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Mermin differential inverse inelastic mean free path of electrons in polymethylmethacrylate

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INTRODUCTION

Electrons continually interact with the matter around us. We use electron beams for our purposes, either on the side of production of materials or on that of their characterization. Let us think to the many applications such as the processing of materials with plasma and to the local melting of materials for joining large components. We use electron beams also in the electron lithography, an important technique utilized for the production of the microelectronics devices. Let us consider the importance of the beams of electrons in the characterization of the materials, performed using techniques such as the electron microscopy and all the electron spectroscopies. Electrons interact with the surfaces of the space-crafts. The plasma–wall interaction in the fusion reactors also involves electron–matter interaction. Electrons play a role also in the cancer proton therapy, where cascades of secondary electrons are produced. These electrons of very low energy are toxic for the human body cells, since they produce damage to the biomolecules due to ionizations/excitations and the resulting break of chemical bonds. Also the secondary electrons which have ultra-low energies – and which, for a long time, were thought to be relatively harmless – are dangerous for the biomolecules due to the so-called "dissociative electron attachment." And, of course, we wish to minimize the effects of the irradiation on the healthy tissues near to the diseased cells.

In all the mentioned cases, the modeling of the interaction of the electrons with the matter is very important, as it can be used to provide a solid theoretical interpretation of the experimental evidences.

The interpretation of the experimental results is often based on the Monte Carlo method (MC). The doping contrast in secondary-electron emission of pn-junctions was investigated by the use of the MC method (Dapor et al., 2008; Rodenburg et al., 2010). The modeling of electron interaction with polymers, and in particular with the polymethylmethacrylate, has been demonstrated to be very important in nano-metrology. Line-scan of resist materials with given geometrical cross-sections deposited on silicon or silicon dioxide, and the corresponding linewidth measurements, obtained with the secondary electrons in the Critical-Dimension Scanning Electron Microscope (CD SEM), require an interpretation that can be performed using MC calculations (Dapor et al., 2010).

The MC method, in turn, requires an accurate description of the differential inverse inelastic mean free path (DIIMFP), in order to calculate the inelastic mean free path (IMFP) and the distribution function for inelastic collisions of electrons causing energy losses less than or equal to given values.

MERMIN THEORY

The Mermin dielectric function (Mermin, 1970) is given by

$$\varepsilon_{\mathrm{M}}\left(\boldsymbol{q},\omega\right) = 1 + \frac{(1+i/\omega\tau)\left[\varepsilon^{0}\left(\boldsymbol{q},\omega+i/\tau\right)-1\right]}{1+(i/\omega\tau)\left[\varepsilon^{0}\left(\boldsymbol{q},\omega+i/\tau\right)-1\right]/\left[\varepsilon^{0}\left(\boldsymbol{q},0\right)-1\right]}, \quad (1)$$

where $\boldsymbol{q}$ is the momentum, ω the frequency, τ the relaxation time, and $\varepsilon^{0}(\boldsymbol{q},\omega)$ the Lindhard dielectric constant (Lindhard, 1954)

$$\varepsilon^{0}(\boldsymbol{q},\omega) = 1 + \frac{4\pi e^{2}}{q^{2}} B(\boldsymbol{q},\omega), \quad (2)$$

$$B(\boldsymbol{q},\omega) = \int \frac{d\boldsymbol{p}}{4\pi^{3}} \frac{f_{\boldsymbol{p}+\boldsymbol{q}/2} - f_{\boldsymbol{p}-\boldsymbol{q}/2}}{\omega - (\epsilon_{\boldsymbol{p}+\boldsymbol{q}/2} - \epsilon_{\boldsymbol{p}-\boldsymbol{q}/2})/\hbar}. \quad (3)$$

In these equations e is the electron charge, $f_{\boldsymbol{p}}$ is the Fermi–Dirac distribution, and $\epsilon_{\boldsymbol{p}}$ the free electron energy.

