ARTICLE IN PRESS

Nuclear Instruments and Methods in Physics Research B xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Plasmon damping in the free-electron gas model of solids

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ARTICLE INFO

Article history:
Received 16 November 2016
Received in revised form 18 May 2017
Accepted 20 May 2017
Available online xxxx

Keywords:
Plasmon damping
Dielectric formalism
Quantum formulation

ABSTRACT

We address the problem of quantifying the decay of plasmons excited in the electron gas of a condensed medium. Within the dielectric formalism, we thoroughly describe the theoretical framework in which we define the damping parameter γ. We present two detailed procedures to assess it as a function of the momentum transfer q, one based on a classical description of the excitation process and the second based on a quantum formulation of it. We present results corresponding to aluminum and magnesium, and compare them with experimental data obtained from the literature.

1. Introduction

Plasmons are collective excitations of the valence electrons of a solid and, along with individual excitations, are one the main causes of the energy loss of an external charged particle traversing a condensed medium [1,2]. The excitation of plasmons and their subsequent decay are of great interest in many branches of fundamental and applied physics [3–5]. Charged-particle spectroscopic techniques, such as EELS, REELS, XPS, etc. [6] show distinct features corresponding to the activation of plasmon modes by the interaction with the incident particle. The shape of the detected signals is determined to a large extent by the way plasmons interact with the medium and ultimately decay. In particular, the width of plasmon characteristic peaks is mainly given by the way the plasmon energy is dissipated.

A widely used quantum model to describe these processes was originally proposed by Lindhard [7], assuming a random phase approximation (RPA) for the free electron gas. This model takes into account both types of electronic excitations (individual and collective), and supplies a very good approximation to the dielectric response of real metals [8]. Within this frame, plasmons decay into a single electron–hole pair and it only occurs when the momentum transfer exceeds a critical value [9,10]. Below this value, plasmons are long-lived excitations with a well defined dispersion relation and negligible decay rate. They can be described using the so-called plasmon pole approximation [11], which models the typical narrow peak centered at a characteristic frequency ω_P and includes a small damping parameter γ to account for its width. In the transition to the individual-excitations regime, the damping increases and the sharp peak widens. Experimental data show that the dependence of γ with the momentum transfer reflects this transition with a threshold behaviour around a critical value [12–14].

The theoretical description of plasmon's decay has been studied in several works and with different approaches [15–18]. However, to our knowledge, an accurate quantitative theoretical model to calculate the damping rate γ remains an open question.

In this work, we explore different methods (from classical to quantum approaches) for quantifying γ in order to determine its value for realistic situations. The paper is structured as follows: in Section 2 we briefly develop the theory related to the response of the electron gas of a solid to the perturbation represented by an external charged particle. Section 3 is devoted to explain the different methods considered for the calculation of γ and to compare their results between them and with experimental data. Finally, we make some concluding remarks in Section 4.

2. Theoretical description of plasmon excitation

As we mentioned above, plasmons are one of the most significant effects in the response of the electron gas in a solid to an external perturbation (e.g., a charged particle). In classical electrodynamic theory, this response is mediated by the complex dielectric function $\epsilon(\mathbf{q},\omega)$, which gives the relation between the induced and the external charge densities in the reciprocal Fourier space with variables $\{\mathbf{q},\omega\}$. These variables are to be identified with

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http://dx.doi.org/10.1016/j.nimb.2017.05.046
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the momentum transfer and the frequency of the excitations. Here we consider an homogeneus and isotropic medium, where the relevant variable is the modulus of the wave vector $q = |\mathbf{q}|$, so the induced charge density $\rho_{\rm ind}$ can be written in terms of the external charge density $\rho_{\rm ext}$ as

$$\rho_{\rm ind}(\mathbf{q},\omega) = \rho_{\rm ext}(\mathbf{q},\omega)\left(\frac{1}{\epsilon(q,\omega)} - 1\right). \quad (1)$$

The zeroes of $\epsilon(q,\omega)$ yield the resonances identified as plasmon modes, which correspond to the poles of the energy loss function $ELF(q,\omega)$:

$$ELF(q,\omega) = \mathrm{Im}\left[-\frac{1}{\epsilon(q,\omega)}\right]. \quad (2)$$

Lindhard's model gives for $\epsilon(q,\omega)$ the following expression in terms of the reduced variables $u = \omega/kv_F$ and $z = q/2k_F$:

$$\epsilon_L(u,z) = 1 + \frac{\chi^2}{z^2}[f_1(u,z) + if_2(u,z)] \quad (3)$$

with v_F and k_F are the Fermi velocity and Fermi wave vector respectively, $\chi = e^2/\pi\hbar v_F$ is a density parameter and f_1 and f_2 are given by

$$f_1(u,z) = \frac{1}{2} + \frac{1}{8z}[g(z-u) + g(z+u)] \quad (4)$$

$$f_2(u,z) = \begin{cases} \frac{\pi}{2}u & z+u<1 \\ \frac{\pi}{8z}(1-(z-u)^2) & |z-u|<1<z+u \\ 0 & |z-u|>1, \end{cases} \quad (5)$$

with

$$g(x) = (1+x^2)\ln\left|\frac{x+1}{x-1}\right|.$$

This formulation divides the plane (q,ω) in three regions corresponding to the allowed excitations due to the energy and momentum transfers from the incident particle. Individual excitations take place in the band region with $|u-z|<1$, where the imaginary part of ϵ is different from zero ($\mathrm{Im}[\epsilon_L] \neq 0$). In the other regions there is no contribution to the *ELF*, except along a line defined by the condition $\epsilon_L = 0$, where the plasmon excitations occur. This line defines the dispersion curve $\omega_{pl}(q)$, as shown in Fig. 1 for a typical metal. This figure presents a map of the *ELF* calculated for aluminum using the Lindhard model for $\epsilon(q,\omega)$. The plasmon resonance $\omega_{pl}(q)$ distinctively shows up in the region $q<q_c, \omega<\omega_c$, being (q_c,ω_c) a critical point determined by the intersection of the plasmon line and the upper boundary of the region of individual excitations; beyond this critical point, the line widens as it enters the region where plasmons are heavily damped by the individual excitations. The width of these resonances is determined by the way the plasmon energy is dissipated.

