Quantization of Non-Radiative Surface Plasma Oscillations

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Non-radiative surface plasma oscillations in a semi-infinite metal are quantized by using a hydrodynamic jellium model for electrons in the metal. In consequence of the quantization, the electron-SP (surface plasmon) vertex function is obtained for the overall region of the surface plasmon wave vector $k$. In the electrostatic limit, it coincides with the usual electrostatic vertex function obtained by others before. In the extremely retarded limit ($k \to 0$), however, it has a different $k$ dependence from the one predicted by a simple extrapolation of the electrostatic vertex function to the long wavelength region. Our vertex function diverges as $k \to 0$, whereas the usual electrostatic one vanish at $k = 0$ if it is extrapolated to $k = 0$. By using our vertex function, the long wavelength behavior of the surface loss intensity of scattered electrons passing through the metal foil is also predicted in the case of the normal incidence.

§ 1. Introduction

Since the surface loss owing to the excitation of a surface plasmon (SP) was observed in a fast electron scattering experiment in thin metal film, many works on the surface plasmon, both theoretical and experimental, have been done extensively. In the early stage of these works, the interaction vertex function of an incident charged particle with a surface plasmon was derived from semi-classical considerations. Such attempts were made as to obtain these vertex function quantum mechanically, but yet within the electrostatic approximation. There has not so far been works which succeeded in quantizing the surface plasma oscillation with the retardation effect taken into account and giving an electron-SP vertex function valid even in the retarded region.

In this paper, the non-radiative surface plasma mode in a semi-infinite metal is quantized with the retardation effect taken into account. The motivation of this quantization is in obtaining various interaction vertex functions of the surface plasmon, for example, with an electron or with a photon, which are valid in an overall region of the wave number $k$ of the surface plasmon. As a result of the quantization, the electron-SP interaction vertex function is presented for an overall region of $k$.

In § 2, the quantization of surface plasma oscillations is carried out by using the hydrodynamic jellium model for electrons in the metal. In consequence of it, fluctuations of all the fields appearing in the model are expressed in terms of surface plasmon creation and annihilation operators. With these fluctuations of the fields, we can write electron-SP interaction Hamiltonian. In § 3, the electron-SP vertex function is obtained for the overall region of SP wave number $k$. It is shown that, in the electrostatic limit, it coincides with the usual electrostatic vertex function obtained first by Stern and Ferrell. In the long wavelength limit ($k \to 0$) where the retardation effect becomes important, however, the vertex function thus obtained shows a different $k$ dependence from the one predicted by a simple extrapolation of the electrostatic vertex function to the retarded region. Our vertex function diverges as $k^{-1/2}$ as $k$ goes to zero, in contrast to the fact that the usual electrostatic vertex function goes to zero as $k$ goes to zero if it is extended to $k = 0$ beyond the validity of the electrostatic approximation. In the final section, our
results are discussed in connection with the inelastic scattering of electrons passing through a metal foil.

§ 2. Quantization of the non-radiative surface plasmon

Our system is an electron gas in a metal with a plane surface. For simplicity, the metal is assumed to occupy a half-space $z < 0$ and to face the vacuum at the boundary surface $z = 0$. The hydrodynamic jellium model for the semi-infinite electron gas is also used. Our basic equations, therefore, are the full set of Maxwell's equations and the hydrodynamic Bloch equation $^{n}$ for the electron gas in the linearized forms:

\begin{align}
\nabla \cdot H &= 0, \quad (2.1) \\
\nabla \cdot E &= 4\pi ne, \quad (2.2) \\
\nabla \times E &= -\frac{1}{c} \frac{\partial H}{\partial t}, \quad (2.3) \\
\nabla \times H &= \frac{1}{c} \frac{\partial E}{\partial t} + \frac{4\pi ne}{c} v, \quad (2.4) \\
n_0 m \frac{\partial v}{\partial t} &= n_0 e E - m_0^2 \beta^2 \nabla n, \quad (2.5)
\end{align}

inside the metal and

\begin{align}
\nabla \cdot H &= 0, \quad (2.6) \\
\nabla \cdot E &= 0, \quad (2.7) \\
\nabla \times E &= -\frac{1}{c} \frac{\partial H}{\partial t}, \quad (2.8) \\
\nabla \times H &= \frac{1}{c} \frac{\partial E}{\partial t}, \quad (2.9)
\end{align}

in the vacuum.

In these equations, $E$, $H$, $n$ and $v$ are fluctuations of the electric field, magnetic field, electron density and average velocity of electrons in the metal respectively; $e$ and $m$ are the charge and mass of an electron; $n_0$ is the electron density in the undisturbed state of the electron gas, and $\beta^2 = \frac{3}{5} v_F^2$ with the Fermi velocity of the electron $v_F$.

