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Charge Response of Jellium Surface through Ripplons

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Abstract

The feasibility of explaining the charge response properties of a jellium surface through excitation of ripplon modes on the electron fluid is examined. It is demonstrated that for a sharp equilibrium density profile the response to the field of an external source through ripplons is different from that through surface plasmons, although in the non-dispersive limit they become identical despite the radical difference in the boundary conditions associated with the two types of excitation. It is suggested that because a free surface is more likely to satisfy the boundary condition for ripplons than for surface plasmons, the surface response properties would be determined more through the excitation of ripplons.

1. Introduction

The charge response properties of a metal surface have been investigated in detail in recent years within the framework of the jellium model. The collective modes of the metal which have been taken into account in this problem are the surface and bulk plasmon modes. The usual boundary condition for obtaining the surface plasmon modes is to have the component of the electron current normal to the surface plane vanish. One may, of course, expect that for a free surface transverse displacements of the surface as in ripples or capillary waves would be likely to occur. In fact, the boundary condition for surface plasmons can only be ensured by maintaining the flatness of the surface with the use of a rigid boundary. The physical situation is closer to the condition in which ripple-like displacements of the surface of the electron gas in jellium occur. The object of this paper is to present an analysis of the charge response behaviour of the jellium surface arising out of the ripples. The analysis is done through the use of the hydrodynamic model of the electron gas.

2. Properties of Ripplons on Jellium Surface

Let us consider a semi-infinite jellium block (from $-\infty < z \leqslant 0$) in which the equilibrium electron number density and that of the background positive charge are both $n_0(\boldsymbol{r})$, given by

$$n_0(\boldsymbol{r}) \equiv n_0\,\theta(-z)\,. \tag{1}$$

The linearized equation of motion for the drift velocity $\boldsymbol{v}$ in the hydrodynamic model is given by

$$mn_0\,\partial\boldsymbol{v}/\partial t = -n_0\nabla\int V(\boldsymbol{r}-\boldsymbol{r}')\,n_1(\boldsymbol{r}')\,\mathrm{d}^3r' - m\beta^2\,\nabla n_1\,. \tag{2}$$

Here n_1 is the small change in electron density, and $V(r) = e^2/r$, the Coulomb potential between the two electrons. The term $m\beta^2 \nabla n_1$ is the pressure that leads to the dispersion of the bulk and surface plasmons through the well-known parameter β, which is of the order of the Fermi velocity. The non-occurrence of the term

$$-n_1 \nabla \int V(\boldsymbol{r}-\boldsymbol{r}')\, n_0(\boldsymbol{r}')\, \mathrm{d}^3 r'$$

is because of cancellation against the positive background. Combined with the equation of continuity

$$\nabla \cdot (n_0 \boldsymbol{v}) = -\partial n_1/\partial t, \tag{3}$$

and the appropriate boundary condition, equation (2) can be solved to give the collective modes associated with the density fluctuations.

Strictly speaking, in the hydrodynamic model one must use an effective potential defined by the equation

$$\nabla V_{\text{eff}}(\boldsymbol{r}) = g(r)\nabla V(r),$$

where $g(r)$ is the radial distribution function. But since in a degenerate high density electron gas $g(r)$ is nearly equal to unity except within a small screening distance around $r = 0$, we shall use the bare Coulomb potential throughout.

Equations (2) and (3) give us

$$\begin{aligned}\frac{\partial^2 n_1}{\partial t^2} &= \frac{n_0}{m}\nabla^2 \int V(\boldsymbol{r}-\boldsymbol{r}')\, n_1(\boldsymbol{r}')\, \mathrm{d}^3 r' + \beta^2 \nabla^2 n_1 \\ &= -\omega_{\mathrm{p}}^2\, n_1(\boldsymbol{r}) + \beta^2 \nabla^2 n_1,\end{aligned} \tag{4}$$

where $\omega_{\mathrm{p}}^2 = 4\pi n_0 e^2/m$. For surface plasmons we can assume that

