Surface-plasmon dispersion relation for the inhomogeneous charge-density medium

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The surface-plasmon dispersion relation is derived for the plane-bounded electron gas when there is an inhomogeneous charge-density distribution in the plasma. The hydrodynamical model is used. Both φ and $d\varphi/dx$ are taken to be continuous at the surface of the slab, where φ is the scalar potential. The dispersion relation is compared with the theoretical works of Stern and Ferrell and of Harsh and Agarwal. It is also compared with the observations of Kunz. A dispersion relation for the volume-plasmon oscillations is derived which resembles the well-known relation of Bohm and Pines.

I. INTRODUCTION

The surface plasmons at the interface of a bulk metal and vacuum were first predicted by Ritchie.[1] They were observed by Powell and Swan.[2,3] Stern and Ferrell[4] found that the surface plasmons at the interface of a metal and its oxide can account for some of the perplexing peaks occurring in the inelastic scattering of fast electrons by metal foils. Bloch[5,6] proposed a quantum-hydrodynamical model which was extended by Ritchie and Wilems.[7]

Recently Smithard[8] and Gariere *et al.*[9] have experimentally studied the surface-plasmon modes of small metallic particles. Kunz,[10] Swan *et al.*,[11] and Kloos[12] have found the dispersion of surface plasmons from the electron-energy-loss measurements. A double-well model has been used by Boardmen *et al.*[13,14] to study the surface plasma oscillations. This problem has also been recently studied by Arakawa *et al.*[15] and Harsh and Agarwal.[16,17]

We propose here a dispersion relation of surface plasma oscillations for the semi-infinite plane-bounded electron gas with inhomogeneous electron density along the normal to the plane (that is, along the x direction). We use the hydrodynamical model.[5–7] The calculated surface-plasmon dispersion relation shows good agreement with other models[1,4,18,19] and also with experiments.[10]

We also obtain a dispersion relation for the volume-plasmon oscillations which agrees with the well-known relation of Bohm and Pines.[18,20]

II. MATHEMATICAL FORMULATION

Consider a uniform positive neutralizing background for the electron gas in a plane-bounded region of thickness a along the positive x direction. The other edges are along the y and z directions. The plane-bounded region has been taken in such a way that the particle density is $n_0+n_0\sin(Kx)$ for the region $0<x<a$ and is zero beyond it. We name the region $0<x<a$ as interior and $x>a$ as exterior, for convenience. The velocity $\mathbf{v}$ and electrostatic potential φ of a hydrodynamic fluid satisfy

$$m\frac{d\mathbf{v}}{dt}=e\nabla\varphi-\nabla\int_0^{n(x,y,z,t)}\frac{dP(n')}{n'}\ , \qquad (1)$$

$$\nabla^2\varphi=4\pi e[n(x,y,z,t)-D_i(x,y,z)]\ , \qquad (2)$$

where m is the electron mass, e is the electron charge, n (and n') the electron concentration, P the Fermi pressure, D_i the ion density, and d/dt a comoving time derivative.

The Fermi pressure $P(n')$, which accounts for the Pauli exclusion principle, is given by

$$P(n)=\frac{(3\pi^2)^{2/3}}{5}\frac{\hbar^2}{m}n^{5/2}\ . \qquad (3)$$

Defining a velocity potential $\Psi(x,y,z,t)$ by

$$\mathbf{v}=-\nabla\Psi\ , \qquad (4)$$

Eq. (1) becomes, on simplification,

$$\frac{d\Psi}{dt}=\tfrac{1}{2}(\nabla\Psi)^2\frac{1}{m}\int_0^{n}\frac{dP(n')}{n'}-\frac{e}{m}\varphi\ . \qquad (5)$$

The equation of continuity is

$$\partial n/\partial t=\nabla\cdot(n\nabla\Psi)\ . \qquad (6)$$

Using the process of linearization, we write

$$n(x,y,z,t)=n_0(x,y,z)+n_1(x,y,z,t)+n_2(x,y,z,t)+\cdots\ , \qquad (7)$$

$$\varphi(x,y,z,t)=\varphi_0(x,y,z)+\varphi_1(x,y,z,t)+\varphi_2(x,y,z,t)+\cdots\ , \qquad (8)$$

$$\Psi(x,y,z,t)=\Psi_1(x,y,z,t)+\Psi_2(x,y,z,t)+\cdots\ , \qquad (9)$$

where it is assumed that for the electron densities $n_0 \gg n_1 \gg n_2$. The particle density decreases as the fluid spreads in the medium. Substituting Eqs. (7)–(9) in Eq. (5), to zeroth order,

$$\int_0^{n_0} \frac{dP(n')}{n'} = e\varphi_0 , \tag{10}$$

and Eqs. (1) and (2) reduce to

$$\tfrac{5}{2} P n_0^{2/3} = e\varphi_0 , \tag{11}$$

$$\nabla^2 \varphi_0 = 4\pi e (n_0 - D_i) . \tag{12}$$

To first order, Eqs. (5), (2), and (6) become

$$\frac{\partial \Psi_1}{\partial t} = -\frac{e}{m}\varphi_1 + \frac{5}{3}\frac{P}{m}\frac{n}{n_0^{1/3}} , \tag{13}$$

$$\nabla^2 \varphi_1 = 4\pi n_1 e , \tag{14}$$

$$\partial n_1/\partial t = \nabla \cdot [n_0(x,y,z)\nabla(\Psi_1)] . \tag{15}$$

For a plane-bounded electron gas, the coordinate axes may be taken so that the particle field or fluid density varies as

$$n_0(x,y,z) = \begin{cases} n_0 + n_0 \sin(Kx), & \text{inside (I)} \\ 0, & \text{outside (II)} \end{cases} \tag{16}$$

where K is a wave vector. The variation of the electron density is only along the normal to the plane (that is, along the positive x axis).