By introducing the electromagnetic scalar and vector potentials, $\phi$ and $A$, and by working in Coulomb gauge $\nabla \cdot A = 0$, the Lagrangian for the system, which gives these equations (2.1)–(2.9), is written as

\begin{align}
L = \iiint d^3 x \left[ \frac{1}{2} n_0 m \left( \frac{\partial u}{\partial t} \right)^2 + \frac{n_0 e}{c} \frac{\partial u}{\partial t} \cdot A + e n_0 \phi \nabla \cdot u + m_0^2 \beta^2 n (\nabla \cdot u) + \frac{m_0^2 \beta^2}{2n_0} n^2 + \frac{1}{8\pi} \left( \frac{1}{c} \frac{\partial A}{\partial t} \cdot \phi \right)^2 - (\nabla \times A)^2 \right], \quad (2.10)
\end{align}

where the velocity of electrons $v$ is expressed in terms of the displacement vector of electrons $u$ as $v = \partial u/\partial t$. 

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All quantities of our concern are decomposed into divergence free (transverse, denoted by subscript $r$) and curl free (longitudinal, denoted by subscript $l$) part as, for example, $E_t = -(1/c)(\partial A/\partial t)$ and $E_l = -\nabla \phi$. Then our basic equations (2.1) to (2.9) can be separated into equations for transverse or longitudinal quantities as follows:

$$\Delta \phi = -4\pi en,$$

$$\nabla \left( \frac{\partial \phi}{\partial t} \right) = 4\pi env_l,$$

$$\frac{1}{c^2} \frac{\partial^2 A}{\partial t^2} - \Delta A = \frac{4\pi en_0}{c} v_t,$$

$$n_0 m \frac{\partial v_l}{\partial t} = -n_0 e \nabla \phi - m \beta^2 v n,$$

$$m \frac{\partial v_l}{\partial t} = - e \frac{\partial A}{\partial t}$$

inside the metal and

$$\frac{1}{c^2} \frac{\partial^2 A}{\partial t^2} - \Delta A = 0,$$

in the vacuum.

So-called non-radiative surface plasma modes are solutions which go to zero as $z \rightarrow \pm \infty$. Non-radiative eigenmodes can be easily obtained by seeking the solutions of type $f(x, t) = g(z) \exp(i(k \cdot r - \omega t))$ with a function $g(z)$ which goes to zero as $z \rightarrow \pm \infty$, where $x = (r, z)$ with $r = (x, y)$ and $k$ is the wave vector parallel to the surface. For $v_l(x, t)$, $\phi(x, t)$, and $n(x, t)$ the function $g(z)$ is of the form $\exp(\gamma z)$ for $z<0$ and vanishes for $z>0$. For $A(x, t)$ or $v_t(x, t)$, the function $g(z)$ is of the form $\exp(\alpha z)$ for $z<0$ and is of the form $\exp(-\delta z)$ or vanishes for $z>0$. Here $\alpha$, $\delta$ and $\gamma$ are related to $k$ and $\omega$ as

$$c^2 \alpha^2 = c^2 k^2 + \omega_p^2 - \omega^2,$$

$$c^2 \delta^2 = c^2 k^2 - \omega^2,$$

$$\beta^2 \gamma^2 = \beta^2 k^2 + \omega_s^2 - \omega^2,$$

where $k = |k|$ and $\omega_s^2 = 4\pi n_0 \epsilon_0^2 / m$. For simplicity we drop subscript $k$ of $\omega_k$, $\alpha_k$, $\delta_k$ and $\gamma_k$ though they are quantities dependent on $k$.

Then the fluctuations due to the non-radiative surface plasma mode can be expanded by these eigenmodes as follows:

$$A(x, t) = \left( \frac{4\pi c^2}{S} \right)^{1/2} \times \left\{ \begin{array}{ll}
\sum_k q_k (i\alpha_k + k\tilde{z}) e^{i\omega_k t} & ; z < 0 \\
\sum_k \bar{q}_k (-i\delta_k - k\tilde{z}) e^{-i\omega_k t} & ; z > 0
\end{array} \right.$$

$$\phi(x, t) = \left( \frac{4\pi}{S} \right)^{1/2} \times \left\{ \begin{array}{ll}
\sum_k \phi_k e^{i\gamma k \cdot r} & ; z < 0 \\
0 & ; z > 0
\end{array} \right.$$
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\[ n(x, t) = -\left(\frac{4\pi}{S}\right)^{1/2} \times \left\{ \begin{array}{ll} \sum_k \frac{k^2 - \omega^2}{4\pi} \phi_k e^{i(k\hat{r} \cdot r + k\gamma z)} & ; z < 0 \\ 0 & ; z > 0 \end{array} \right. \]  

(2.20)

\[ v_n(x, t) = -\left(\frac{4\pi}{S}\right)^{1/2} \times \left\{ \begin{array}{ll} \frac{1}{4\pi\varepsilon_0} \sum_k \phi_k (i\omega k + \gamma k^\prime) e^{i(k\hat{r} \cdot r + k\gamma z)} & ; z < 0 \\ 0 & ; z > 0 \end{array} \right. \]  