In the context of the plasmon-pole approximation (PPA) the dielectric function is represented as [11]:

$$\epsilon_{PPA}(q,\omega) = 1 - \frac{\omega_P^2}{\omega(\omega+i\gamma) + \omega_P^2 - \omega_q^2}, \quad (6)$$

Here, ω_P is the resonant plasma frequency, and $\omega_q = \omega_{pl}(q)$ is the dispersion relation. The damping is introduced here with γ playing the role of the imaginary part of a complex frequency ($\omega \to \omega + i\gamma$) and gives the width of the plasmon resonance in the *ELF*. A usual approach for the dispersion relation is given by $\omega_q^2 = \omega_P^2 + \beta^2q^2 + \alpha^2q^4$ where β is a typical velocity, related to the

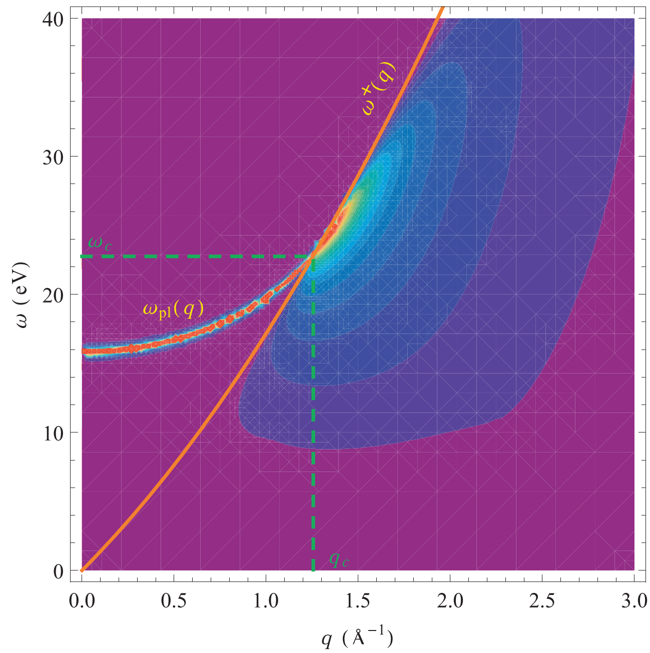

Fig. 1. Map of the ELF using Lindhard's model. The plasmon dispersion relation $\omega_{pl}(q)$ emerges in the $q<q_c$ region with a sharp but finite width since we have considered a complex frequency $\omega = \omega + i\eta$ in the Lindhard's equation for $\epsilon(q,\omega)$. The line $\omega^+(q)$ limits the region where individual excitations are allowed from that where they are forbidden.

Fermi speed as $\beta^2 = (3/5)v_F^2$, and $\alpha = \hbar/2m_e$. Notice that, in the limit $q \to 0$ we obtain from Eq. (6) the well-known Drude's approximation [19] which describes the non-dispersive case $\omega_q = \omega_P$:

$$\epsilon(\omega) = 1 - \frac{\omega_P^2}{\omega(\omega+i\gamma)}. \quad (7)$$

Continuing the analysis of Fig. 1, we observe that in the transition zone it is still possible to follow the plasmon line beyond the critical point, in a sort of fuzzy dispersion relation $\tilde{\omega}_{pl}(q)$; energy loss spectra will show a wide peak but with a defined maximum at a certain value $\tilde{\omega}_q$. In this sense we will be able to determine the value of γ as a function of q for $q>q_c$. In the following we explore different approaches to accomplish this task.

3. Determination of the damping parameter γ

3.1. The plasmon decay process

The damping of plasmons can be easily visualized through the following thought experiment. Let us consider an external charge density, oscillating in an arbitrary direction x with amplitude A, that is switched off at $t=0$,

$$\rho_{\rm ext}(\mathbf{r},t) = Ae^{ik_1x-\omega_1t}e^{\eta t}H(t), \quad (8)$$

with $H(t)=1$ for $t \leqslant 0$ and $H(t)=0$ for $t>0$, and let η be a smallness parameter that will be taken as zero at the end of the calculation. If we take k_1 and ω_1 as those corresponding to a plasmon (using the dispersion relation $\omega_1 = \omega_{pl}(k_1)$), we ensure that it will excite a pure plasmon mode of frequency ω_1 in the x direction. Now, since the perturbation is switched off at $t=0$, we can study how the plasmon decays for $t>0$.

We write the external charge density in Fourier space

$$\rho_{\rm ext}(\mathbf{q},\omega) = \frac{A(2\pi)^3\delta(\mathbf{q}_\perp)\delta(q_x-k_1)}{i(\omega-\omega_1-i\eta)} \quad (9)$$