

Coherent double-plasmon excitation in solids

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Abstract

Coherent double-plasmon mean free paths of swift electrons passing through an electron gas are studied, following a many-body perturbation-theoretic analysis. Both linear and quadratic response functions are evaluated in the full random-phase-approximation, and theoretical predictions for this second-order process are given as a function of the projectile velocity. The validity of previously studied analytical estimations is discussed, and comparison with experiments is made.

1. Introduction

Energy-loss spectra of fast electrons in transmission experiments have been successfully explained in terms of plasmons [1], quanta of collective excitations of conduction electrons in the target with an energy $\hbar\omega_p$. Experimental data show multiple-plasmon peaks, which are mainly attributed to plural single excitations. This multiple single-plasmon excitation is due to the fact that the penetrating electron interacts a number of times with the medium, with a single plasmon excitation per interaction. Thus, the probability for multiple single excitations obeys a Poisson distribution.

Ashley and Ritchie [2] were the first to consider the possibility of coherent double-plasmon excitation, i.e., creation of two plasmons in a single interaction of the probe electron with the target. They calculated the relative probability of this event as compared to the single-plasmon case in a nonlinear version of the RPA originated by Pines and Bohm for treatment of collective phenomena. After this work, experimental investigations performing electron-energy-loss spectra suggested evidence for the double-plasmon process [3,4]. Since then, experimental and theoretical investigations have been performed [5–9], without full agreement.

This work intends to be part of a many-body theoretical investigation on nonlinear contributions to multiple excitations in solids. In particular, in this paper we follow the lines of a previous work reported by Pitarke and Ritchie [10], where, for the first time, a diagrammatic many-body perturbation theoretic analysis was developed to deal with

the double-plasmon problem. In their work, they calculated coherent double-plasmon inverse mean free paths for swift electrons passing through an electron gas by using the low- q limit of the RPA linear and quadratic response functions, thus neglecting plasmon dispersion. In this paper, we derive double-plasmon inverse mean free paths by evaluating linear and quadratic response functions in the full random-phase-approximation (RPA). In this way, further improvement on the investigation of multiple excitations in solids is fulfilled.

2. General approach

We consider, in the jellium model, a metal of density n , through which a swift electron passes, and focus on processes in which a single virtual excitation of the electron gas decays into two real subsequent excitations. A many-body perturbation analysis of this problem, up to first order in the charge of the penetrating particle and to lowest order in the effective interaction, thus neglecting local field corrections, leads to the following expression for the inverse mean free path of a double excitation (we use atomic units throughout, i.e., $e = m = \hbar = 1$) [10]:

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

where v is the probe electron velocity, q is the four momentum transferred to the system by creating a virtual excitation that decays into two real excitations of four momenta q_1 and $q - q_1$, and q_0 and q_f are the initial and

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final momenta of the penetrating electron ($q = q_0 - q_f$). ϵ_q represents the well-known linear RPA response function, ϵ_q^{-1} being represented diagrammatically by an infinite sum of empty double vertices [11,12], and M_{q,q_1}^s , represented diagrammatically by empty triple vertices [13–15], is the symmetrized quadratic response function, as defined in Ref. [16].

In particular, we follow Ref. [10] to derive the contribution to the inverse mean free path coming from the excitation of a double plasmon, and find:

$$\lambda_{2p}^{-1} = \frac{2^7 \pi^2}{(2\pi)^2 v^2} (F_1 + F_2), \quad (2)$$

where F_1 and F_2 are:

$$F_1 = \int_{q_{\min}}^{q_c} \frac{dq}{q} \int_0^{q_c-q} dq_1 f_1(q, q_1) \quad (3)$$

and

$$F_2 = \left[\int_{q_{\min}}^{q_c} \frac{dq}{q} \int_{q_c-q}^{q_c} dq_1 + \int_{q_c}^{2q_c} \frac{dq}{q} \int_{q-q_c}^{q_c} dq_1 \right] \times f_2(q, q_1), \quad (4)$$

with

$$f_1(q, q_1) = 2\pi \int_{-1}^1 d\mu \left| \frac{\partial \text{Re}(\epsilon_{q_1, \omega_1})}{\partial \omega_1} \right|_{\omega_1 = \omega_{q_1}}^{-1} \times \left| \frac{\partial \text{Re}(\epsilon_{q_2, \omega_2})}{\partial \omega_2} \right|_{\omega_2 = \omega_{q_2}}^{-1} \times \frac{|\epsilon_{q, \omega_{q_1} + \omega_{q_2}}|^{-2} |M_{q, \omega_{q_1} + \omega_{q_2}; q_1, \omega_{q_1}}^s|^2}{q^2 |q - q_1|^2}, \quad (5)$$