(2.21)

\[ v_r(x, t) = -\left(\frac{4\pi}{S}\right)^{1/2} \times \left\{ \begin{array}{ll} \frac{e}{m} \sum_k a_k (i\omega k + k\hat{z}) e^{i(k\hat{r} \cdot r + k\gamma z)} & ; z < 0 \\ 0 & ; z > 0 \end{array} \right. \]  

(2.22)

\[ E(x, t) = -\left(\frac{4\pi}{S}\right)^{1/2} \times \left\{ \begin{array}{ll} \sum_k \left[ \phi_k (i\omega k + k\hat{z}) e^{i(k\hat{r} \cdot r + k\gamma z)} + \phi_\ast (i\omega k + \gamma k^\prime) e^{i(k\hat{r} \cdot r + k\gamma z)} \right] e^{ik\cdot r} & ; z < 0 \\ \sum_k \phi_k (-i\delta k + k\hat{z}) e^{-i\omega t - i(k\hat{r} \cdot r + k\gamma z)} & ; z > 0 \end{array} \right. \]  

(2.23)

\[ H(x, t) = i\left(\frac{4\pi e^2}{S}\right)^{1/2} \times \left\{ \begin{array}{ll} (-1) \sum_k a_k (a^2 - \omega^2) \hat{k} \times \hat{z} e^{i(k\hat{r} \cdot r + k\gamma z)} & ; z < 0 \\ \sum_k \phi_k (k^2 - \omega^2) \hat{k} \times \hat{z} e^{-i\omega t - i(k\hat{r} \cdot r + k\gamma z)} & ; z > 0 \end{array} \right. \]  

(2.24)

where \( \hat{k} = \hat{k}/|\hat{k}| \), \( \hat{z} \) is a unit vector of positive \( z \) direction and \( S \) is the area of the boundary surface \( z = 0 \). To ensure that \( \phi \) and \( A \) are real, coefficients must satisfy

\[ \phi_k = \phi_{-k}, \quad a_k = a_{-k}, \quad q_k = q_{-k}. \]  

(2.25)

Besides the continuities of \( H \) and the tangential component of \( E \) at the boundary surface \( z = 0 \), we require the normal component of the electronic velocity \( v_z \) vanishes at \( z = 0 \), that is, electrons in the metal are reflected specularly at the boundary surface,

\[ \hat{q}_k = -\frac{\alpha}{\beta} \hat{q}_k \quad \text{and} \quad \phi_k \]  

(2.26)

\[ q_k = -\frac{a^2 - k^2}{k^2 - \omega^2} q_k \]  

(2.27)

\[ \phi_k = \frac{k}{\gamma} \omega^2 q_k. \]  

(2.28)

Eliminating \( q_k \) and \( \hat{q}_k \) in Eqs. (2.18)~(2.24) by using Eqs. (2.26)~(2.28), we have the following expressions:

\[ A(x, t) = \left(\frac{4\pi e^2}{S}\right)^{1/2} \times \left\{ \begin{array}{ll} \sum_k \phi_k \frac{\gamma}{k_0^2} (i\omega k + k\hat{z}) e^{i(k\hat{r} \cdot r + k\gamma z)} & ; z < 0 \\ \sum_k \phi_k \frac{\gamma}{k_0^2} (a^2 - \omega^2) (-i\delta k + k\hat{z}) e^{-i\omega t - i(k\hat{r} \cdot r + k\gamma z)} & ; z > 0 \end{array} \right. \]  

(2.18')
\[ E(x, t) = -\left(\frac{4\pi}{S}\right)^{1/2} \]
\[ \times \left[ \sum_k \phi_k \left( \frac{\alpha^2 - k^2}{k^2 - \delta^2} - \frac{\delta}{k} \right)^{-1} \left( i\alpha \hat{k} + k \hat{z} \right) e^{i\alpha x} + (i\hat{k} \hat{z}) e^{i\alpha x} \right] e^{ikr}; z < 0 \]
\[ \sum_k \phi_k \left( \frac{\alpha^2 - k^2}{k^2 - \delta^2} - \frac{\delta}{k} \right)^{-1} \left( -i\delta \hat{k} + k \hat{z} \right) e^{-i\delta x - ikr}; z > 0 , \]
\[ (2.23') \]
\[ H(x, t) = -i \left(\frac{4\pi e^2}{S}\right)^{1/2} \times \left[ \sum_k \phi_k \frac{\gamma}{k \omega_0} (\alpha^2 - k^2) \hat{k} \hat{z} e^{i\alpha x}; z < 0 \right] \]
\[ \sum_k \phi_k \frac{\gamma}{k \omega_0} (\alpha^2 - k^2) \hat{k} \hat{z} e^{-i\delta x + ikr}; z > 0 , \]
\[ (2.24') \]
\[ v(x, t) = v_l + v_r \]
\[ = \left(\frac{4\pi}{S}\right)^{1/2} \frac{1}{4\pi n_0 e} \times \left[ \sum_k \phi_k \left( (i\hat{k} \hat{z}) e^{i\alpha x} - \frac{\gamma}{k} (i\alpha \hat{k} + k \hat{z}) e^{i\alpha x} \right) \right] e^{ikr}; z < 0 \]
\[ ; z > 0 , \]
\[ (2.29) \]
\[ u(x, t) = \left(\frac{4\pi}{S}\right)^{1/2} \frac{1}{4\pi n_0 e} \times \left[ \sum_k \phi_k \left( (i\hat{k} \hat{z}) e^{i\alpha x} - \frac{\gamma}{k} (i\alpha \hat{k} + k \hat{z}) e^{i\alpha x} \right) \right] e^{ikr}; z < 0 \]
\[ ; z > 0 , \]
\[ (2.30) \]