$$n_1 \propto \exp(\mathrm{i}\,\boldsymbol{\kappa}\cdot\boldsymbol{\rho})\exp(-\gamma|z|)\exp\{-\mathrm{i}\,t\,\omega_{\mathrm{SP}}(\boldsymbol{\kappa})\}, \tag{5}$$

where $\boldsymbol{\rho} \equiv (x, y)$ is the two-dimensional surface coordinate and $\boldsymbol{\kappa}$ the corresponding two-dimensional wave vector. Thus equation (4) becomes

$$\omega_{\mathrm{SP}}^2(\boldsymbol{\kappa}) = \omega_{\mathrm{p}}^2 + \beta^2(\kappa^2 - \gamma^2). \tag{6}$$

Here γ can be evaluated from the boundary condition $v_z|_{z=0} = 0$, or from equation (2), i.e.

$$-m\beta^2 \frac{\partial n_1}{\partial z}\bigg|_{z=0} = n_0 \frac{\partial}{\partial z}\int V(\boldsymbol{r}-\boldsymbol{r}')\, n_1(\boldsymbol{r}')\, \mathrm{d}^3 r' \bigg|_{z=0}. \tag{7}$$

Using equation (5) for n_1 in (7) we get $m\beta^2\gamma = 2\pi n_0 e^2/(\gamma+\kappa)$, or

$$\gamma(\gamma+\kappa) = \omega_{\mathrm{s}}^2/\beta^2; \qquad \omega_{\mathrm{s}}^2 = 2\pi n_0 e^2/m \equiv \tfrac{1}{2}\omega_{\mathrm{p}}^2. \tag{8}$$

Solving for γ from equation (8) and using it in (6) we get

$$\gamma_\kappa = -\tfrac{1}{2}\kappa + \tfrac{1}{2}\{\kappa^2 + (4\omega_{\mathrm{s}}^2/\beta^2)\}^{\frac{1}{2}}, \qquad \omega_{\mathrm{SP}}^2(\boldsymbol{\kappa}) = \tfrac{1}{4}\{\beta\kappa + (\beta^2\kappa^2 + 4\omega_{\mathrm{s}}^2)^{\frac{1}{2}}\}^2. \tag{9a, b}$$

This result for the surface plasmon frequency was obtained by Ritchie and Marusak (1966).

For ripples with small amplitude an appropriate form for n_1 is obtained by assuming that the disturbed density $n(r)$ of the electrons has the form

$$n(\mathbf{r},t) = n_0\,\theta\{s(\boldsymbol{\rho},t)-z\} \approx n_0\,\theta(-z) + n_0\,\delta(z)\,s(\boldsymbol{\rho},t),$$

where $s(\boldsymbol{\rho},t)$ is the transverse displacement of the surface as a function of the surface coordinate $\boldsymbol{\rho}$. Then we have

$$n_1(\mathbf{r},t) = n_0\,\delta(z)\,s(\boldsymbol{\rho},t). \tag{10}$$

The boundary condition is

$$v_z\,|_{z=0} = \partial s/\partial t. \tag{11}$$

With the choice

$$s(\boldsymbol{\rho},t) \propto \exp[\mathrm{i}\{\boldsymbol{\kappa}.\boldsymbol{\rho} - \omega_{\mathrm{R}}(\boldsymbol{\kappa})t\}], \tag{12}$$

equation (2) becomes

$$mn_0\,\omega_{\mathrm{R}}^2(\boldsymbol{\kappa}) = n_0^2\,e^2\frac{\partial}{\partial z}\int\frac{\delta(z')\exp\{\mathrm{i}\,\boldsymbol{\kappa}.(\boldsymbol{\rho}'-\boldsymbol{\rho})\}\,\mathrm{d}^2p'\,\mathrm{d}z'}{|\mathbf{r}-\mathbf{r}'|}\Bigg|_{z=0} + m\beta^2 n_0\,\delta'(z)$$

$$= 2\pi n_0^2\,e^2$$

or

$$\omega_{\mathrm{R}}^2(\boldsymbol{\kappa}) = \omega_{\mathrm{s}}^2. \tag{13}$$

Here the integration over z' includes the δ function at the surface. Note that the pressure term drops out because of the occurrence of $\delta'(z)$.

