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Inelastic interactions of low-energy electrons with biological media

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Abstract

A study of inelastic interactions of low-energy electrons with biological media is of fundamental importance in the understanding of radiation-induced actions leading to the biological damage. In the present work, electron energy loss properties in liquid water and solid DNA were calculated based on the dielectric response theory for valence band electrons and atomic models for inner-shell electrons. For electron interactions with the valence band, an extended Drude dielectric function was fitted to experimental optical data and then checked by sum rules. The valance band was considered as composed of many subbands with different oscillator strength, binding energy and damping coefficient. The real and imaginary parts of the dielectric function and the energy loss function were all checked. For electron interactions with inner shells, the generalized oscillator strength was calculated using the sum-rule-constrained binary-encounter model and the local-plasma approximation. The differential inverse mean free paths for the valence band and inner shells were calculated using the Born approximation. Exchange effect was estimated using an approach based on the Moller cross-section. Results of the present work were compared with similar calculations by other authors.

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Keywords: Electron; Inelastic interaction; Water; DNA

1. Introduction

A study of the inelastic interactions of low-energy (<10 keV) electrons with biological media, such as DNA and water, is of fundamental importance in the understanding of radiation-induced actions, both direct and indirect, leading to the biological damage. Monte Carlo simulations of these actions can be made using cross-sections of such interactions. For instance, an investigation of the DNA strand breaks caused by Auger electrons requires input data on the differential inverse mean free path (DIMFP) of these electrons in water and DNA [1].

In the present work, electron inverse mean free path (IMFP), stopping power (SP), and continuous slowing down approximation (CSDA) range in liquid water and DNA were calculated using the dielectric response function for the valence band and the generalized oscillator strength (GOS) for inner shells. For electron interactions with the valence band, an extended Drude dielectric function [2] was fitted to experimental optical data and then checked by sum rules. The valance band was considered as composed of many subbands with different oscillator strength, binding energy and damping coefficient. The real and imaginary parts of the dielectric function and the energy loss function were all examined and checked. For electron interactions with inner shells, the GOS was calculated using either the sum-rule-constrained binary-encounter model [3] or the local-plasma approximation [4]. DIMFPs for the valence band and inner shells were calculated using the quantum mechanical Born approximation. Exchange effect was estimated using an approach based on the Moller cross-section.

It showed that the difference in calculated results between liquid water and DNA was shown to arise mainly from inner shells. The exchange effect was important for electron energies below ~1 keV.

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2. Methods

For inelastic interactions, the response of a condensed medium to the energetic electron may be described by the dielectric function, $\varepsilon(k,\omega)$, for valence band electrons and by the GOS, $df/d\omega$, for inner-shell electrons. Here k and ω are, respectively, the momentum and energy transfers. Note that atomic units are used for quantities and expressions unless otherwise specified. Considering an energetic electron of energy E to interact with the valence band of the medium, the DIMFP of this electron for energy loss ω is given in the Born approximation as [5]:

$$\mu(E,\omega)=\frac{1}{E}\int_{k_-}^{k_+}\frac{1}{k}\,\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right]\mathrm{d}k, \tag{1}$$

where $k_\pm=\sqrt{2E}\pm\sqrt{2E-2\omega}$ are obtained from the conservations of energy and momentum and $\mathrm{Im}[-1/\varepsilon]$ is the energy loss function.

Assuming the valence band is composed of several subbands, each ith subband is characterized by its own oscillator strength A_i, damping constant γ_i, and excitation energy ω_i. Using the extended Drude model [2], the optical energy loss function may be given by

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(0,\omega)}\right]=\sum_i\frac{\omega_i^2\gamma_i\omega}{(\omega^2-\omega_i^2)^2+\gamma_i^2\omega^2}. \tag{2}$$