Now the Lagrangian (2.10) is expressed in terms of \( \phi_k \) and \( \dot{\phi}_k \) as
\[ L = \sum_k \left[ -\frac{1}{4\omega_0^2 F(k)} \phi_k \dot{\phi}_k - F(k) \phi_k \dot{\phi}_k \right] , \]
\[ (2.31) \]
where
\[ F(k) = \left[ \frac{\gamma^2}{k^2} a^2 + k^2 + \frac{\gamma^2}{k^2} \frac{\omega_0^2 - \omega^2}{\omega_0^2} \frac{\alpha^2 - k^2}{\omega_0^2} \frac{\gamma^2 + k^2}{\gamma} \right]^{-1} , \]
\[ (2.32) \]
and
\[ G(k) = \frac{\omega_0^2 + \omega^2}{4\gamma^2} \frac{\gamma^2 - k^2}{4\gamma} + \gamma^2 + k^2 + k \left( \frac{\delta}{k} \frac{\gamma^2 - k^2}{\gamma} - \frac{\delta}{k} \right) \]
\[ + \left( \frac{\delta}{k} \frac{\gamma^2 - k^2}{\gamma} - \frac{\delta}{k} \right)^2 \frac{\gamma^2 + k^2}{4\gamma} + \left( \frac{\delta}{k} \frac{\gamma^2 - k^2}{\gamma} - \frac{\delta}{k} \right)^2 \frac{\gamma^2 + k^2}{4\gamma} . \]
\[ (2.33) \]
Defining the canonical momenta \( \pi_k \) conjugate to the variables \( \phi_k \) as
\[ \pi_k = \frac{\partial L}{\partial \dot{\phi}_k} = -\frac{1}{2\omega_0^2 F(k)} \dot{\phi}_k , \]
\[ (2.34) \]
we find Hamiltonian \( H \) of the system as
\[ H = -\sum_k [\omega_0^2 F(k) \pi_k \pi_k + G(k) \phi_k \dot{\phi}_k] . \]
\[ (2.35) \]
We introduce the creation and annihilation operators for the surface plasmon field, \( a_k \) and \( a_k^\dagger \), which are defined by
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\[ \phi_k = \left( \frac{\hbar \omega_p^2 F(k)}{\omega} \right)^\frac{1}{2} (a_k - a_{-k}^\dagger), \]

\[ \pi_k = i \left( \frac{\hbar \omega}{4 \omega_p^2 F(k)} \right)^\frac{1}{2} (a_k + a_{-k}^\dagger), \]

(2.36)

and satisfy the commutation relations

\[ [a_k, a_{k\prime}] = [a_k^\dagger, a_{k\prime}^\dagger] = 0, \]

\[ [a_k, a_{k\prime}^\dagger] = \delta_{k, k\prime}. \]

(2.37)

In terms of these operators, our Hamiltonian (2.35) can be written as

\[ H = \frac{1}{2} \sum_k \hbar \omega_k (a_k a_k^\dagger + a_{-k}^\dagger a_k), \]

provided that \( \omega_k \) satisfies the relation,

\[ \omega^2 = 4 \omega_p^2 F(k) G(k). \]

(2.38)

In virtue of relation (2.17), Eq. (2.38) is rewritten as

\[ [\gamma (a \delta - k^2) + (a - \delta) k^2] Z(k) = 0, \]

(2.39)

where

\[ Z(k) = \gamma (k^2 - a \delta)(k^2 - \delta^2) [\delta (a - \delta)(a^2 + k^2) + (a^2 - k^2)^2] \]

\[ + \frac{a - \delta}{a + \delta} k^2 [\delta (a^2 + k^2)(a^2 - k^2)^2 + \delta (a^2 + k^2)(k^2 - \delta^2)^2] \]

\[ - 4 \alpha \delta (a + \delta)(k^4 - \delta^2)(k^2 - a \delta). \]

