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A model dielectric function for low and very high momentum transfer

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ABSTRACT

A model dielectric function is derived for TiO_2 based on reflection electron energy loss spectroscopy data and photoabsorption cross sections. The model is based on a set of Mermin oscillators. The input data is dominated by excitations at low momentum transfer, i.e. near the optical limit. Surprisingly the dielectric function derived at low momentum transfer describes the Compton profile quite well, while approaches based on Drude oscillators fail dramatically. The link between the dielectric function in the high-momentum transfer limit and a Compton profile is discussed. The underlying reason why the Mermin approach, which is based on a free electron model, is successful in describing the Compton profile is tentatively discussed.

1. Introduction

There is a lot of interest recently in methods to obtain the dielectric function $\epsilon(\boldsymbol{q},\omega)$ (with $\boldsymbol{q}$ the momentum transfer and ω the energy loss) of materials over an extended range of energy and momentum [1–4]. These quantities are the basis for the calculations of the inelastic mean free path of electrons and the stopping power of fast ions in matter. More generally the dielectric function is an essential ingredient of the description of electron–electron correlation in matter. Extraction of the dielectric function based on reflection electron energy loss spectroscopy(REELS) is usually based on extended Drude or Drude–Lindhard (D–L) models [5,6]. In its simplest form this approach does not give any broadening of the loss function (described by $\mathrm{Im}[-1/\epsilon(\mathbf{q},\omega)]$) with increasing momentum transfer, in clear contrast to experiment [7,8]. Agreement with the experiment can be improved by introducing a q-dependent broadening term [9,10].

This restriction is not present in the Mermin loss function [11]. It is based on a free-electron model and the width of the loss feature depends on $\mathbf{q}$, while maintaining the Bethe (or the related Tho mas–Reiche–Kuhn) sum rule for all $\boldsymbol{q}$ as well as the Kramers–Kronig sum rule. These desirable properties made the Mermin description of the valence band dielectric function the basis for the determination of the stopping of ions in matter [12].

A scattering electron interacts coherently over a distance of the order of $1/q$. At small momentum transfer the scattered electron interacts thus with a rather large volume of the target and one probes long-range density fluctuations (plasmons). At very large momentum transfer the projectile interacts coherently only with a small volume, containing only a single electron. In that case one can describe the interaction as a binary collision of the projectile and a target electron. The loss function at these large q values is usually referred to as the 'Bethe ridge'. Here the loss function reveals information about the target electron momentum distribution and can be considered a Compton profile [13]. In this paper we aim at obtaining a simultaneous description of both limiting conditions with a single dielectric function.

For a free electron gas, Lindhard derived a dielectric function that described both the collective behaviour at small q values and the single-particle excitations at large q values. The Mermin dielectric function is an extension of the Lindhard dielectric function that allows for a finite width of the peak in the loss function due to collective excitations at low q. There are several approaches to derive a dielectric function when the free electron approximation does not apply, e.g. by Penn [14] and Ashley [15], and a comparison of different approaches is given in Refs. [16,17].

In recent days it has become popular to describe the loss function of a wide range of materials by a sum of Mermin loss functions for small q excitations. A rigorous justification for the use of a sum of Mermin loss functions for materials that are far from free-electron like is usually not given. The heart of this paper is the investigation to what extent such an approach can still give a reasonable description of the loss function at both low and high q values.

In the next section we will revisit some of the properties of the Mermin loss function using the simple case of a carbon film as an example and demonstrate that both a more traditional electron energy loss spectrum and a Compton profile can be described in

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a uniform way by the Mermin loss function. Subsequently, we investigate more quantitatively, using TiO_2 as a test case, if we can obtain a set of Mermin oscillators that simultaneously describes the feature-rich spectrum of a reflection electron energy loss spectroscopy (REELS) measurement (extended to higher energy loss values by photo-absorption measurements) as well as its Compton profile. Moreover the obtained dielectric function is in agreement simultaneously with the Bethe and Kramers–Kronig sum rules. Finally it is discussed if the reasonable agreement obtained can be understood in terms of some kind of a 'local density approximation'.

