The Internal Structure of Plasmons

I. Egri
Institut für Theoretische Physik RWTH Aachen, Federal Republic of Germany

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The concept of an internal wave function for the plasmon is introduced and discussed. Explicit results are presented for two and three dimensions in the degenerate and nondegenerate limit, respectively. The reciprocal Fermi wavevector $K_F^{-1}$ and the thermal wavelength $K_r^{-1}$ are found to define the length scale and hence the size of the plasmon. A possible surface anomaly is indicated.

0. Introduction

The plasmon is a collective excitation of a system of electrons. As such, it is a coherent superposition of electron-hole pairs, corresponding to electronic transitions from occupied to unoccupied states. The expansion coefficients, which describe the contribution of individual electron-hole pairs, give rise to an internal structure of the plasmon. This is in complete analogy to the exciton problem, as has already been noted by Ferrell [1], but is usually overlooked in the conventional treatment of the plasmon. Apart from the intrinsic interest in this problem, there now seems to be experimental evidence for the finite size of the plasmon [2].

In view of this fact, it may be useful to present a comprehensive study of the plasmon's wavefunction. This is the purpose of the present paper. We first establish the connection of our method with the usual derivation of the plasmon's frequency. Then, we calculate explicitly the plasmon's wavefunction in two and three dimensions, in the degenerate and non-degenerate limit, respectively. Finally, qualitative arguments are given how the internal structure of the plasmon may lead to the observed surface anomaly [2]. An attempt to incorporate this effect into the interpretation of energy loss measurements quantitatively, has been made recently by Stahl [3].

1. Theory

Conventionally, the plasmon is defined along either of the two following lines. On the one hand, Maxwell's equations imply for any longitudinal eigenmode of the system that the dielectric function vanishes at the corresponding frequency:

$$\varepsilon(q, \omega(q)) = 0. \quad (1.1)$$

For an electron gas, Eq. (1.1) defines the plasma frequency $\omega_p(q)$. On the other hand, one may consider the equation of motion for a density wave in the electron gas and obtain

$$\frac{d^2}{dt^2} \rho_q = -(\omega_p(q))^2 \rho_q. \quad (1.2)$$

In certain approximations, the two definitions yield identical results for the plasmon frequency.

The second definition can be carried over into a microscopic quantum-mechanical treatment, if $\rho_q$ is regarded as an operator and the time derivative is replaced by the commutator with the Hamiltonian $\mathcal{H}$. Equation (1.2) then reads

$$[[\rho_q, \mathcal{H}], \mathcal{H}] = (\hbar \omega_p(q))^2 \rho_q. \quad (1.3)$$

In what follows, we will also use an equation of motion approach, similar to (1.3) but there is one important difference: we distinguish between the operator for a density wave $\rho_q$ and the operator for a plasmon, $S_q$. As will be seen later, $\rho_q = S_q$ holds in the limit of vanishing $q$ only. Making this distinction, we need not take the second time derivative but can use the following definition

$$i \hbar \dot{S}_q = [S_q, \mathcal{H}] = \hbar \omega_p(q) S_q. \quad (1.4)$$
This is the familiar definition of an elementary excitation \[4\]. It constitutes an eigenvalue equation for the plasmon operator \( S_\mathbf{q} \) and the plasma frequency \( \omega_\mathbf{p}(\mathbf{q}) \).

Once \( S_\mathbf{q} \) is known, we obtain the plasmon state by
\[
|\Psi_\mathbf{p}(\mathbf{q})\rangle = S_\mathbf{q}^* |\Psi_\mathbf{0}\rangle
\]
where \( |\Psi_\mathbf{0}\rangle \) is a suitable ground state, and the plasmon wavefunction is given, by definition, as
\[
f_\mathbf{q}(r_e, r_h) = \langle \Psi(r_e, r_h) | \Psi_\mathbf{p}(\mathbf{q}) \rangle.
\]

Here
\[
|\Psi_\mathbf{p}(\mathbf{q})\rangle = \sum_{r_e, r_h} f_\mathbf{q}(r_e, r_h) |\Psi_\mathbf{0}\rangle
\]
is a state, where an electron-hole pair situated at \( r_e \) and \( r_h \), respectively, has been created from the ground state. Therefore, \( f_\mathbf{q}(r_e, r_h) \) is the probability amplitude of finding an electron at \( r_e \) and a hole at \( r_h \), provided the system is in the plasmon state. This definition is identical to the one given earlier \[2\]:
\[
|\Psi_\mathbf{p}(\mathbf{q})\rangle = \sum_{r_e, r_h} f_\mathbf{q}(r_e, r_h) |\Psi_\mathbf{0}\rangle
\]
which shows that the plasmon state is a coherent linear combination of electron-hole pairs.

