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How the choice of model dielectric function affects the calculated observables

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ABSTRACT

It is investigated how the model used to describe a dielectric function (i.e. a Mermin, Drude, Drude–Lindhard, Levine–Louie with relaxation time dielectric function) affects the interpretation of a REELS experiment, the calculation of the electron inelastic mean free path as well proton stopping and straggling. Three dielectric functions are constructed that are based on different models describing a metal, but have identical loss functions in the optical limit. A loss function with the same shape, but half the amplitude, is used to derive four different model dielectric functions for an insulator. From these dielectric functions we calculate the differential inverse mean free path, the mean free path itself, as well as the stopping force and straggling for protons. The similarity of the underlying physics between proton stopping, straggling and the electron inelastic mean free path is stressed by describing all three in terms of the differential inverse inelastic mean free path. To further highlight the reason why observed quantities depend on the model dielectric function used we study partial differential inverse inelastic mean free paths, i.e. those obtained by integrating over only a limited range of momentum transfers. In this way it becomes quite transparent why the observable quantities depend on the choice of model dielectric function.

1. Introduction

Many processes, in particular the interaction of charged particles with matter, can be described in terms of the dielectric function $\epsilon(\omega,q)$ with q the momentum and ω the energy transfer. Unfortunately $\epsilon(\omega,q)$ is generally unknown although it can be measured directly in a transmission electron energy loss experiment [1,2] (or inelastic X-ray scattering experiment [3]) or calculated from first principle [4]. When this information is not available one has to use model dielectric functions. In that case only a limited number of parameters are required to obtain the dielectric function everywhere in (ω,q) space. Parameters for the model are usually determined by fitting the Energy Loss Function $\mathrm{Im}[-1/\epsilon(\omega,q)]$ at $q=0$ (from now on referred to as ELF) to optical data, which are much more widely available [5]. Subsequently, one can then calculate frequently used quantities such as the electron inelastic mean free path (IMFP) [6] or ion stopping and straggling from the dielectric function by a (weighted) integration of the loss function over (ω,q) space [7]. The results of these calculations are only valid if the model, describing how the dielectric function varies away from $q=0$, is correct. Here we want to study systematically how the outcome of such calculations depends on the model used.

For this purpose we construct different model dielectric functions that coincide at $q=0$. Then we calculate the aforementioned observables as well as the differential inelastic inverse mean free path (DIIMFP), the central quantity in REELS (Reflection Electron Energy Loss Spectroscopy). Such an approach will obviously show the differences of the calculated quantities for the different models, but it is often more difficult to pinpoint the origin of these differences. In all cases the calculated quantity is obtained by a weighted integration of the loss function over all accessible q values. To gain insight in the nature of the differences we integrate over only a fraction of allowed q values, and see for what range of q values the contribution to the calculated quantity of the models differ.

The main aim of this paper is to get some insight in what the consequences are of adoption one of the available model dielectric functions for the interpretation of their experiment, and under what condition this assumption is crucial for the outcome of the derived parameter(s), in particular the differential inverse electron mean free path (DIIMFP) which relates to REELS experiments, the

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IMFP in electron spectroscopy and ion stopping and straggling. The approach described will highlight the similarity of the underlying physics of the electron IMFP and ion stopping and straggling.

In the context of the IMFP of water many issues explored here where recently discussed by Shinotsuka et al. [8]. The work of Nikjoo et al. [9] describes the state-of-the-art of our knowledge of the interaction of charged particles with matter in the context of medical physics.

2. Model dielectric functions

2.1. Metals

Here we present briefly the model dielectric functions we will be using. Most of them have been described extensively in the literature before, e.g. see Ref. [9–13].

The first one is the Drude dielectric function $\epsilon^{\mathrm{D}}(\omega,q)$ with $\epsilon_1^{\mathrm{D}}(\omega,q)$ and $\epsilon_2^{\mathrm{D}}(\omega,q)$ the corresponding real and imaginary part:

$$\epsilon_1^{\mathrm{D}}(\omega,q)=\epsilon_b-\sum_i\frac{A_i(\omega^2-\omega_i(q)^2)}{(\omega^2-\omega_i(q)^2)^2+\Gamma_i^2\omega^2} \tag{1}$$

$$\epsilon_2^{\mathrm{D}}(\omega,q)=\sum_i\frac{A_i\Gamma_i\omega}{(\omega^2-\omega_i(q)^2)^2+\Gamma_i^2\omega^2} \tag{2}$$

here A_i (in units of (energy)^2) relates to the density of electrons with binding energy ω_i. Γ_i determines the width of the excitation. ϵ_b is the background dielectric constant due to the polarizability of the core electrons. Such a model dielectric function was used for the interpretation of REELS experiments by Tung et al. [14], Kwei et al. [15] and Werner et al. (e.g. [16]). The energy of oscillator i can depend on q (dispersion). This dependence will be assumed here to have a simple form (using atomic units):

$$\omega_i(q)=\omega_i(0)+\alpha_i\frac{q^2}{2} \tag{3}$$

but more complex dependencies (e.g. full dispersion [17]) could be used as well. The case of $\alpha_i=1$ is often referred to as 'free-electron dispersion'. For deeper levels α_i is often chosen much smaller than 1. For metals there is one component (representing the conduction electrons) with $\omega_i(0)=0$. For a free-electron metal this is the only component and the loss function $\mathrm{Im}[-1/\epsilon(\omega,0)]$ has then a maximum at $\sqrt{A_1}$. Such a free electron plasma will have, away from $q=0$, a maximum in $\mathrm{Im}[-1/\epsilon(\omega,q)]$ at $\sqrt{A_1+\omega_1(q)^2}$. The dispersion of this peak is thus different from a free electron dispersion, even with $\alpha_1=1$. If there is more than one component in the dielectric function then different components will 'repel' each other and peak positions are somewhat from $\sqrt{A_i+\omega_i(q)^2}$ [11].

