surface plasmon decaying into electron-hole pairs in the metal. We find that this process only occurs over a finite interval of wave vector, and at large values of wave vector. It is interesting that the same feature is found in plasmon decay in three dimensions [49]. Our calculation contained two new features: (i) we used the Brillouin formula [37] to normalize the electromagnetic field traveling along the surface and (ii) we included the spin rotation term [40] in the electron current. The latter contribution has no effect at large wave vector of the surface plasmon. Our main result, obtained by a two-dimensional numerical integral, is contained in Fig. 1, which shows the dimensionless lifetime as a function of $x = q/k_F$, where k_F is the Fermi wave vector of the metal. We have not found a figure of this type in any prior publication.

APPENDIX

Here we derive the expression for the electron current operator $j_\mu(\mathbf{r},t)$ in the metal. We include the spin rotation terms derived in Sakurai [40]. The current for spin one-half Fermions is written in terms of derivatives and Pauli matrices $(\sigma_x,\sigma_y,\sigma_z)$:

$$j_x = \frac{e\hbar}{2mi}\left\{\chi_f^\dagger\left[\mathcal{I}\frac{\partial}{\partial x} - i\sigma_y\frac{\partial}{\partial z} + i\sigma_z\frac{\partial}{\partial y}\right]\chi_i - \left[\chi_i^\dagger\left[\mathcal{I}\frac{\partial}{\partial x} - i\sigma_y\frac{\partial}{\partial z} + i\sigma_z\frac{\partial}{\partial y}\right]\chi_f\right]^\dagger\right\}, \tag{A1}$$

$$j_y = \frac{e\hbar}{2mi}\left\{\chi_f^\dagger\left[\mathcal{I}\frac{\partial}{\partial y} - i\sigma_z\frac{\partial}{\partial x} + i\sigma_x\frac{\partial}{\partial z}\right]\chi_i - \left[\chi_i^\dagger\left[\mathcal{I}\frac{\partial}{\partial y} - i\sigma_z\frac{\partial}{\partial x} + i\sigma_x\frac{\partial}{\partial z}\right]\chi_f\right]^\dagger\right\}, \tag{A2}$$

$$j_z = \frac{e\hbar}{2mi}\left\{\chi_f^\dagger\left[\mathcal{I}\frac{\partial}{\partial z} - i\sigma_x\frac{\partial}{\partial y} + i\sigma_y\frac{\partial}{\partial x}\right]\chi_i - \left[\chi_i^\dagger\left[\mathcal{I}\frac{\partial}{\partial z} - i\sigma_x\frac{\partial}{\partial y} + i\sigma_y\frac{\partial}{\partial x}\right]\chi_f\right]^\dagger\right\}, \tag{A3}$$

where $\mathcal{I}$ is the 2×2 identity tensor, and the wave functions contain two-dimensional spinors (χ_i,χ_f). The electrons scatter from the initial state $(\mathbf{k},k_i)$ to a final state $(\mathbf{k}+\mathbf{q},k_f)$. The initial wave functions have the form $\rho = (x,y,0)$:

$$\phi_i(\mathbf{r}) = \sqrt{\frac{2}{AL}}e^{i\mathbf{k}\cdot\rho}\sin(k_i z)\chi_i, \tag{A4}$$

and a similar form for the final wave function: $\chi_{i,f}$ are the spin eigenfunctions. The current operator becomes

$$j_x = \frac{e\hbar}{mLA}e^{-i\rho\cdot\mathbf{q}}\{[(\chi_f^\dagger\chi_i)(2k_x + q_x)s_i s_f - i(\chi_f^\dagger\sigma_z\chi_i)q_y s_i s_f - (\chi_f^\dagger\sigma_y\chi_i)(k_i c_i s_f + k_f s_i c_f)\}, \tag{A5}$$

$$j_y = \frac{e\hbar}{mLA}e^{-i\rho\cdot\mathbf{q}}\{(\chi_f^\dagger\chi_i)(2k_y + q_y)s_i s_f + i(\chi_f^\dagger\sigma_z\chi_i)q_x s_i s_f + (\chi_f^\dagger\sigma_x\chi_i)(k_i c_i s_f + k_f s_i c_f)\}, \tag{A6}$$

$$j_z = \frac{e\hbar}{miLA} e^{-i\rho\cdot\mathbf{q}}\{(\chi_f^\dagger\chi_i)(k_ic_is_f - k_fs_ic_f) - (\chi_f^\dagger\sigma_x\chi_i)q_ys_is_f + (\chi_f^\dagger\sigma_y\chi_i)q_xs_is_f\}, \tag{A7}$$