From Eq. (2.39), a dispersion

\[ \gamma (a \delta - k^2) + (a - \delta) k^2 = 0 \]

(2.40)

is derived. This dispersion relation multiplied by \((a + \delta)\) is written as

\[ \gamma (a + \delta) \omega^2 - (\gamma \delta + k^2) \omega_p^2 = 0, \]

(2.40')

which coincides with the one derived by the corresponding classical theory.\(^5\) In the retarded region \(ck/\omega_p < 1\), we find \(k/\gamma < 1\) because \(\beta k/\omega_p \ll ck/\omega_p < 1\), so that Eq. (2.40) is reduced to the equation

\[ a \delta - k^2 = 0. \]

This is solved for \( \omega \) to give the well-known dispersion relation

\[ \omega^2 = \frac{1}{2} (\omega_p^2 + 2c^2 k^2 - [\omega_p^2 + 4c^2 k^2]^{1/2}), \]

(2.41)

which was first derived by Stern and quoted by Ferrell\(^6\) for semi-infinite free electron gas with the retardation effect included, and also derived from different approaches, the surface impedance matching approach\(^7\) and the linear-response correlation-function approach.\(^8\) It is known that this dispersion relation is in good agreement with the
observed one for the surface polariton (the surface plasmon in the retarded region $ck/\omega_p \ll 1$). In the electrostatic region of $k$, $\omega_p/\beta > k > \omega_p/c$, the dispersion relation (2.40) gives the well-known linear dispersion relation

$$\omega^2 = \frac{\omega_p^2}{2} \left( 1 + \frac{\sqrt{2} \beta k}{\omega_p} \right).$$  

(2.42)

Although there remains the possibility of the existence of other branches due to $Z(k)$ factor in Eq. (2.39), in this paper we restrict our interest to the surface plasmon whose dispersion relation is given by Eq. (2.40). Other branches which come from

$$Z(k) = 0$$

will be discussed in a forthcoming paper.

Now all our fluctuations of fields are expressed by surface plasmon operators, $a_k$ and $a_k^\dagger$:

$$\phi(x, t) = \left(\frac{4\pi}{S}\right)^{1/2} \times \left[ \sum_k \frac{\hbar \omega_p^2 F(k)}{\omega} \right]^{1/2} \left( a_k - a_k^\dagger \right) e^{i z + i k \cdot r},$$

$$z < 0$$

$$z > 0,$$

(2.43)

$$A(x, t) = -i \left(\frac{4\pi c^2}{S}\right)^{1/2} \times \left[ \sum_k \frac{\hbar \omega_p^2 F(k)}{\omega} \right]^{1/2} \left( \frac{\omega^2}{\omega_p} \frac{\gamma}{k} (i \alpha k + \Delta \Delta)(a_k + a_k^\dagger) e^{i z + i k \cdot r},$$

$$z < 0$$

$$z > 0,$$

(2.44)

$$E(x, t) = -\left(\frac{4\pi}{S}\right)^{1/2} \times \left[ \sum_k \frac{\hbar \omega_p^2 F(k)}{\omega} \right]^{1/2} \left( \frac{\omega^2}{\omega_p^2} \frac{\gamma}{k} (i \alpha k + \Delta \Delta)(a_k - a_k^\dagger) e^{i z - i k \cdot r},$$

$$z < 0$$

$$z > 0,$$

(2.45)

$$H(x, t) = -\left(\frac{4\pi c^2}{S}\right)^{1/2} \times \left[ \sum_k \frac{\hbar \omega_p^2 F(k)}{\omega} \right]^{1/2} \left( \frac{\omega^2}{\omega_p^2} \frac{\gamma}{k} (\alpha^2 - k^2) \Delta \Delta (a_k + a_k^\dagger) e^{-i z + i k \cdot r},$$

$$z < 0$$

$$z > 0,$$

(2.46)
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\[ n(x, t) = - \left( \frac{4\pi}{S} \right)^{1/2} \times \left\{ \sum_i \left[ \frac{\hbar v_F^2 F(k)}{\omega} \right]^{1/2} \gamma^2 \left[ \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right] \right\}^i \times \left( \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right)^i ; z < 0 \]
\[ u(x, t) = - \left( \frac{4\pi}{S} \right)^{1/2} \times \left\{ \sum_i \left[ \frac{\hbar v_F^2 F(k)}{\omega} \right]^{1/2} \gamma^2 \left[ \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right] \right\}^i \times \left( \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right)^i ; z > 0, \]
\[ v(x, t) = i \left( \frac{4\pi}{S} \right)^{1/2} \times \left\{ \sum_i \left[ \frac{\hbar v_F^2 F(k)}{\omega} \right]^{1/2} \gamma^2 \left[ \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right] \right\}^i \times \left( \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right)^i ; z < 0 \]
\[ w(x, t) = i \left( \frac{4\pi}{S} \right)^{1/2} \times \left\{ \sum_i \left[ \frac{\hbar v_F^2 F(k)}{\omega} \right]^{1/2} \gamma^2 \left[ \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right] \right\}^i \times \left( \frac{a_k - a_k^* \overline{a}_k \overline{a}_k} {4\pi e} \right)^i ; z > 0, \]

\[ 2.47 \]

\[ 2.48 \]

\[ 2.49 \]