A second model is often referred to as the Drude–Lindhard (DL) model [18]. Here ϵ^{DL} is defined in terms of $1/\epsilon(\omega,q)$ rather than $\epsilon(\omega,q)$ itself:

$$\mathrm{Im}\left[\frac{-1}{\epsilon^{\mathrm{DL}}(\omega,q)}\right]=\sum_i C_i\frac{\omega\Gamma_i\omega_i(0)^2}{(\omega^2-\omega_i(q)^2)^2+\omega^2\Gamma_i^2} \tag{4}$$

and for the real part:

$$\mathrm{Re}\left[\frac{1}{\epsilon^{\mathrm{DL}}(\omega,q)}\right]=1+\sum_i C_i\frac{(\omega^2-\omega_i(q)^2)\omega_i(0)^2}{(\omega^2-\omega_i(q)^2)^2+\omega^2\Gamma^2} \tag{5}$$

In the context of REELS such an approach is used e.q. in the QUASES package [19,20]. $\omega_i(q)$ is again defined as in Eq. (3) but now $\alpha_i=1$ implies that a peak disperses indeed in the same way as a free electron. There is no interaction between different oscillators, as there is in the Drude case, as now the dielectric function is defined in terms of the loss function itself.

For a free electron metal the DL dielectric function is at $q=0$ equivalent to the Drude dielectric function if $C_1=1$ and $\omega_1^{\mathrm{DL}}=\sqrt{A_1}$, but away from $q=0$ their dispersion will differ somewhat. For a metal $\mathrm{Re}\left[\frac{1}{\epsilon^{\mathrm{DL}}(0,0)}\right]$ should correspond to 0, as DC fields are completely screened (i.e. $\epsilon_1(0,0)=\infty$). This implies that $\sum_i C_i=1$.

These loss functions have their roots in classical physics. Lindhard derived, based on quantum physics, a dielectric function for a free electron gas $\epsilon^{\mathrm{L}}(\omega,q)$ [21] (see appendix for details). Here the loss function consists of a delta function (describing collective excitations, or 'plasmons') and a continuous part (describing single-particle excitations). Mermin added relaxation time to the Lindhard dielectric function which transforms the delta function to a peak with finite width [22]:

$$\epsilon^{\mathrm{M}}(\omega,q)=1+\frac{(1+i\Gamma/\omega)(\epsilon^{\mathrm{L}}(\omega+i\Gamma,q)-1)}{1+i\Gamma/\omega[\epsilon^{\mathrm{L}}(\omega+i\Gamma,q)-1]/[\epsilon^{\mathrm{L}}(0,q)-1]} \tag{6}$$

Abril et al. used a sum of Mermin loss functions to fit a optical data [23] or REELS data (e.g. [24]) to describe the proton stopping. Denton et al. used the Mermin dielectric function to calculate the electron inelastic mean free path [25]. Da et al. used a large number of positive and negative Mermin oscillator to fit the ELF of Cu and calculate its inelastic mean free path [26]. At $q=0$ the Mermin loss function coincides with the DL loss function with the same parameters. The Mermin loss function has dispersion 'build in' and away from $q=0$ the width of peak in $\mathrm{Im}[-1/\epsilon(\omega,q)]$ increases and becomes much larger than the width of the corresponding DL Loss function (see e.g. [27]). In this paper we will consider a simple model dielectric function consisting of two components. One component causes a peak in the ELF function at 15 eV. The second component corresponds to a peak at 80 eV. This model could be seen as a very crude model of Al where the first peak corresponds to the free-electron plasmon peak, and the second peak is due to the (combined) 2p and 2s electrons. The coefficients of these components are shown in Table 1 and the loss function and ϵ_1, ϵ_2 are reproduced in Fig. 1. The coefficients are chosen such that the ELF (Energy Loss Function at $q=0$) of all three model dielectric functions are identical.

2.2. Insulators

In an insulator the band gap has the effect of moving the loss features to higher energies. However, the dielectric function should remain compliant with sum rules, e.g. the Bethe sum rule:

$$\frac{1}{2\pi^2}\int_0^\infty\omega'\mathrm{Im}\left[\frac{-1}{\epsilon(\omega',0)}\right]d\omega'=N, \tag{7}$$

with N the number of electrons per unit volume. When using the DL or Mermin model, the shift of the loss function to higher energy due to band gap means that its area, and hence the coefficient(s) C_i, should decrease in order to comply with the sum rule. Hence for an insulator $\sum_i C_i<1$. This has also as a consequence that $\mathrm{Re}[1/\epsilon(0,0)]$ has values between 0 and 1, and hence $\epsilon_1(0,0)$ is finite and larger than 1 as required for an insulator.

An insulator with a loss function with a peak at the same energy as the loss function of a metal will thus have a lower electron density. We constructed 4 model loss functions with half the electron density of our metal loss function for simplicity, but with the peaks in the ELF at the same energy loss position (and the same width). The values of the parameters used are given in Table 2. To get half the electron density within the Drude model one has to halve the A_i parameters. To retain the same peak position one has to increase