ENERGY LOSS FUNCTION

Let us now consider a superposition of free and bound oscillators. For any oscillator, the energy loss function (ELF) is given by the opposite of the imaginary part of the inverse of the Mermin dielectric function:

$$\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{M}}\left(\omega_{\mathrm{i}},\gamma_{\mathrm{i}};\boldsymbol{q},\omega\right)}\right] = \frac{\varepsilon_{\mathrm{M}2}}{\varepsilon_{\mathrm{M}1}^{2}+\varepsilon_{\mathrm{M}2}^{2}}. \quad (4)$$

where

$$\varepsilon_{M} = \varepsilon_{M_1} + i\varepsilon_{M_2} \quad (5)$$

and ω_{i}, and γ_{i} are, respectively, the frequency and the damping constant associated to each specific oscillator. A linear combination of Mermin-type ELFs, one per oscillator, allows to calculate the electron ELF for $q = 0$, for any specific material

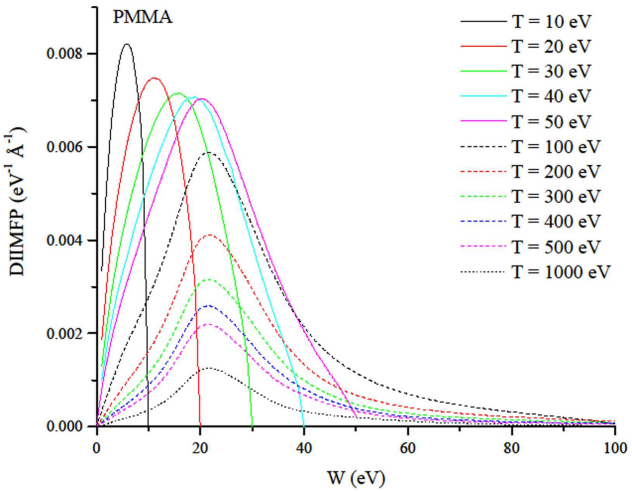

FIGURE 1 | Mermin DIIMFP of electrons in PMMA for selected values of the incident electron kinetic energy *T* in the range 10–1000 eV.

(Planes et al., 1996; Abril et al., 1998; de Vera et al., 2011)

$$\mathrm{Im}\left[\frac{-1}{\varepsilon\left(q=0,\omega\right)}\right] = \sum_{\mathrm{i}} A_{\mathrm{i}}\,\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{M}}\left(\omega_{\mathrm{i}},\gamma_{\mathrm{i}};q=0,\omega\right)}\right]. \quad (6)$$

In this equation, A_{i}, ω_{i}, and γ_{i} are determined looking for the best fit of the available experimental optical ELF. Actually, as both Mermin and Drude–Lorentz oscillators converge on the same values in the optical limit (i.e., for $q=0$) (de la Cruz and Yubero, 2007),

$$\mathrm{Im}\left[\frac{-1}{\varepsilon\left(q=0,\omega\right)}\right] = \sum_{\mathrm{i}} A_{\mathrm{i}}\,\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{M}}\left(\omega_{\mathrm{i}},\gamma_{\mathrm{i}};q=0,\omega\right)}\right] = \sum_{\mathrm{i}} A_{\mathrm{i}}\,\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{D}}\left(\omega_{\mathrm{i}},\gamma_{\mathrm{i}};q=0,\omega\right)}\right], \quad (7)$$

where the Drude–Lorenz functions are given by (Ritchie and Howie, 1977)

$$\mathrm{Im}\left[\frac{-1}{\varepsilon_D(\omega_i,\gamma_i;\,q=0,\omega)}\right] = \frac{\gamma_i\omega}{(\omega_i^2-\omega^2)^2+(\gamma_i\omega)^2}, \quad (8)$$

the best fit can also be performed using a linear combination of Drude–Lorentz functions, instead of Mermin functions.

