

Double plasmon excitation in an electron gas

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A many-body perturbation-theoretic calculation of the double-plasmon excitation in an electron gas is presented, using Feynman diagrams. The low- q limit of the inverse mean free path for this second order process is evaluated in the random-phase-approximation, and it is compared to previous theoretical and experimental estimations.

It is well known that electron-energy-loss measurements performed in transmission experiments [1] have been successfully explained in terms of plasmons, i.e., collective excitations of valence electrons of the target with an energy $\hbar\omega_p$. Experiments show that after passing thin foils electrons suffer not only one loss, but, in general, multiples of $\hbar\omega_p$, depending on the thickness of the specimen. The major contribution to this multiple plasmon excitation comes from the fact that the penetrating electron interacts a number of times with the medium, with a single plasmon excitation per interaction. The probability for multiple excitations has, therefore, usually been described by a Poisson distribution, assuming zero probability for nonlinear multiple plasmon excitation events occurring in a single interaction of the probe electron with the target.

Ashley and Ritchie [2] were the first to calculate the probability for nonlinear double plasmon excitation. They employed a version of the random-phase-approximation (RPA) originated by Pines and Bohm for treatment of collective phenomena, neglected dispersion in the plasmon energy and used second order perturbation theory to show that the probability for double plasmon excitation might be large enough for low electron density metals.

Subsequently, experimental investigations of the single and double plasmon contributions to the $2\hbar\omega_p$ peak in the aluminum loss spectrum were reported [3,4], and the results suggested evidence for the double plasmon process. Since then, experimental as well as theoretical investigations have been performed [5–9]; however, not only theoretical predictions but also experimental results disagree and a full theoretical treatment of the problem is still missing.

In this paper we present a many-body perturbation-theoretic treatment of nonlinear processes leading to double excitations in the electron gas, and derive general expressions for the inverse mean free path for these processes, which involve the linear response function of the medium and, also, the quadratic response function represented by the triple vertex recently studied [10,11] to investigate Z_1^3 corrections to the energy loss of charged particles travelling through matter. In particular, we focus on the double plasmon excitation, consider the low- q limit of the RPA linear and quadratic response functions, evaluate in this limit the inverse mean free path after neglectation of plasmon dispersion, and compare with previous theoretical and experimental estimations.

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We consider an electron gas of density n through which a fast electron of speed $v \gg e^2/\hbar$ passes, and focus on processes in which a single virtual excitation of the electron gas decays into two real subsequent excitations.

The matrix element of the S -matrix corresponding to this process can be represented in momentum space as (we use atomic units throughout, i.e., $e = m = \hbar = 1$):

$$S_{fi} = \text{diagram} = -i\Omega^{-2} V_q V_{q_1} V_{q-q_1} M_{q,q_1}. \quad (1)$$

In this expression $q_1 = (p_1 - s_1, \omega_{p_1} - \omega_{s_1})$, $q - q_1 = (p_2 - s_2, \omega_{p_2} - \omega_{s_2})$, $\omega_p = p^2/2$ is the energy of an electron with momentum p , and it is understood that both (p_1, s_1) and (p_2, s_2) lie in the (unoccupied, occupied) part of momentum space. Ω is the normalization volume, V_q , represented by wavy lines, gives account of the effective interaction [12], i.e.,

$$V_q = \frac{4\pi}{q^2} \epsilon_q^{-1}, \quad (2)$$

ϵ_q being the linear response function of the medium, that is,

$$\epsilon_q = 1 - \frac{4\pi}{q^2} A_q, \quad (3)$$

A_q , represented by a double vertex, is a sum over all proper polarization subdiagrams, and M_{q,q_1} , represented by the triple vertex diagram, is the quadratic response function. In the well-known random-phase-approximation (RPA), A_q and M_{q,q_1} are approximated by the empty bubble [14,15] and the empty triangle diagram [10,13], respectively.

The transition rate for a double excitation of the electron gas is, then, given by the following expression:

$$\gamma = \sum_{p_1} \sum_{p_2} \sum_{s_1} \sum_{s_2} \frac{|S_{fi}|^2}{T}, \quad (4)$$

T being the interaction time, and after introduction of Eq. (1) into Eq. (4) we find for the inverse mean free path

$$\lambda^{-1} = \frac{1}{v} \lim_{T \rightarrow \infty} \gamma = \frac{2^7}{(2\pi)^3 v} \int d^4 q \int d^4 q_1 \left[\text{Im}(-\epsilon_{q_1})^{-1} \text{Im}(-\epsilon_{q-q_1})^{-1} \frac{|\epsilon_q|^{-2} \frac{1}{2} |M_{q,q_1}^s|^2}{q^4 q_1^2 |q - q_1|^2} \delta(q^0 - \frac{1}{2} q_0^2 + \frac{1}{2} q_f^2) \right], \quad (5)$$

where $M_{q,q-q_1}^s$ is the symmetrized quadratic response function:

$$M_{q,q_1}^s = \frac{1}{2} (M_{q,q_1} + M_{q,q-q_1}), \quad (6)$$

and q_0 and q_f represent the initial and final momentum of the penetrating electron ($q = q_0 - q_f$).