In the present work, Eq. (2) was fitted to experimental optical data to determine the parameters A_i, γ_i and ω_i. The real and imaginary parts of the dielectric function were inspected in the fittings. To ensure the accuracy of fitted parameters, the validities of the sum rules [6]

$$\int_0^\omega\omega'\varepsilon_2(0,\omega')\,\mathrm{d}\omega'=2\pi^2NZ(\omega) \tag{3}$$

and

$$\int_0^\omega\omega'\,\mathrm{Im}\left[\frac{-1}{\varepsilon(0,\omega')}\right]\mathrm{d}\omega'=2\pi^2NZ'(\omega) \tag{4}$$

were checked, where N is the molecular density and $Z(\omega)$ and $Z'(\omega)$ are the corresponding effective numbers of electrons per molecule contributed by excitations up to the energy transfer ω. These sum rules were applied by setting the upper limits of integration to both finite and infinite values. In the case of finite-range sum rules, $Z(\omega)$ and $Z'(\omega)$ were compared, at any given energy transfers, between results calculated from present fittings and determined from optical data. For infinite-range sum rules, the satisfaction of the present fittings was checked by the condition $4\pi NZ_m=\sum A_i$, where Z_m is the number of contributing electrons per molecule from the valence band. Fitting values were adopted until all sum rules were satisfied correctly. Note that $Z(\omega)$ and $Z'(\omega)$ merge to a constant value Z_m at very large ω. The energy loss function in Eq. (2) may be extended to any momentum transfers by replacing ω_i in the denominator of Eq. (2) by $\omega_i+k^2/2$.

There are influential effects, which contribute to the DIMFP of low-energy electrons. These include the corrections due to correlation and exchange. With the dielectric function fitted to optical data, the influence of correlation is already incorporated. The exchange correction can be estimated using a semi-empirical scheme based on the Moller differential cross-section. The exchange-corrected DIMFP is given by [7]

$$\mu_{\mathrm{ex}}(E,\omega)=\mu(E,\omega)+\mu(E,E-\omega)-[\mu(E,\omega)\mu(E,E-\omega)]^{1/2} \tag{5}$$

where the maximum energy loss becomes half of the electron energy.

The energy loss function of a weakly interacting system, such as inner shells in the condensed medium, is composed of GOS of molecules. The GOS of the ith inner shell is related to the energy loss function by [8]

$$\left[\frac{\mathrm{d}f(k)}{\mathrm{d}\omega}\right]_i=\frac{\omega Z}{2\pi^2n_i}\,\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right] \tag{6}$$

where n_i is the electron density of the ith inner shell and Z is the atomic number. For low-Z materials, a sum-rule-constrained binary-encounter model [3] may be easily applied to determine the GOS. This model involves the use of a binary-encounter GOS with empirical parameters determined by sum rules. For high-Z materials, the local-plasma approximation [4] is readily applicable to derive the GOS. This approximation assumes a local plasmon excitation, dependent on the electron density distribution, determined from the Hartree–Fock–Slater model. The molecular GOS is then obtained from the component atomic GOSs using the mixture rule.

The IMFP, SP and CSDA range for an energetic electron can be calculated using the DIMFP. Detailed formulas for these calculations are available elsewhere [9].

3. Results and discussion

In the present work, the energy loss function of Eq. (2) was fitted to optical data of liquid water [10]. The GOS of the oxygen K-shell was calculated using the sum-rule-constrained binary-encounter model. Fig. 1 shows the results of present calculations for the SP of liquid water as a function of electron energy. The dashed and dotted curves are the contributions from the valence band excluding and including the exchange effect, respectively. The chain curve is the contribution from the oxygen K-shell. The sum of these two contributions including the exchange effect, i.e. the solid curve, is compared to results (symbols) calculated by other authors [11,12]. Notice that circles and triangles are the results calculated without [12] and with [11] the exchange effect, respectively. The present results agree quite well with theoretical data of Emfietzoglou [11]. A comparison of the electron CSDA range in liquid water is made in Fig. 2 for the results calculated presently (curve), given in Watt [13] (squares), and recommended by the ICRU [14] (triangles).

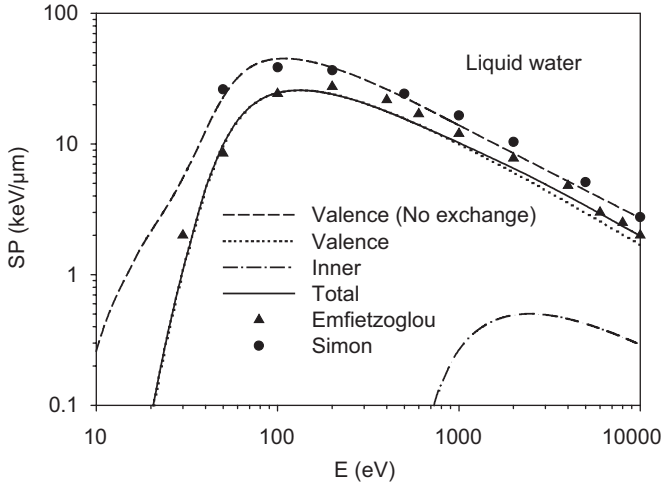

Fig. 1. A plot of the SP of liquid water as a function of electron energy. Curves and symbols are the results of present work: (triangles) [11] and (circles) [12].