We thus arrive at the conclusion that the frequencies of the ripples on the jellium surface with a step-function equilibrium density profile do not show any wave number dependence. In fact the ripplon dispersion formula becomes identical with that for the surface plasmons in the non-dispersive ($\beta = 0$) limit. This coincidence is basically a consequence of the analytic properties of the Coulomb potential. It may be noted that the boundary conditions used in deriving the surface plasmon and the ripplon dispersion formulae are quite different.

In the analysis presented here for the ripplons, the electron gas is treated as incompressible. In fact it is possible, for a general fluid system, to obtain a dispersion formula for the ripples in a series of powers of the compressibility—a good approximation for small compressibility (Jewsbury and Mahanty 1979; Ogale *et al.* 1982). Since the compressibility of a high density electron gas is small, compressibility corrections will not be discussed here.

3. Force on an External Charge

The charge response properties of the jellium surface are best discussed in terms of the induced potential and the corresponding force experienced by an external charged particle. In view of the result of the previous section one would expect to get the induced potential and force from the known results for surface plasmons in the non-dispersive ($\beta \to 0$) limit, although this approach would mask the essential difference in the boundary conditions in the two cases. A more direct approach using ripplons all through is just as straight forward.

Consider a point charge at $\boldsymbol{r}_0 \equiv (\boldsymbol{\rho}_0, z_0)$ outside the metal surface, with its magnitude given by $Q\exp(-\mathrm{i}\,\omega t)$. Equation (2) is then modified to the form

$$mn_0\,\partial\boldsymbol{v}/\partial t = -e\,n_0(-\nabla\phi)-m\beta^2\nabla n_1\,, \tag{14}$$

where

$$\phi = -e\int\frac{n_1(\boldsymbol{r}')\,\mathrm{d}^3r'}{|\boldsymbol{r}-\boldsymbol{r}'|}+\phi_{\mathrm{ext}}\,, \tag{15}$$

with $\phi_{\mathrm{ext}}(\boldsymbol{r},t) = Q\exp(-\mathrm{i}\,\omega t)/|\boldsymbol{r}-\boldsymbol{r}_0|$. Using equation (10) for n_1 corresponding to only ripple-type surface disturbances we get

$$\begin{aligned}\phi &= -en_0\int\frac{s(\boldsymbol{\rho}')\,\mathrm{d}^2\rho'}{|\boldsymbol{r}-\boldsymbol{\rho}'|}+\frac{Q\exp(-\mathrm{i}\,\omega t)}{|\boldsymbol{r}-\boldsymbol{r}_0|}\\ &= -2\pi en_0\int\exp(\mathrm{i}\,\boldsymbol{\kappa}\,.\,\boldsymbol{\rho}-\kappa|z|)\,s(\boldsymbol{\kappa})\frac{\mathrm{d}^2\kappa}{\kappa}+\frac{Q\exp(-\mathrm{i}\,\omega t)}{|\boldsymbol{r}-\boldsymbol{r}_0|}.\end{aligned} \tag{16}$$

Here $s(\boldsymbol{\kappa})$ is the two-dimensional Fourier transform of $s(\boldsymbol{\rho})$,

$$s(\boldsymbol{\kappa}) = \int\frac{\mathrm{d}^2\rho}{(2\pi)^2}\exp(-\mathrm{i}\,\boldsymbol{\kappa}\,.\,\boldsymbol{\rho})\,s(\boldsymbol{\rho})\,. \tag{17}$$