In particular, the contribution to the inverse mean free path coming from the excitation of a double plasmon can be obtained from the following expression:

$$\lambda_{2p}^{-1} = \frac{2^8 \pi^2}{(2\pi)^3 v} \int d\mathbf{q} \int d\mathbf{q}_1 f(\mathbf{q}, \mathbf{q}_1) \theta(q_c - |\mathbf{q}_1|) \theta(q_c - |\mathbf{q}_2|) \delta(2\omega_{q_1} + 2\omega_{q_2} - q_0^2 + q_f^2), \quad (7)$$

where

$$f(\mathbf{q}, \mathbf{q}_1) = \left| \frac{\partial \text{Re}(\epsilon_{q_1, \omega_1})}{\partial \omega_1} \right|_{\omega_1=\omega_{q_1}}^{-1} \left| \frac{\partial \text{Re}(\epsilon_{q_2, \omega_2})}{\partial \omega_2} \right|_{\omega_2=\omega_{q_2}}^{-1} \frac{|\epsilon_{q, \omega_{q_1} + \omega_{q_2}}|^{-2} |M_{q, \omega_{q_1} + \omega_{q_2}; q_1, \omega_{q_1}}^s|^2}{q^4 q_1^2 |q - q_1|^2}. \quad (8)$$

In these equations we have defined $q_2 = q - q_1$, $\theta(x)$ represents the Heaviside function, q_c is the critical wave vector where the plasmon dispersion enters the electron-hole pair excitation region, and ω_q is given by the dispersion relation:

$$\omega_q^2 \approx \omega_0^2 + \frac{3}{5} q_F^2 q^2 + \left(\frac{1}{4} + \frac{3}{7} \frac{q_F^4}{\omega_0^2}\right) q^4 + O(q^6), \quad (9)$$

ω_0 and q_F being the plasma frequency and the Fermi momentum of the electron gas, respectively.

The integrals of Eq. (7) can be simplified if one assumes that dispersion can be neglected in the plasmon energies ω_{q_1} and ω_{q_2} inside the delta function. Then, both $|q_0|$ and $|q_f|$ are fixed, $q \, dq = q_0 q_f \sin \theta \, d\theta$, θ being the angle between q_0 and q_f , and one can write

$$\gamma_{2p} = \frac{2^7 \pi^2}{(2\pi)^2 v} (F_1 - F_2), \quad (10)$$

where we have followed ref. [2] in defining

$$F_1 = \left[\int_{q_{\min}}^{q_c} \frac{dq}{q} \int_0^{q_c} dq_1 + \int_{q_c}^{2q_c} \frac{dq}{q} \int_{q-q_c}^{q_c} dq_1 \right] f_1(q, q_1), \quad (11)$$

and

$$F_2 = \left[\int_{q_{\min}}^{q_c} \frac{dq}{q} \int_{q_c-q}^{q_c} dq_1 + \int_{q_c}^{2q_c} \frac{dq}{q} \int_{q-q_c}^{q_c} dq_1 \right] f_2(q, q_1), \quad (12)$$

with

$$f_1(q, q_1) = 2\pi \int_{-1}^1 d\mu \left| \frac{\text{Re}(\epsilon_{q_1, \omega_1})}{\omega_1} \right|_{\omega_1=\omega_{q_1}}^{-1} \left| \frac{\text{Re}(\epsilon_{q_2, \omega_2})}{\omega_2} \right|_{\omega_2=\omega_{q_2}}^{-1} \frac{|\epsilon_{q, \omega_{q_1}+\omega_{q_2}}|^{-2} |M_{q, \omega_{q_1}+\omega_{q_2}; q_1, \omega_{q_1}}^s|^2}{q^2 |q - q_1|^2}, \quad (13)$$

$$f_2(q, q_1) = 2\pi \int_{-1}^{\alpha} d\mu \left| \frac{\text{Re}(\epsilon_{q_1, \omega_1})}{\omega_1} \right|_{\omega_1=\omega_{q_1}}^{-1} \left| \frac{\text{Re}(\epsilon_{q_2, \omega_2})}{\omega_2} \right|_{\omega_2=\omega_{q_2}}^{-1} \frac{|\epsilon_{q, \omega_{q_1}+\omega_{q_2}}|^{-2} |M_{q, \omega_{q_1}+\omega_{q_2}; q_1, \omega_{q_1}}^s|^2}{q^2 |q - q_1|^2}, \quad (14)$$

and

$$q_{\min} = v - \sqrt{v^2 - 4\omega_0}, \quad (15)$$

μ and α being

$$\mu = \cos(q, q_1) \quad (16)$$

and

$$\alpha = \frac{q^2 + q_1^2 - q_c^2}{2qq_1}, \quad (17)$$

respectively.