From Eqs. (13)–(16),

$$\frac{\partial \Psi_1}{\partial t} = -\frac{e}{m}\varphi_1 + \frac{v_F^2}{3n_0} n_1 , \tag{17}$$

$$\nabla^2 \varphi_1 = 4\pi n_1 e , \tag{18}$$

$$\partial n_1/\partial t = n_0 \nabla^2 \Psi_1 , \tag{19}$$

where $v_F = (5P/m)n_0^{2/3} = (\hbar/m)^2(3\pi^2 n_0)^{2/3}$ for the absolute temperature $T=0$. Eliminating Ψ_1 between Eqs. (17) and (19), and using Eq. (18), we obtain

$$\left[\frac{\partial^2}{\partial t^2} + \omega_p^2 - \beta^2 \nabla^2 \right] n_1(x,y,z,t) = 0 , \tag{20}$$

where $\omega_p^2 = (4\pi n e^2/m)$ and $\beta^2 = v_F^2/3$. Equation (20) represents the condition for the volume-plasmon oscillations. Due to rectangular symmetry we can write

$$n_1(x,y,z,t) = n_1(x,y,z)e^{-i\omega_k t} , \tag{21}$$

$$n_1(x,y,z) = \sum_l X_l(x) W(y,z) . \tag{22}$$

We can interpret ω_p and ω_k as the circular frequencies of plasma oscillations and flow of charged particles, respectively. In view of Eq. (20), $X_l(x)$ in the expansion (22) satisfies

$$\partial^2 X_l/\partial x^2 = [(K')^2 - K^2] X_l(x) , \tag{23}$$

where $K^2 = (\omega_k^2 - \omega_p^2)/\beta^2$ and K' is a constant. Its solution is

$$X_l = A e^{\pm lx} \tag{24}$$

where $l^2 = [(K')^2 - K^2]$ and A is a constant.

We have taken the boundary conditions of finiteness of X_l at the origin. Therefore, Eq. (18) implies

$$\varphi_1(x,y,z,t) = \varphi_1(x,y,z)e^{-i\omega_k t} , \tag{25}$$

$$\varphi_1(x,y,z) = \sum_l \varphi_l(x) W(y,z) , \tag{26}$$

$$(\partial^2 \varphi_l/\partial x^2) - (K')^2 \varphi_l(x) = 4\pi e X_l(x) . \tag{27}$$

Rewrite Eq. (27) by adding $K^2\varphi_l(x)$ on both sides,

$$(\partial^2 \varphi_l/\partial x^2) + [K^2 - (K')^2]\varphi_l(x) = R X_l(x) + K^2 \varphi_l(x) , \tag{28}$$

where $R = 4\pi e$.

The interior solution of Eq. (28) is of the form

$$\varphi_l^{(\text{int})}(x) = -RK^{-2} X_l(x) + A' e^{K'x}, \quad 0 < x < a . \tag{29}$$

In the exterior region, there are no real charges ($n_1 = 0$), and the $\varphi_l(x)$ obeys $\nabla^2 \varphi_l = 0$, or

$$\varphi_l^{(\text{ext})}(x) = B e^{-K'x} , \quad x > a . \tag{30}$$

Using the boundary conditions at $x = a$,

$$\begin{aligned} &\varphi_l^{(\text{int})}(x) = \varphi_l^{(\text{ext})}(x) , \quad \text{(I)} \\ &\partial \varphi_l^{(\text{int})}/\partial x = \partial \varphi_l^{(\text{ext})}/\partial x , \quad \text{(II)} . \end{aligned} \tag{31}$$

We have from Eqs. (29)–(31)

$$A' = \frac{R}{2K^2 K' e^{K'a}} [K' X_l(a) + X_l'(a)] , \tag{32}$$

where $X'(a) = [\partial X(x)/\partial x]_{x=a}$. Thus the complete solution for the interior is

$$n_1^{(\text{int})}(x,y,z) = \sum_l X_l(x) W(y,z) , \tag{33}$$

$$\varphi_l^{(\text{int})}(x,y,z) = -RK^{-2} X_l(x) + \frac{\text{R}e^{K'x}}{2K^2 K' e^{K'a}} [K' X_l(a) + X_l'(a)] . \tag{34}$$