$$f_2(q, q_1) = 2\pi \int_{\alpha}^1 d\mu \left| \frac{\partial \text{Re}(\epsilon_{q_1, \omega_1})}{\partial \omega_1} \right|_{\omega_1 = \omega_{q_1}}^{-1} \times \left| \frac{\partial \text{Re}(\epsilon_{q_2, \omega_2})}{\partial \omega_2} \right|_{\omega_2 = \omega_{q_2}}^{-1} \times \frac{|\epsilon_{q, \omega_{q_1} + \omega_{q_2}}|^{-2} |M_{q, \omega_{q_1} + \omega_{q_2}; q_1, \omega_{q_1}}^s|^2}{q^2 |q - q_1|^2}, \quad (6)$$

and

$$\alpha = \frac{q^2 + q_1^2 - q_c^2}{2qq_1}. \quad (7)$$

$q_{\min}$ is the lowest allowed momentum for plasmon excitation, and μ is the angle between q and q_1 .

Eqs. (2)–(6) are exactly equivalent to Eqs. (10)–(12) of Ref. [10], though they happen to be more feasible for

numerical computation. Also, the approximate value of $q_{\min}$ given by Eq. (15) of Ref. [10] has been replaced by its exact numerical value in the computations presented in this paper.

3. Results and discussion

Eq. (1) was deduced under the assumptions of the random-phase-approximation. The full RPA result is obtained when the explicit expressions of ϵ_q and M_{q,q_1} are introduced into Eq. (2). The explicit expression for ϵ_q was first obtained by Lindhard [17], and explicit expressions for M_{q,q_1} are also available [16].

For high velocities of the probe electron, the target can be considered to be an electron gas at rest. This is known as the static electron gas model, in which the momenta of all the electrons become equal to zero, and the plasmon frequency can be written as

$$\omega_q^2 = \omega_p^2 + \frac{1}{4} q^4. \quad (8)$$

In the plasmon-pole approximation [18],

$$\omega_q^2 = \omega_p^2 + \frac{3}{5} q^2 q^2 + \frac{1}{4} q^4, \quad (9)$$

and double-plasmon excitation probabilities derived in this approximation are equivalent to the result obtained within a nonlinear quantum hydrodynamical model of the electron gas, as demonstrated in Ref. [19].

In the low- q limit both static electron gas and plasmon-pole approximations to linear and quadratic response functions coincide with the low- q limit of full RPA response functions [16], and the integrations involved in Eqs. (3)–(6) can be performed analytically to find, after some algebra, the following expression [20].

$$\lambda_{2p}^{-1} = \frac{q_c^5}{36\pi\omega_p^2 v^2} \left[\frac{308}{375} + \frac{\gamma}{25} - \frac{221}{450} \gamma^2 + \mathcal{O}(\gamma^3) \right], \quad (10)$$

where $\gamma = q_{\min}/q_c$.

This expression can be simplified if we take $q_c \approx \omega_p/q_F$, and find:

$$\lambda_{2p}^{-1} \sim 0.164 \frac{\sqrt{r_s}}{36\pi v^2}. \quad (11)$$

We have performed the calculation of λ_{2p}^{-1} as a function of the probe electron velocity for aluminium, $r_s = 2.07$, and we have, also, evaluated λ_{1p}^{-1} in the same approximations, in order to compare with λ_{2p}^{-1} , and to obtain the relative magnitude $\lambda_{2p}^{-1}/\lambda_{1p}^{-1}$.

In Fig. 1 λ_{2p}^{-1} is plotted by a solid line in the full RPA, together with results obtained in the static electron gas, plasmon-pole and low- q limit approximations; low- q limit results have been plotted from Eqs. (10) and, also, from

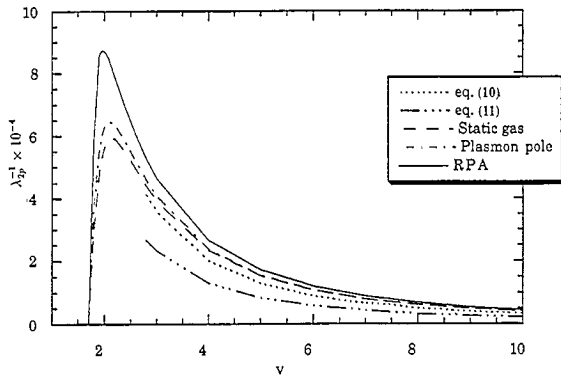


Fig. 1. Double-plasmon inverse mean free path, as a function of the velocity of the projectile. The solid line represents the result obtained after introduction of full RPA response functions into Eq. (2). Dashed and dashed-dotted lines represent the result obtained in the static electron gas and plasmon-pole approximations, respectively, and dotted and dashed-dotted-dotted-dotted lines, results obtained from Eqs. (10) and (11), respectively.