2. A simple illustration for carbon films

First let us introduce the topic considering the experimental results for a very thin (35 Å thick) free-standing carbon film. Two scattering measurements were done with the same electron spectrometer employing very different kinematical conditions [18,19]. The experiment was done in a transmission geometry and is shown schematically in Fig. 1, the scattering angle Θ_s was ≈45°.

In the first experiment the incoming electron energy was 25 keV, and outgoing electrons with an energy loss up to 100 eV were detected. The obtained spectrum is shown in Fig. 2(A). It shows a sharp peak near zero energy loss (the elastic peak, due to electrons deflected from a nucleus), and a broader, less intense feature at larger energy loss. The latter is due to electrons that created an electronic excitation in the film. The maximum momentum of an electronic excitation created under these conditions is much smaller than the momentum required to deflect a 25 keV electron over 45°. Therefore there are no projectile electrons scattered directly into the analyser by such an electronic excitation. All the detected electrons have scattered elastically from a nucleus as well. The situation is as in a normal REELS experiment [20]. The inelastic mean free path (IMFP) of a 25 keV electron in graphite is ≈270 Å [21]. Under these conditions (film thickness much smaller than the IMFP) the probability for multiple inelastic excitations is small and the shape of the loss part of the spectrum can be directly compared to the shape of the DIIMP (differential inelastic inverse mean free path). The (bulk) DIIMP $W_b(\omega, E_0)$ is related to the dielectric function by:

$$W_b(\omega, E_0) = \frac{1}{\pi E_0}\int_{q_-}^{q_+}\frac{dq}{q}\mathrm{Im}\left[\frac{-1}{\varepsilon(q,\omega)}\right] \quad (1)$$

with the limits of integration given by: $q_\pm = \sqrt{2mE_0} \pm \sqrt{2m(E_0-\omega)}$ [20].

To model $\mathrm{Im}[-1/\varepsilon(\boldsymbol{q},\omega)]$ one can use either a D–L oscillator with 3 parameters, ω_p (plasmon energy at $q=0$), γ (width of this plasmon) and α (dispersion of the loss feature with q):

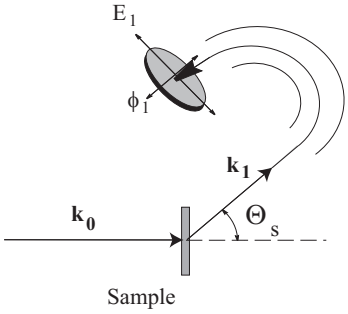

Fig. 1. A schematic view of the carbon film measurement done in a transmission geometry. The scattering angle Θ_s is 45°, $\mathbf{k}_0$ and $\mathbf{k}$ are the momentum of the incoming and detected electron respectively. The analyser resolves both the energy E_1 and azimuthal angle ϕ_1 of the detected electron.