To obtain the surface-plasmon dispersion relation, we now introduce the hydrodynamical condition of zero electronic velocity normal to the surface. The acceleration $\dot{\mathbf{v}}_1$ of electrons at $x = a$ is given by

$$\dot{\mathbf{v}}_1 = \frac{e}{m}\nabla \varphi_1 - \frac{\beta^2}{n_0}\nabla n_1 = 0 . \tag{35}$$

From (33)–(35),

$$\frac{X(a)}{X'(a)} = \frac{2}{K'[1+\sin(Ka)]} \left[\frac{\beta^2 K^2}{\omega_p^2} + \tfrac{1}{2}[1+\sin(Ka)] \right] . \tag{36}$$

If ϵ_+ is the dielectric constant of the medium in which the rectangular metal slab is embedded and ϵ_- is the dielectric constant of the dielectric slab, then (36) can be modified to

$$\frac{X(a)}{X'(z)}=\frac{(1\pm\epsilon_{\pm})}{K'[1+\sin(Ka)]}\times\left[\frac{\beta^2K^2}{\omega_p^2}+\left[1\pm\frac{\epsilon_{\mp}}{1\pm\epsilon_{\pm}}\right][1+\sin(Ka)]\right] . \quad (37)$$

In deriving the dispersion relation (37), the boundary conditions at $x=a$ have been taken as

$$\varphi_l^{(\mathrm{int})}(x)=\varphi_l^{(\mathrm{ext})}(x) ,$$
$$\epsilon_+\left[\frac{\partial\varphi_l^{(\mathrm{int})}(x)}{\partial x}\right]=\epsilon_-\left[\frac{\partial\varphi_l^{(\mathrm{ext})}(x)}{\partial x}\right] . \quad (38)$$

III. RESULTS

A. Surface-plasmon dispersion relation

(i) If we put $\epsilon_{\pm}=\pm1$, $\sin(Ka)=0$, and $K'=0$ in (37), the well-known results of Stern and Ferrell[2] may be obtained in the long-wavelength limit as

$$\omega_k=\omega_p/\sqrt{2} . \quad (39)$$

At $\epsilon_{\pm}=\pm1$, (37) represents the general characteristic of surface-plasmon oscillations.

(ii) For a fixed electron density (varying the thickness a), (36) is plotted for Mg ($n=8.6\times10^{22}$ electrons/cm^{-3}) and compared with the experimental data of Ritchie and Kunz.[10] As $K\to0$ and $K'=0$, the surface mode approaches $\omega_p/\sqrt{2}$. Curves D and E are monotonically decreasing, and curve E is well fitted with Ritchie's[1,20] and Kunz's[10] results (Fig. 1).

(iii) Surface-plasmon dispersion relation (36) has also been compared with the surface-plasmon dispersion relation obtained by the x-ray emission spectra technique,[17] for the plane-bounded electron gas (Fig. 1, curve C).

B. Volume-plasmon dispersion relation

From (18) and (20)

$$\omega_K^2=\omega_p^2+\tfrac{1}{3}v_F^2K^2 , \quad (40)$$

where ω_K is the angular frequency of the free charges, ω_p is the frequency of the plasmon oscillations, and v_F is the Fermi velocity. Dispersion relation (40) closely resembles the Pines formula[18,20]

$$\omega_K^2=\omega_p^2+\tfrac{3}{5}v_F^2K^2 . \quad (41)$$

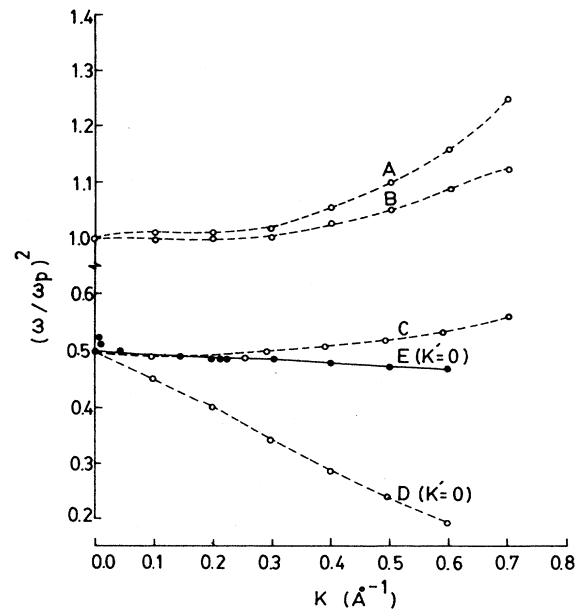

FIG. 1. Plot of $(W/W_p)^2$ as a function of K. Curve A, volume-plasmon dispersion curve [Eq. (40)]; curve B, volume-plasmon dispersion according to Bohm and Pines (Refs. 18 and 20); curve C, surface-plasmon dispersion curve for x-ray emission spectra in case of semi-infinite-plane boundary (Ref. 17); curve D, surface-plasmon dispersion curve using the hydrodynamical model, in-plane bounded electron gas, Eq. (36), for comparatively large thickness and $K'=0$; curve E, the surface-plasmon dispersion curve using the hydrodynamical model, in-plane bounded electron gas, Eq. (36), for comparatively low thickness and $K'=0$. The solid points are the experimental data of Kunz (Ref. 10).

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