Plasmons can only be excited for a momentum transfer smaller than q_c , so that linear and quadratic RPA response functions might be well approximated by their low- q limit, to give after an expansion in powers of $|q|$ and $|q_1|$:

$$\epsilon_{q, \omega} = 1 - \frac{\omega_0^2}{\omega^2} (1 + O[q^2]) \quad (18)$$

and

inction, q_c is the
ation region, and

$$M_{q,q_1}^s = -\frac{n}{4} \left[\frac{q^4}{\omega_1(\omega - \omega_1)\omega^2} - \frac{q_1^4/\omega_1 + |q - q_1|^4/(\omega - \omega_1)}{\omega_1(\omega - \omega_1)\omega} \right. \\ \left. + \frac{q^2(q_1^2/\omega_1^2 + |q - q_1|^2/(\omega - \omega_1)^2)}{\omega^2} + \frac{q_1^2|q - q_1|^2}{\omega_1^2(\omega - \omega_1)^2} \right]. \quad (19)$$

In addition, one can approximate ω_q of Eq. (9) by ω_0 and find

$$\gamma_{2p} = \frac{1}{2^4 3^2 \pi^2 n q_0} (F_1 - F_2), \quad (20)$$

where F_1 and F_2 are given by Eqs. (11) and (12) with

$$f_1(q, q_1) = [2q_1^2 - \frac{1}{2}q(q^2 + q_1^2)] - \left[\frac{(q^2 + q_1^2)^2}{4q_1} - q^2 q_1 \right] \ln \left| \frac{q - q_1}{q + q_1} \right| \quad (21)$$

and

$$f_2(q, q_1) = q[q_1^2 - \frac{1}{4}(q^2 + q_1^2)] \left[\frac{q^2 + q_1^2 - 1}{2qq_1} + 1 \right] + [8q_1^3 - \frac{1}{4}q^2 q_1] \left[\left(\frac{q^2 + q_1^2 - 1}{2qq_1} \right)^2 - 1 \right] \\ - 4q_1^3 \left[\left(\frac{q^2 + q_1^2 - 1}{2qq_1} \right)^4 - 1 \right] + \left[\frac{(q^2 + q_1^2)^2}{4q_1} - q^2 q_1 \right] \ln \left(\frac{q + q_1}{q_c} \right). \quad (22)$$

Finally, assuming that the velocity of the incident electron is large enough as compared to the velocity of target electrons, $q_{\min}$ of Eq. (15) can be approximated by $2\omega_0/v$, the integrals involved in Eqs. (11) and (12) may be performed analytically under the assumption that the quantity $\gamma = q_{\min}/q_c$ is a small quantity, and we find

$$\lambda_{2p}^{-1} = \frac{q_c^5}{36\pi\omega_0^2 v^2} [0.963555 - \frac{7}{5}\gamma^2 + O(\gamma^3)]. \quad (23)$$

On the other hand, it is easy to show under the same assumptions that the contribution to the S -matrix corresponding to the process in which a single excitation of the electron gas is produced leads to the well-known result for the inverse mean free path for the single plasmon emission:

$$\lambda_p^{-1} = \frac{\omega_0}{v^2} \ln \frac{v q_c}{\omega_0}, \quad (24)$$

and we find, therefore, the following result for the double plasmon loss relative to the single plasmon loss:

$$\frac{\lambda_{2p}^{-1}}{\lambda_p^{-1}} \sim \frac{q_c^5}{36\pi\omega_0^3} \frac{0.96}{\ln(v q_c/\omega_0)} = 9.78 \times 10^{-4} r_s^2 \left(\ln \frac{2}{\gamma} \right)^{-1}, \quad (25)$$

where we have assumed $q_c = \omega_0/q_F$ and $\gamma = 2q_F/v$, and r_s represents the radius of the sphere that contains on average one valence electron. For aluminum ($r_s = 2.07$) and an incident electron energy of 40 keV, we find $\lambda_{2p}^{-1}/\lambda_p^{-1} \sim 8.78 \times 10^{-4}$.

The intensity of the double plasmon measured relative to the single plasmon intensity in aluminum after bombardment of 60 keV electrons was found by Missell and Atkins [3] to be less than 0.02, Spence and Spargo [5] reported a value of 0.03 in their investigation and Batson and Silcox [8] found a value of 0.07, in approximate agreement with the calculations developed by Ashley and Ritchie [2], who found for aluminum a relative intensity of 0.04, and by Srivastava et al. [7], who found a value of 0.02. However, other experiments seem to indicate the excitation of two plasmons in one event as less probable [6,9]; in particular, most recent experiments performed by Schattschneider et al. [9] with 40 keV incident electrons impose an upper limit of 5.0×10^{-3} onto the relative probability of double plasmon excitation, which is much smaller than the former theoretical predictions but is not in contradiction with our preliminary result.

In conclusion, a general expression for the double plasmon creation probability which can be used for non-zero momentum transfer has been devised, following procedures of many body perturbation

theory, this expression has been evaluated in the low- q and high-velocity limits of the RPA, and a preliminary result has been obtained that is much lower than previous theoretical predictions though it is consistent with most recent experimental evidence. In future work we shall investigate the reliability of the low- q limit approximation for the linear and quadratic response functions of the electron gas, and a detailed comparison with previous theoretical treatments will also be made; work in this direction is now in progress.

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