Substituting equation (16) in (14) and using the boundary condition (11), we get

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}t^2}+\omega_{\mathrm{s}}^2\right)s = \frac{Qe}{m}\frac{z_0}{\{(\boldsymbol{\rho}-\boldsymbol{\rho}_0)^2+z_0^2\}^{3/2}}\exp(-\mathrm{i}\,\omega t)\,,$$

or

$$s(\boldsymbol{\rho},t) = \frac{Qe}{m}\frac{z_0}{\{(\boldsymbol{\rho}-\boldsymbol{\rho}_0)^2+z_0^2\}^{3/2}}\frac{\exp(-\mathrm{i}\,\omega t)}{\omega_{\mathrm{s}}^2-\omega^2}\,. \tag{18}$$

Hence the induced potential $\phi_{\mathrm{I}}(\boldsymbol{r},t)$, which is the first term in equation (16), is given by

$$\begin{aligned}\phi_{\mathrm{I}}(\boldsymbol{r},t) &= -en_0\int\frac{s(\boldsymbol{\rho}')\,\mathrm{d}^2\rho'}{|\boldsymbol{r}-\boldsymbol{\rho}'|}\\ &= -Q\frac{\omega_{\mathrm{s}}^2}{\omega_{\mathrm{s}}^2-\omega^2}\frac{\exp(-\mathrm{i}\,\omega t)}{\{(\boldsymbol{\rho}-\boldsymbol{\rho}_0)^2+(|z|+|z_0|)^2\}^{1/2}}\,.\end{aligned} \tag{19}$$

In the static limit ($\omega\to 0$) this reduces to the image charge result, and for finite ω the strength of the image charge is modified in the expected way.

This result may be compared with the expression for the surface plasmon response (Mahanty and Paranjape 1977)

$$\begin{aligned}\phi_{\mathrm{I}}(\boldsymbol{r}) = -\frac{Q\exp(-\mathrm{i}\,\omega t)}{2\pi}\int\frac{\mathrm{d}^2\kappa}{\kappa}\exp\{\mathrm{i}\,\boldsymbol{\kappa}\,.\,(\boldsymbol{\rho}-\boldsymbol{\rho}_0)-\kappa(|z|+|z_0|)\}\\ \times\frac{\omega_s^2}{\beta^2\gamma_\omega(\kappa+\gamma_\omega)-\omega_{\mathrm{s}}^2}\,,\end{aligned} \tag{20}$$

with $\gamma_\omega = \kappa^2+(\omega_{\mathrm{p}}^2-\omega^2)/\beta^2$. This reduces to (19) in the non-dispersive ($\beta\to 0$) limit.

For a charge Q moving with velocity $\boldsymbol{v}$ in the electrostatic or non-retarded approximation we have

$$\phi_{\text{ext}}(\boldsymbol{r}, t) = \frac{Q}{|\boldsymbol{r}-\boldsymbol{r}_0+\boldsymbol{v}t|} = \frac{Q}{2\pi^2}\int \frac{\mathrm{d}^3k}{k^2}\exp\{\mathrm{i}\boldsymbol{k}.(\boldsymbol{r}-\boldsymbol{r}_0+\boldsymbol{v}t)\}\,. \tag{21}$$

Proceeding as before we get

$$s(\boldsymbol{\rho}, t) = \frac{Qe}{m}\frac{1}{2\pi^2}\int \frac{\mathrm{d}^3k}{k^2}\mathrm{i}\,k_3\exp\{\mathrm{i}\boldsymbol{\kappa}.(\boldsymbol{\rho}-\boldsymbol{\rho}_0)-\mathrm{i}\,k_3 z_0\} \times \exp\{\mathrm{i}(\boldsymbol{k}.\boldsymbol{v})t\}/\{\omega_{\mathrm{s}}^2-(\boldsymbol{k}.\boldsymbol{v})^2\}\,, \tag{22}$$

$$\phi_{\mathrm{I}}(\boldsymbol{r},t) = -\frac{Qe^2n_0}{m}\frac{1}{\pi}\int \frac{\mathrm{d}^3k}{k^2}\mathrm{i}\,k_3\frac{\exp\{\mathrm{i}\boldsymbol{k}.(\boldsymbol{r}-\boldsymbol{r}_0+\boldsymbol{v}t)\}}{\omega_{\mathrm{s}}^2-(\boldsymbol{k}.\boldsymbol{v})^2} \times\{\exp(-\kappa|z|-\mathrm{i}\,k_3 z)\}/\kappa\,. \tag{23}$$