It is the function \( f_\mathbf{q}(r_e, r_h) \) which determines the internal structure of the plasmon. Its calculation is the purpose of the present paper.

We start by expanding the field operators in (1.7) into plane wave states:
\[
\psi(r) = (\Omega)^{-1/2} \int d^3k e^{i\mathbf{k}\cdot r} c_k.
\]

Here, \( \Omega \) is the volume of the system. In this basis, the Hamiltonian is given by
\[
\mathcal{H} = \sum_k \hbar^2 K^2 2m c_k^+ c_k + \frac{1}{2} \sum_q V_q \rho_q \rho_q - q
\]
with
\[
V_q = \frac{\epsilon^2}{\Omega \epsilon_0 q^2}
\]
and \( \rho_q \) is the density
\[
\rho_q = \sum_k c_k^+ c_{k+q} = \sum_k S_{q}(k).
\]

In the last line, we have introduced the electron-hole-pair operator \( S_q(k) \). In contrast to papers relating to the high density electron gas \[5, 6\], we do not introduce separate operators for electrons below and above \( K_F \) and keep the Hamiltonian state-independent.

This is necessary because we want to treat the non-degenerate case as well. Hence, the sum in (1.12) runs over all \( k \). The plasmon operator is now defined as:
\[
S_q = \sum_k \sigma_q(k) S_q(0)(k)
\]
where \( \sigma_q(k) \) is to be determined by the condition (1.4).

The commutator \( [S_q, \mathcal{H}] \) is straightforward to obtain
\[
[S_q, \mathcal{H}] = \sum_k \left( \sigma_q(k) \frac{\hbar^2}{2m} [(k+q)^2 - K^2] S_q(k) \right)
\]
\[
+ \frac{1}{2} \sum_{k', q'} V_q \sigma_q(k) \{ S_q(k') c_{k+q} - c_{k+q}^+ c_{k+q} \}
\]
where \( \{A, B\} = AB + BA \).

We see that the definition (1.4) cannot be satisfied exactly, due to the presence of the operators \( c_k^+ c_k \) in (1.14). This is in accordance with the approximate nature of the concept of elementary excitations and we linearize (1.14) by applying the random phase approximation: we replace \( c_k^+ c_k \) by its average:
\[
c_k^+ c_k \rightarrow \langle c_k^+ c_k \rangle = f(k) \delta_{k,k'}
\]
where \( f(k) \) is the Fermi function.

Inserting (1.15) into (1.14) and employing the definition (1.4) we arrive at an integral equation for \( \sigma_q(k) \):
\[
\sigma_q(k) = \frac{V_q}{\hbar \omega_p(q) - \frac{\hbar^2}{2m} [(k+q)^2 - K^2]} \cdot \sum_{k'} (f(k') - f(k' + q)) \sigma_q(k').
\]

Multiplying by \( f(k) - f(k + q) \) and summing over \( k \) we obtain the following equation for \( \omega_p(q) \):
\[
1 = \sum_k \frac{f(k) - f(k + q)}{\hbar \omega_p(q) - \frac{\hbar^2}{2m} [(k + q)^2 - K^2]}
\]
This is the famous RPA result for the plasmon frequency of an electron gas. It has been discussed extensively in the literature \[5, 7\]. It yields the same result for the frequency as (1.1), if Lindhard's dielectric function \[8\] is used. The integral Eq. (1.16) is solved by
\[
\sigma_q(k) = \frac{h \omega_p(0)}{h \omega_p(q) - \frac{\hbar^2}{2m} [(k + q)^2 - K^2]}
\]