§ 3. The electron-SP vertex function

In the preceding section, the non-radiative surface plasma mode was quantized and all the fluctuations of the fields were expressed in terms of the surface plasmon creation and annihilation operators, \( a_k^* \) and \( a_k \). Then the interaction vertex function of the surface plasmon with a charged particle is determined without ambiguity by substituting the expressions (2·43) and (2·44) for \( \phi(x, t) \) and \( A(x, t) \) into the electron-SP interaction Hamiltonian,

\[ H_{el-sp} = \iiint d^3x \rho_e(x, t) \phi(x, t) - \frac{1}{C} \iiint d^3x j_e(x, t) \cdot A(x, t). \]

Here \( \rho_e(x, t) \) and \( j_e(x, t) \) are the charge and current density of the incident electron respectively and given by

\[ \rho_e(x, t) = \frac{1}{V} \sum_{\mathbf{p}} \left[ \mathbf{b}^\dagger \mathbf{b} \right] \left( \mathbf{E}(\mathbf{p}) + \overline{\mathbf{E}(\mathbf{p})} \right) e^{-i(\mathbf{p} \cdot \mathbf{r} - \omega t + \mathbf{k} \cdot \mathbf{x}} \right] \]

and

\[ j_e(x, t) = \frac{1}{V} \sum_{\mathbf{p}} \left[ \mathbf{b}^\dagger \mathbf{b} \right] \left( \mathbf{E}(\mathbf{p}) + \overline{\mathbf{E}(\mathbf{p})} \right) e^{-i(\mathbf{p} \cdot \mathbf{r} - \omega t + \mathbf{k} \cdot \mathbf{x}} \right] \]

where \( \mathbf{b}^\dagger \) and \( \mathbf{b} \) are the creation and annihilation operators of an incident electron with the momentum \( \mathbf{p} \) and energy \( \mathbf{E}(\mathbf{p}) \) respectively and \( V \) is the volume of the whole system. The incident electron is assumed to have a large velocity so that it is treated as a spinless relativistic particle. Therefore, the electron energy \( \mathbf{E}(\mathbf{p}) \) in Eqs. (3·2) and (3·3) is given by

\[ \mathbf{E}(\mathbf{p}) = \left( m^2 c^4 + \mathbf{p}^2 c^2 \right)^{1/2}. \]
For convenience, we introduce the kinetic energy of the incident and scattered electrons, \( E = \tilde{E}(p) - mc^2 \) and \( E' = \tilde{E}(p') - mc^2 \).

The transition matrix element \( \langle p'; k|H_{el-ws}|p \rangle \) for the electron from the state of momentum \( p \) to that of momentum \( p' \) by exciting a surface plasmon with wave vector \( k \) is calculated for the case of normal incidence, \( p = -pE \);

\[
\langle p'; k|H_{el-ws}|p \rangle = \frac{S}{V} \left( \frac{4\pi \varepsilon^2 \hbar \omega_p}{S} \right)^{1/2} \gamma F(k)^{1/2} \frac{\omega_p}{\omega} \frac{\partial \hat{p}}{m \Delta E} \\
\times \left[ \frac{\omega^2}{\omega^2} + \frac{1 + \Delta \hat{p}/2p + (\hbar \Delta \sigma^2)/2p \Delta \sigma + i(h\Delta \sigma/\Delta \hat{p})}{\alpha^2 + (\Delta p/\hbar)^2} \right] \nu \omega_p^2 - \frac{1 + \Delta \hat{p}/2p + (\hbar \Delta \sigma^2)/2p \Delta \sigma - i(h\Delta \sigma/\Delta \hat{p})}{\omega^2} \frac{\Delta E(\tilde{E}(p') - \tilde{E}(p))}{2p \Delta \sigma^2} \left[ 1 - i(\Delta p/\hbar \gamma) \right].
\] (3.5)

Here \( \Delta \hat{p} = p_x - p_{x'} < 0 \) and \( \Delta E = \tilde{E}(p) - \tilde{E}(p') = \hbar \omega_p \). For the case of small angles of scattering, \( \theta = \hbar k/p \ll 1 \), the following relations hold from energy-momentum conservation, with \( \theta_k = \Delta E(E + mc^2)/E(E + 2mc^2) \);

\[
\frac{\Delta \hat{p}}{p} = \theta_k \left[ 1 + \frac{1}{2} \frac{\theta_k}{\theta_k} \left( \frac{mc^2}{E} + E \right)^2 + \frac{\theta_k^2}{\theta_k} \right],
\]

\[
\frac{\Delta E(\tilde{E}(p') - \tilde{E}(p))}{2p \Delta \sigma^2} = \left( \frac{1 - \theta_k}{2} \right) \frac{\theta_k^2}{\theta_k}.
\] (3.6)