The force on the moving charge is then (for $\boldsymbol{r}\to\boldsymbol{r}_0-\boldsymbol{v}t$)

$$\boldsymbol{F} = Q(-\nabla\phi_{\mathrm{I}}) = -\frac{Q^2e^2n_0}{m}\frac{1}{\pi}\int\frac{\mathrm{d}^3k\;k_3\,\boldsymbol{k}\exp(-\kappa|z|-\mathrm{i}\,k_3 z)}{\kappa k^2\{\omega_{\mathrm{s}}^2-(\boldsymbol{k}.\boldsymbol{v})^2\}}\,. \tag{24}$$

The integral can be done and the resulting functions are complicated. In the case when v_3 vanishes, i.e. the motion of the charged particle is parallel to the jellium surface, we get the results obtained earlier by Muscat and Newns (1977) from the non-dispersive surface plasmon response

$$\begin{aligned} F_{\|}(z_0) &= -\frac{Q^2e^2n_0}{m}\frac{1}{\pi}\int\frac{\mathrm{d}^2\kappa\;\mathrm{d}k_3\;k_3\,\boldsymbol{\kappa}\exp(-\kappa|z_0|-\mathrm{i}\,k_3 z_0)}{\kappa(\kappa^2+k_3^2)\{\omega_{\mathrm{s}}^2-(\boldsymbol{\kappa}.\boldsymbol{v})^2\}} \\ &= -(Q^2/4z_0)(2\kappa_{\mathrm{c}}z_0)^2\mathrm{K}_0(2\kappa_{\mathrm{c}}z_0)\,, \end{aligned} \tag{25}$$

$$\begin{aligned} F_{\perp}(z_0) &= -\frac{Q^2e^2n_0}{m}\frac{1}{\pi}\int\frac{\mathrm{d}^2\kappa\;\mathrm{d}k_3\;k_3^2\exp(-\kappa|z_0|-\mathrm{i}\,k_3 z_0)}{\kappa(\kappa^2+k_3^2)\{\omega_{\mathrm{s}}^2-(\boldsymbol{\kappa}.\boldsymbol{v})^2\}} \\ &= -Q^2\kappa_{\mathrm{c}}^2[1+\tfrac{1}{2}\pi\{\mathrm{L}_1(2\kappa_{\mathrm{c}}z_0)-\mathrm{I}_1(2\kappa_{\mathrm{c}}z_0)\}]\,. \end{aligned} \tag{26}$$

where L_1 is the Struve function, K_0 and I_1 are the modified Bessel functions, and $\kappa_{\mathrm{c}} = \omega_{\mathrm{s}}/v$. Here $F_{\perp}(z_0)$ goes to the image force limit $-Q^2/4z_0^2$ for $v\to 0$ or $\kappa_{\mathrm{c}}\to\infty$, and goes to zero for $v\to\infty$. The damping force $F_{\|}(z_0)$ as a function of v has a maximum for $2\kappa_{\mathrm{c}}z_0 \approx 1{\cdot}6$, or for $v\approx 1{\cdot}25\,\omega_{\mathrm{s}}z_0$, and the value at maximum is $0{\cdot}481$ times the image force.

4. Conclusions

It is shown here that the charge response behaviour of the jellium surface due to ripples on it leads to the image force on a stationary external charge, and a damping force on a moving external charge. The expression for the force is identical with what one gets by considering the response due to the surface plasmons in the non-dispersive limit, although the boundary conditions in the two situations are radically different.

Since the free surface of the jellium electron gas is likely to respond through ripple-like displacements more easily than through the rigid boundary condition corresponding to surface plasmons, it would perhaps be appropriate to regard the image potential and associated effects as arising out of the ripplon modes.