Eq. (11) in which q_c has been approximated by ω_p/q_F . Though at high velocities of the ion both the static electron gas and plasmon-pole approximations give, as hoped, an excellent account of the full RPA result, introduction of the low- q limit gives a result with a similar dependence on v as the full RPA result but much smaller. It is well-known that single-plasmon excitation probabilities are very well reproduced by introducing the low- q limit of the linear response function; however, momentum transfers involved in coherent double-plasmon excitations are much higher, up to $2q_c$, and virtually excited electron-hole pairs may decay, via a triple vertex, into two real low- q excitations, plasmons, so that the low- q limit of double-plasmon excitation probabilities is not suitable for quantitative purposes. Nevertheless, Eq. (10) gives, in the high-velocity limit, the correct dependence on the velocity of the projectile.

It is interesting to notice that both single and double plasmon excitation probabilities exhibit, in the high velocity limit, the same dependence on v , which is a consequence of the fact that they are both linear with respect to the external perturbation and proportional, therefore, to the square of the projectile charge. This is obvious from Fig. 2, where both the full RPA single and double plasmon inverse mean free paths have been plotted, the double-plasmon inverse mean free path having been rescaled by a factor of $\approx 10^2$. In the low-velocity regime, however, the nonlinearity of the medium giving rise to non-zero double plasmon excitation probabilities results in these probabilities being relatively greater as the velocity diminishes.

Finally, in Fig. 3 the ratio $\lambda_{2p}^{-1}/\lambda_{1p}^{-1}$ is plotted by a solid line, together with results derived from Eqs. (10) and (11). In the high velocity limit the step of these curves diminishes, showing, again, that both single and double

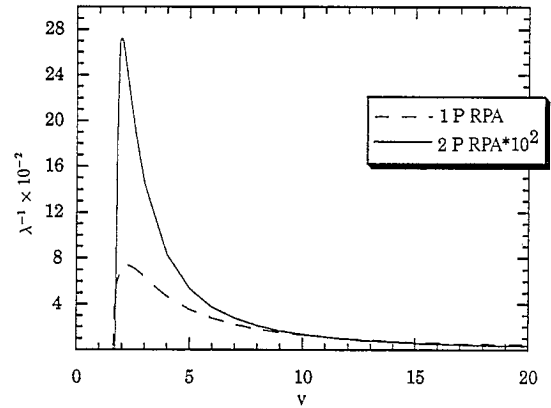


Fig. 2. Inverse mean free path for double and single plasmon excitations, versus the velocity of the projectile, evaluated in the full RPA. Double-plasmon inverse mean free paths have been multiplied by a factor of $\approx 10^2$.

plasmon inverse mean free paths present similar dependences on the velocity of the projectile, while as the velocity decreases double plasmon excitation probabilities become relatively more important. In particular, for 40 keV incident electrons ($v \approx 51.27$) we find, in the full RPA, a ratio of 1.93×10^{-3} , while Eqs. (10) and (11) result in ratios of 1.36×10^{-3} and 8.94×10^{-4} , respectively. Our full RPA result is in agreement with most recent experiments performed by Schattschneider et al. [9] with 40 keV incident electrons in aluminum, who imposed an upper limit of 5.0×10^{-3} onto the relative probability of double plasmon excitation.

In conclusion, double plasmon excitation probabilities for aluminum have been evaluated in the full RPA, and the reliability of the previously studied low- q limit and, also, static electron gas and plasmon-pole approximations has

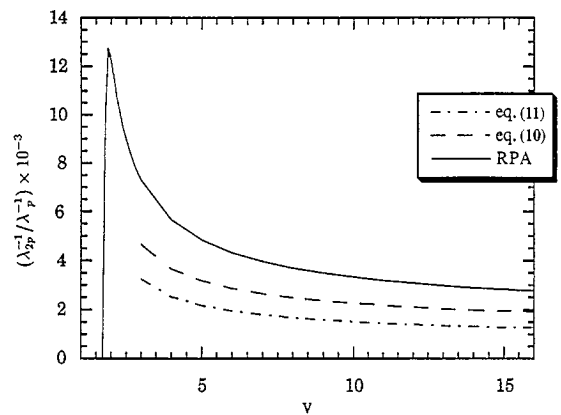


Fig. 3. Double-plasmon relative to single-plasmon inverse mean free path, as a function of the velocity of the projectile, in the full RPA (solid line) and, also, from Eqs. (10) (dashed line) and (11) (dashed-dotted line)

been investigated. In future work we shall, also, investigate multiple excitations in the electron gas, coming from both electron–hole pairs and plasmons.

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