In the electrostatic region, \( \omega_p/k \gg \omega_p/c \), the dispersion relation of the surface plasmon is given by Eq. (2.42) and also the following estimations:

\[
\frac{k}{\alpha} \approx 1, \quad \frac{k}{\gamma} \approx \sqrt{2} \beta k \omega_p, \quad \gamma^2 F(k)/k = \frac{1}{2} \left( 1 + \frac{\beta k}{\sqrt{2} \omega_p} \right),
\] (3.7)

are obtained up to the first order in \( \beta k/\omega_p \) and \( \omega_p/c \). Therefore, in this electrostatic region, the interaction vertex function (3.5) becomes

\[
\langle p'; k|H_{el-ws}|p \rangle = \frac{S}{V} \left( \frac{4\pi \varepsilon^2 \hbar \omega_p}{S} \right)^{1/2} \frac{mc^2 + E}{mc^2 + (\Delta \hat{p}/\hbar)^2},
\] (3.8)

where only the lowest order term in \( \beta k/\omega_p \) and \( \omega_p/c \) is remained and \( \omega_1 = \omega_p/\sqrt{2} \). This vertex function coincides with the one derived first by Stern and Ferrell in the electrostatic approximation by semi-classical argument,

\[
\langle p'; k|H_{el-ws}|p \rangle = \frac{S}{V} \left( \frac{4\pi \varepsilon^2 \hbar \omega_p}{S} \right)^{1/2} \frac{k^{1/2} (\Delta \hat{p}/\hbar)^2}{k + (\Delta \hat{p}/\hbar)^2},
\] (3.9)

except for a relativistic correction factor.

In the extremely retarded region of \( k, \beta k/\omega_p \ll p/mc < 1 \), instead of Eqs. (3.6) and (3.7), the dispersion relation becomes

\[
\omega = ck
\] (3.10)

and the estimations...
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\[
\frac{k}{a} \approx \frac{ck}{\omega_p}, \quad \frac{\delta}{k} \approx \left(1 - \frac{\beta}{c}\right) \frac{ck}{\omega_p}, \quad \frac{k}{\gamma} \approx \frac{\beta}{c} \frac{ck}{\omega_p},
\]

\[
\frac{\gamma^2 F(k)}{k} \approx \left(\frac{ck}{\omega_p}\right)^3,
\]

are obtained in the lowest order approximation in \(ck/\omega_p\). Keeping only the lowest order term in \(ck/\omega_p\), we obtain the transition matrix element in the extremely retarded region as

\[
\langle p'; k|H_{el.-sp}|p \rangle = \frac{S}{V} \left(\frac{4 \pi e^2 \hbar}{S}\right)^{1/2} \left(\frac{mc^2 + E}{mc^2}\right) \left(\frac{v}{c}\right)^2 \frac{c}{(\omega_p k)^{1/2}},
\]

where \(v\) is the velocity of the incident electron with momentum \(p\). It should be noted that Eq. (3·12) comes from the vacuum part of the second integral of the interaction Hamiltonian (3·1), \(- (1/c) \int \! df d^3x \hat{\varphi}(x) \cdot \mathbf{A}(x)\), which has not been taken into account in the works so far. It is also noted that the \(k\) dependence of the vertex function (3·12), where the retardation effect is included, is greatly different from that of the electrostatic one, (3·8) or (3·9). The vertex function (3·12) diverges in the long wavelength limit \(k \to 0\), whereas the usual electrostatic one, (3·8) or (3·9), goes to zero as \(k \to 0\) if it is simply extrapolated to the retarded region \(k \ll \omega_p/c\). This divergence in the long wavelength limit is characteristic of the scattering processes where a zero mass quanta is exchanged. In our case, the surface plasmon satisfying the dispersion relation \(\omega \approx ck\) in the long wavelength limit \(k \to 0\) plays a role of this zero mass quanta.