For the present work, the parameters provided by de Vera et al. (2011) obtained by calculating the best fit of the Ritsko et al. (1978) experimental optical data, were used.

Once the values of the best fit parameters have been established, the extension out of the optical domain ($q\neq 0$) can be obtained by (Planes et al., 1996; Abril et al., 1998; de Vera et al., 2011)

$$\mathrm{Im}\left[\frac{-1}{\varepsilon\left(q,\omega\right)}\right] = \sum\nolimits_i A_{\mathrm{i}}\,\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{M}}\left(\omega_{\mathrm{i}},\gamma_{\mathrm{i}};q,\omega\right)}\right]. \quad (9)$$

DIFFERENTIAL INVERSE INELASTIC MEAN FREE PATH AND INELASTIC MEAN FREE PATH

The ELF allows, in turn, the computation of the DIIMFP, given by

$$\frac{d\lambda_{\mathrm{inel}}^{-1}}{d\hbar\omega} = \frac{1}{\pi a_0 T}\int_{q-}^{q+}\frac{dq}{q}\,\mathrm{Im}\left[\frac{-1}{\varepsilon(q,\omega)}\right], \quad (10)$$

where a_0 is the Bohr radius, T is the kinetic energy of the incident electrons and

$$q_{\pm} = \sqrt{\frac{2m}{\hbar^2}}\left(\sqrt{T}\pm\sqrt{T-\hbar\omega}\right). \quad (11)$$

The Mermin DIIMFP of electrons in PMMA is represented in **Figure 1**, for kinetic energies of the incident electrons in the range from 10 to 1000 eV.

The inverse of the integral of every curve presented in **Figure 1** provides, for each kinetic energy T, the IMFP. According to de la Cruz and Yubero (2007), the values of the IMFP calculated using the Tanuma, Powell, and Penn (TPP) empirical predictive formula (Tanuma et al., 1994) are systematically higher than the corresponding values calculated within the Mermin theory. For PMMA, according to our calculation, when $T=100$ eV, the Mermin IMFP is equal to 6.3 Å while when $T=1000$ eV, it is equal to 27.6 Å. According to TPP, the IMFP of PMMA is equal to 7.9 Å for $T=100$ eV and to 33.7 Å for $T=1000$ eV (Tanuma et al., 1994). Also approaches based on the Drude–Lorentz theory (Emfietzoglou et al., 2013; Dapor, 2014a,b; Dapor et al., 2015) provide values of the IMFP systematically higher than those obtained using the Mermin theory. The IMFP of PMMA calculated according to the Drude–Lorentz theory is equal to 10.1 Å for $T=100$ eV and to 33.5 Å for $T=1000$ eV (Dapor, 2014a).

It is not simple to express an opinion about the different approaches used today to calculate the DIIMFP, and hence the IMFP, due to the lack of experimental data and their quite large uncertainty (Dapor et al., 2015). Roberts et al. (1980) provided the experimental values of 29 ± 4 Å for 1196 eV electrons and 33 ± 5 Å for 1328 eV electrons in PMMA. On the one hand, also taking into account of the experimental uncertainty, de Vera et al. (2011) have shown that these values are closer to those predicted by the Mermin theory than to those predicted by the TPP formula, usually taken as a reference and provided, among other IMFP data, by the NIST database. On the other hand, the number of experimental data about electron IMFP in PMMA seems to be definitely too small to judge. Since Mermin theory is much more accurate it is preferable, even if we conclude that, at the moment, both Mermin theory and TTP empirical predictive formula are compatible with the available experimental data about IMFP in PMMA.

CONCLUSION

Calculations of the DIIMFP of electron in materials are of paramount importance

for modeling the transport of electrons in solid targets. In this paper, we presented the calculations of the DIIMFP of electrons in PMMA based on the Mermin theory (Mermin, 1970) and on the de Vera et al. (2011) best fit of the Ritsko et al. (1978) optical data.

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