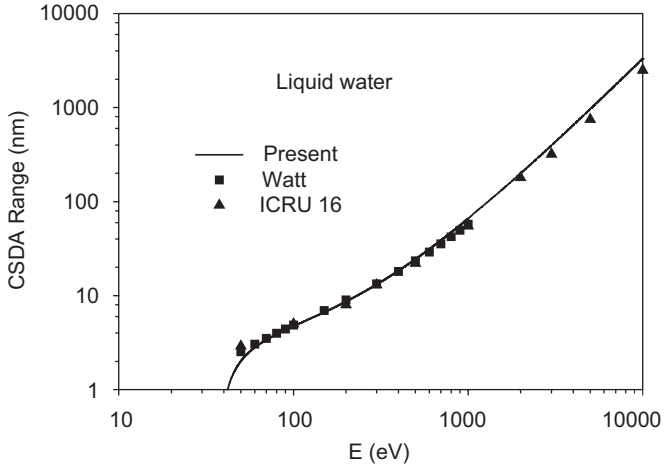

Fig. 2. CSDA range of electrons in liquid water. Curves and symbols are the results of present work: (squares) [13] and (triangles) [14].

It reveals that an extranuclear electron of energy less than ~5 keV (with CSDA range ~1 μm) is unable to reach the DNA within the cell nucleus. Therefore, to induce damage to the DNA molecule, low-energy electrons should be emitted within the cell nucleus. Thus, a DNA-bound Auger emitter is required for the effective killing of cells.

The energy loss function of Eq. (2) was also fitted to optical data of calf thymus DNA [15]. Considering the molecular formula C20H27N7O13P2 [12] for the typical DNA nucleotides, the GOS of the carbon, nitrogen, oxygen or phosphorous K-shell was calculated using the original derivation of the sum-rule-constrained binary-encounter model [16]. The GOS of the phosphorous 2p-subshell was calculated using the reconstructed formula of the sum-rule-constrained binary-encounter model [3]. The GOS of the phosphorous 2s-subshell was estimated using the local-plasma approximation. Fig. 3 is a plot of electron

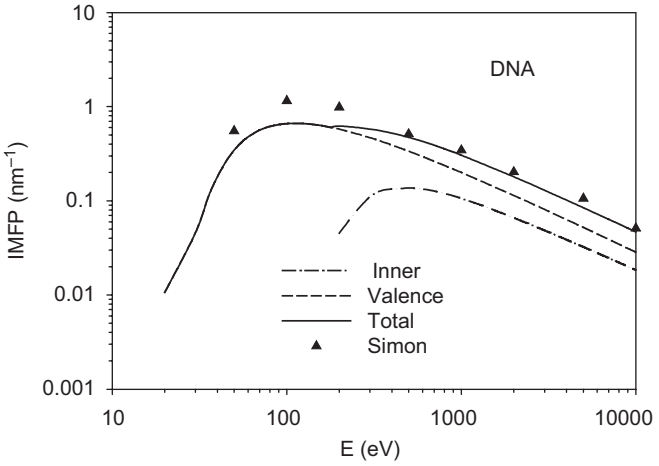

Fig. 3. IMFP of electrons in DNA. Curves and symbols are the results of present work and published data [12].

IMFP in DNA calculated is the present work (curves) and by other authors (symbols) [12]. The dashed, chain and solid curves are the results of contributions from the valence band, inner shells and the sum of them. Other theoretical data were calculated without the exchange effect. Although not shown here, the results of present calculations for the valence band are very close for DNA and liquid water. The contribution from inner shells, however, is larger for DNA than liquid water.

4. Conclusions

Energy loss properties of low-energy (<10 keV) electrons in liquid water and DNA were calculated using the dielectric function for the valence band and the GOS for inner shells. Results of these calculations were compared with similar calculations by other authors. It showed that the difference between liquid water and DNA was mainly due to inner shells. The exchange effect was important for electron energies below about 1 keV.

The present data can be used to simulate the radiation actions by low-energy electrons. Since all ionizing radiations produce low-energy electrons, it is conceived that much of the critical damage arises from the track-ends of these electrons.

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