$$s_{i,f} = \sin(k_{i,f}z), \quad c_{i,j} = \cos(k_{i,j}z). \tag{A8}$$

Multiply the above current by the vector potential of surface plasmons:

$$\vec{j}\cdot\vec{A} = \frac{e\hbar}{mLA}\sum_{\mathbf{k},\mathbf{q}} e^{-i\mathbf{q}\cdot\rho} C^\dagger_{\mathbf{k}+\mathbf{q},k_f} C_{\mathbf{k},k_i} \times \sum_{\mathbf{Q}'} \frac{B_0c^2}{\omega_{q'}^2} e^{\gamma' z}(M_x + M_y + M_z), \tag{A9}$$

$$M_x + M_y = (a_{\mathbf{q}'}\xi + a^\dagger_{\mathbf{q}'}\xi^*)p\{(\chi_f^\dagger\chi_i)\mathbf{q}\cdot(2\mathbf{k}+\mathbf{q})s_is_f + [\chi_f^\dagger(q_y\sigma_x - q_x\sigma_y)\chi_i](k_fc_fs_i + k_ic_is_f)\}, \tag{A10}$$

$$M_z = \frac{q^2}{\varepsilon}(a_{\mathbf{q}'}\xi - a^\dagger_{\mathbf{q}'}\xi^*)\{(\chi_f^\dagger\chi_i)(k_ic_is_f - k_fc_fs_i) - s_is_f[\chi_f^\dagger(q_y\sigma_x - q_x\sigma_y)\chi_i]\}. \tag{A11}$$

It is interesting that some of the spin-rotation terms have canceled out. Now we evaluate the integral $\int d^3r$ in V_2. The two-dimensional integral over (x,y) sets $\mathbf{q} = \pm\mathbf{q}'$ in the current and the vector potential. The z integrals give

$$\int_{-L}^0 dze^{\gamma z}s_is_f = \frac{\gamma}{2}(1 \pm e^{-\gamma L})V_-, \tag{A12}$$

$$\int_{-L}^0 dze^{\gamma z}(k_ic_is_f - k_fc_fs_i) = \frac{k_-k_+}{2}(1 \pm e^{-\gamma L})V_-, \tag{A13}$$

$$\int_{-L}^0 dze^{\gamma z}(k_ic_is_f + k_fc_fs_i) = -\frac{\gamma^2}{2}(1 \pm e^{-\gamma L})V_-, \tag{A14}$$

$$V_- = \left[\frac{1}{\gamma^2 + k_-^2} - \frac{1}{\gamma^2 + k_+^2}\right], \tag{A15}$$

$$k_\pm = k_i \pm k_f. \tag{A16}$$

The exponent $\exp(-\gamma L)$ also has a factor of $\exp(-iLk_\pm) = \pm 1$. We consider a thick slab, and calculate the dispersion at one surface: so set $\exp(-\gamma L) = 0$. Also note the identity $p\gamma = q^2$. The interaction is now

$$V_2 = -\sum_{\mathbf{k},\mathbf{q},k_i,k_f} C^\dagger_{\mathbf{k}+\mathbf{q},k_f} C_{\mathbf{k},k_i}(a_\mathbf{q} - a^\dagger_{-\mathbf{q}})M_T, \tag{A17}$$

$$M_T = \frac{e\hbar(qc)^2B_0}{2mL\omega_q^2}V_-\left\{(\chi_j^\dagger\chi_i)\left[\mathbf{q}\cdot(2\mathbf{k}+\mathbf{q}) + \frac{1}{\varepsilon}k_+k_-\right] + \gamma[\chi_f^\dagger(q_y\sigma_x - q_x\sigma_y)\chi_i]\left(1 + \frac{1}{\varepsilon}\right)\right\}. \tag{A18}$$

In calculating Π, we will average over spin arrangements in $\langle|M_T|^2\rangle$

$$\frac{1}{2}\sum_{ss'}\langle|M_T|^2\rangle = T\left(\frac{e\hbar(qc)^2B_0}{2mL\omega_q^2}V_-\right)^2, \tag{A19}$$

$$T = [\mathbf{q}\cdot(2\mathbf{k}+\mathbf{q}) + \frac{k_-k_+}{\varepsilon}]^2 + q^2\gamma^2\left(1 + \frac{1}{\varepsilon}\right)^2. \tag{A20}$$

The last term is due to spin rotation.

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