§ 4. Discussion

The angular distribution of inelastic scattering intensity of fast electrons suffering from the surface energy loss in passing through a metal foil has been measured by several people,\(^1\) and it is well known that the differential surface loss intensity of fast electrons behaves as \(\theta^{-3}\) with the scattering angle \(\theta\) in a region \(\theta \gg \theta_E\).\(^{5,10}\) This angular dependence was predicted by Stern and Ferrell\(^1\) by using the electrostatic electron-SP vertex function (3·9). The differential surface loss probability \(\nu(\theta, \phi) d\Omega\) of an electron (with velocity \(v\)) entering parallel to the normal of the metal surface of area \(S\) is given by

\[
\nu(\theta, \phi) d\Omega = \frac{V}{S} \frac{2\pi}{\hbar} |\langle p'; k|H_{el.-sp}|p \rangle|^2 \rho(E) d\Omega
\]

with the differential density of states

\[
\rho(E) = \frac{V p^2}{(2\pi)^2 \hbar^2} \frac{1}{v} d\Omega.
\]

Substitution of Eq. (3·9) into Eq. (4·1) yields the scattering coefficient \(\nu(\theta, \phi)\):

\[
\nu(\theta, \phi) = \frac{e^2}{4\pi \hbar^2} \frac{\theta_E}{(\theta^2 + \theta_E^2)^{3/2}}.
\]

This scattering coefficient (4·3) behaves as \(\theta^{-3}\) for large angles \(\theta \gg \theta_E\). Our interaction vertex (3·5) becomes Eq. (3·8) in the electrostatic region, \(\omega_p/\beta \gg k \gg \omega_p/c\), and also gives the scattering coefficient.
which gives the same \( \theta^{-3} \) behavior for \( \theta \gg \theta_0 \) as Eq. (4·3) does. In the inelastic scattering of fast electrons (\( E = 30 \sim 50 \) keV) passing through metal foil (Al or Mg, for example), \( \theta_0 \) is of order of \( 10^{-3} \sim 10^{-4} \) radian and this \( \theta^{-3} \) behavior has been confirmed experimentally over a certain range of \( \theta \), \( 10^{-3} \sim 10^{-2} \) radian.\(^1\)

It was usually said, by extrapolating the electrostatic scattering coefficient to the retarded region, that the surface loss intensity of scattered electrons should vanish at \( \theta = 0 \) in the case of normal incidence.\(^1\) There is, however, no justification of extrapolating the electrostatic vertex function to the extremely retarded region. Since our vertex function (3·5) was obtained with the retardation effect taken into account, the vertex function (3·12), the long wavelength limit of the vertex function (3·5), may be expected to give more reliable information of the scattering coefficient in the region near \( k = 0 \). Our vertex function (3·12) gives the scattering coefficient,

\[
\mu(\theta, \phi) = \frac{e^2}{\pi \hbar v} \left( \frac{m c^2 + E}{m c^2} \right) \left( \frac{m c^2}{E} \right) \frac{1}{\hbar \omega_0} \frac{1}{\theta}, \tag{4·5}
\]

in the extremely retarded region \( k \ll \omega_0/c \), that is, in the small angle region \( \theta \ll \theta_0 = \hbar \omega_0/cp \).

We note that the scattering coefficient (4·5) diverges inversely with \( \theta \) as \( \theta \) goes to zero. This divergence is due to the fact that in the long wavelength limit the dispersion relation of the surface plasmon becomes \( \omega \approx \omega_0 k \) and therefore the electron scattered without deflection can excite a soft plasmon with the energy \( \hbar c k \approx \omega_0 k \approx 0 \).

The angular distribution of inelastically scattered electrons passing through a metal foil was measured mostly in the region \( \theta \gg \theta_0 \) and the \( \theta^{-3} \) dependence of the differential surface loss intensity was confirmed in the region \( \theta \gg \theta_0 (\approx \theta_0) \). In the small angle region \( \theta \ll \theta_0 \), however, the experimental data are very poor and not accurate owing to the finite aperture of the primary beam. Therefore we do not have so far enough experimental data to check the characteristic \( \theta^{-1} \) behavior of the surface loss intensity (4·5). But, from a few available loss intensity data\(^9\) measured near \( \theta \approx \theta_0 \), we can say that they do not give any indications that the loss intensity approaches zero as \( \theta \) decreases to zero. Near \( \theta \approx \theta_0 \), it is yet increasing as \( \theta \) decreases, and it rather behaves as \( \theta^{-1} \) which fits a simple extrapolation of our equation (4·5) to the boundary region \( \theta \approx \theta_0 \). It is awaited to carry out experiments in the region \( \theta < \theta_0 \) with enough accuracy to check Eq. (4·5).

We conclude this section by commenting on the total characteristic surface loss probability. It is known that the scattering coefficient (4·3) in the electrostatic approximation gives the characteristic surface loss probability \( P \) as

\[
P = \int_0^\pi \mu(\theta, \phi) 2 \pi \theta d\theta = \frac{\pi e^2}{2 \hbar v}. \tag{4·6}
\]

For the electron beam with \( E = 34 \) keV, the value of \( P \) calculated from Eq. (4·6) is 3.3 %. If the integrand in the region \( \theta < \theta_0 \) is replaced by Eq. (4·5), the value of \( P \) is enhanced slightly to 3.4 %. Experimental value of \( P \) for Mg foil of a thickness 100 A is 3~4 \%,\(^10\) so that this change is not appreciable experimentally.
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References

2) E. S. Stern and R. A. Ferrell, Phys. Rev. 120 (1960), 130.