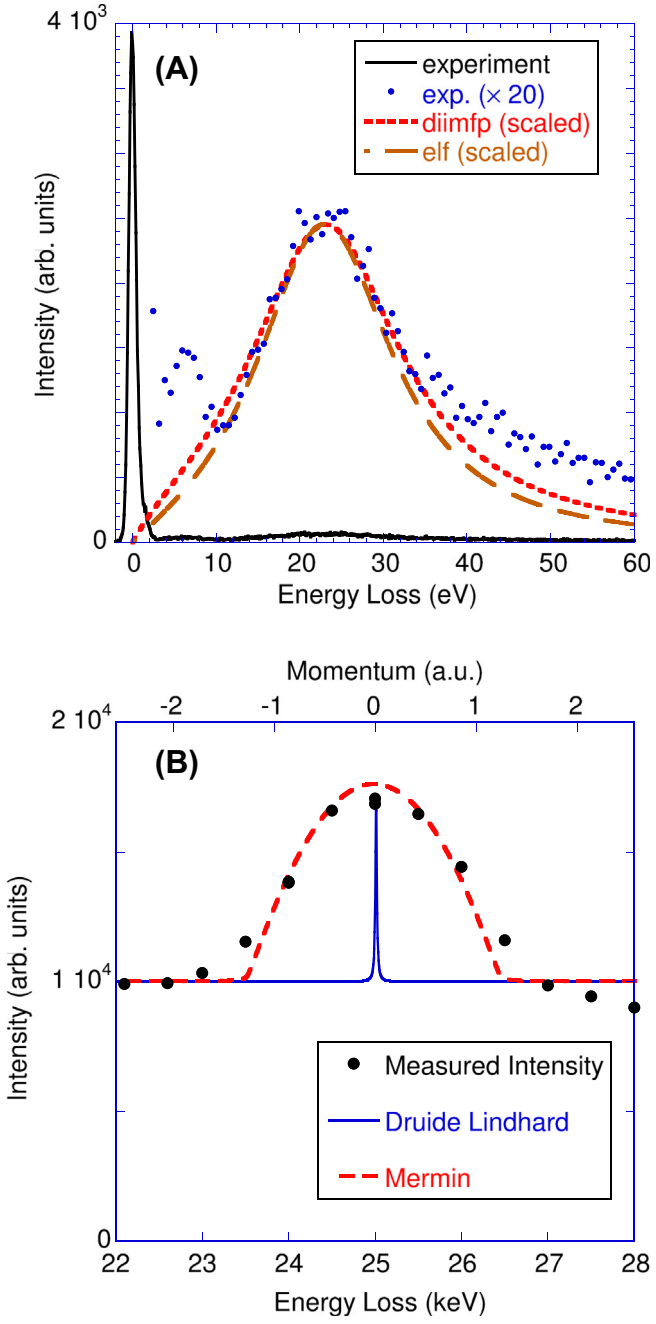

Fig. 2. (A) shows the energy loss spectrum for a 35 Å thick carbon film. The incoming energy was 25 keV. The shape of the loss spectrum can be described by the DIIMFP of a single Mermin oscillator centred at an energy loss of 25 eV and a width γ of 20 eV. The shape of the DIIMFP (short dashed line) is very close to the shape of the energy loss function $\mathrm{Im}[-1/\epsilon(q=0,\omega)]$ (ELF, long dashed line). A D–L based dielectric function with the same parameters has a very similar DIIMFP. In (B) the incoming energy was 50 keV and the spectrum was measured near $\omega = 25$ keV. The shape of this spectrum resembles $1/\epsilon(q=42.9,\omega)$ for the Mermin loss function, superimposed on a rather constant background. In contrast the width of the D–L loss function at $q = 42.9$ a.u. is orders of magnitude smaller.

$$\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{DL}}(\boldsymbol{q},\omega)}\right] = \frac{\gamma\omega_p^2\omega}{(\omega^2-\omega_q^2)^2+\gamma^2\omega^2}\Theta(\omega-E_{\mathrm{gap}}) \quad (2)$$

with $\omega_q = \omega_p + \alpha q^2$ and $\Theta(\omega - E_{\mathrm{gap}})$ the step function assuring that no excitations are possible within the bandgap of a semiconductor. Alternatively one can use a Mermin oscillator:

$$\varepsilon_{\mathrm{M}}(q,\omega) = 1 + \frac{(1+i\gamma\omega)[\epsilon_{\mathrm{L}}(q,\omega+i\gamma)-1]}{1+i\gamma/\omega[\epsilon_{\mathrm{L}}(q,\omega+i\gamma)-1]/[\epsilon_{\mathrm{L}}(q,0)-1]}\Theta(\omega-E_{\mathrm{gap}}) \quad (3)$$