As seen in equation (19), the image of a stationary point charge formed due to ripplon response is a point image, so that there will be a divergence in the self-energy of the charge as $z_0 \to 0$. This, of course, is a spurious divergence since for very small values of z_0 an upper cut-off must be used in the κ integrals that arise through the Fourier transforms (such as in equation 16), beyond which the continuum hydrodynamic model is not valid. The image due to the response through surface plasmons has a finite extension for non-zero β, so that the self-energy does not diverge for $z_0 \to 0$ even without any cut-off in κ space.

A complete analysis of the charge oscillation modes of the jellium surface must take into consideration the equilibrium density profile $n_0(z)$ which is not a step function. This has been done in detail for the surface plasmon modes (Equiluz *et al.* 1975, and references cited therein). In principle, the solution of equation (2) with the appropriate density profile $n_0(z)$ will give the density oscillation modes. The method of solution indicated in Section 2 is based on specific assumptions on the form of n_1 for the surface plasmon and the ripplon modes—the actual modes may be expected to be a mixture of both. For the electron gas in jellium, however, the frequencies of surface plasmons and ripplons at small wave numbers are essentially the same and so are the associated density fluctuations. Hence, either of them could be used to work out the surface response due to an external charge, particularly at larger values of its distance from the surface.

An analysis of ripplon dispersion including a realistic equilibrium density profile can be made by assuming that the entire surface profile undergoes transverse displacements, that is,

$$n_0(\boldsymbol{r}) = n_0\, T(-z), \tag{27}$$

where $T(-z)$ is a function that falls to zero from unity rapidly across the boundary, and $z = 0$ corresponds to the point at which $T''(-z)$ vanishes. The above form of the ripple implies

$$n(\boldsymbol{r}) = n_0\, T\{s(\boldsymbol{\rho}, t) - z\} \approx n_0\, T(-z) + n_0\, s(\boldsymbol{\rho}, t)\, T'(-z) + \dots,$$

so that

$$n_1 = n_0\, T'(-z)\, s(\boldsymbol{\rho}, t). \tag{28}$$

Equation (2) along with the boundary condition (11) leads to

$$m n_0\, \omega_{\mathrm{R}}^2(\boldsymbol{\kappa}) = n_0^2 e^2 \frac{\partial}{\partial z} \int \frac{T'(-z') \exp\{\mathrm{i}\boldsymbol{\kappa} . (\boldsymbol{\rho}' - \boldsymbol{\rho})\}\, \mathrm{d}^2\rho'\, \mathrm{d}z'}{|\boldsymbol{r} - \boldsymbol{r}'|} \bigg|_{z=0}$$

$$+ m\beta^2 n_0\, T''(-z)\,|_{z=0},$$

or

$$\omega_{\mathrm{R}}^2(\boldsymbol{\kappa}) = \omega_{\mathrm{s}}^2 \int_{-\infty}^{\infty} T'(z') \exp(-\kappa |z'|)\, \mathrm{d}z'. \tag{29}$$

We obviously have $\omega_{\mathrm{R}}^2(\boldsymbol{\kappa}) \to \omega_{\mathrm{s}}^2$ for $\kappa \to 0$, while for higher values of κ the value of $\omega_{\mathrm{R}}^2(\boldsymbol{\kappa})$ diminishes. Although $\omega_{\mathrm{R}}^2(\boldsymbol{\kappa})$ for these ripples appears to diminish to zero for $\kappa \to \infty$, there would be a practical upper limit for κ beyond which, as pointed out above, the hydrodynamic model breaks down. Since $T'(-z)$ is almost a δ function, the previous analysis is useful as a semi-quantitative description of phenomena associated with ripples.

When the external charged particle is an electron, then for sufficiently large z_0 the above theory is valid. But for small z_0 exchange corrections become important—these corrections would reduce its self-energy from its value when exchange is neglected. Incorporation of this correction in the hydrodynamic model is difficult.

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