Calculated electronic energy loss of swift proton and helium ion beams in liquid water

Isabel Abril[1], Rafael Garcia-Molina[2], Cristian D. Denton[1]
and Dimitris Emfietzoglou[3]

[1] *Departamento de Física Aplicada, Universitat d'Alacant, Apartat 99, E-03080 Alacant, Spain*

[2] *Departamento de Física-CIOyN, Universidad de Murcia, Apartado 4021, E-30080 Murcia, Spain*

[3] *Medical Physics Laboratory, University of Ioannina Medical School, Ioannina 451 10, Greece*

Abstract
The electronic energy loss of swift proton and helium beams in liquid water is theoretically evaluated. Our model is based in the dielectric formalism, taking into account the charge exchange of the projectile during its travel through the target. The electronic properties of liquid water are described by the MELF-GOS model, where the outer electron excitations are represented by a sum of Mermin functions fitted to the experimental data in the optical limit, whereas the inner-shell electron excitations are modelled by the corresponding atomic generalized oscillator strength. The inverse mean free path, the stopping power and the energy loss straggling are calculated, showing a reasonably good agreement with the available experimental data.

I. Introduction
By using the dielectric formalism we theoretically calculate the energy loss of swift proton and helium beams impinging in liquid water. This formalism reasonably accounts for the electronic excitations produced in the bombarded materials by the passage of fast particles. We have used an improved description of the energy loss function corresponding to liquid water by means of an empirical fitting to available optical data for the outer electrons and by generalized oscillator strengths to take into account electron excitations from the inner-shells. This model also fulfils physical constraints such as the *f*-sum rule, and provides the mean ionization energy I of liquid water. It is worth to mention the suitability of this model in the description of the target energy loss function in the whole energy-momentum plane. A more detailed analysis and discussion of the different dielectric descriptions of the liquid water is provided in [Emfietzoglou 2008]. Finally, we present our calculated results for the inverse mean free path (IMFP), the stopping power and energy loss straggling of proton and helium beams in liquid water, which are compared to experimental data when available.

II. Theoretical model
When energetic particles impinge on a material, most of their energy is lost exciting the electrons of the solid (i.e., inelastic energy loss). These energy loss processes can be characterize by the following magnitudes: the inverse mean free path (IMFP), Λ^{-1}, the stopping power, S_{p}, and the energy loss straggling Ω^2. The IMFP represents the probability per unit path length that an incident particle will suffer a process of inelastic interaction with the stopping medium, the stopping power is the mean energy lost by the projectile per unit

path length, while the energy loss straggling is the variance in the energy-loss distribution per unit path length.

At low and intermediate projectile energies it is necessary to consider the processes of electron capture from and loss to the target that give a continuous charge exchange of the projectile in their path through the solid, since it means a change in their energy loss. Therefore for a projectile with atomic number Z_1 and velocity v that bombards a target, the average IMFP, the stopping power and the energy loss straggling can be obtained from a weighted sum of the partials IMFP, Λ_q^{-1}, stopping power, $S_{\mathrm{p},q}$, and energy loss straggling Ω_q^2 for each charge q of the projectile,

$$\Lambda^{-1} = \sum_{q=0}^{Z_1} \phi_q \Lambda_q^{-1} \ , \tag{1}$$

$$S_{\mathrm{p}} = \sum_{q=0}^{Z_1} \phi_q S_{\mathrm{p},q} \ , \tag{2}$$

$$\Omega^2 = \sum_{q=0}^{Z_1} \phi_q \Omega_q^2 \ , \tag{3}$$

where ϕ_q is the probability to find the projectile in a given charge state q. Since the charge equilibrium is reached in a few femtoseconds after the projectile penetrates into the target, we assume that ϕ_q are the charge-state fractions at equilibrium, which depend on the target, the projectile and their velocity; we obtain ϕ_q from the CasP 3.1 code [Grande 2005]. For compound targets, such as water, this code applies Bragg rule to their constituents to find the final charge fractions.

The dielectric formalism, which based in the first Born approximation, provides the following expressions for the inverse mean free path, Λ_q^{-1}, the stopping power, $S_{\mathrm{p},q}$, and the energy loss straggling Ω_q^2 for a projectile with charge state q moving through a material

$$\Lambda_q^{-1} = \frac{2e^2}{\pi v^2} \int_0^{\infty} \frac{dk}{k} \rho_q^2(k) \int_0^{kv} d\omega \, \mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right] , \tag{4}$$

$$S_{\mathrm{p},q} = \frac{2e^2}{\pi v^2} \int_0^{\infty} \frac{dk}{k} \rho_q^2(k) \int_0^{kv} d\omega \, \omega \, \mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right] , \tag{5}$$

$$\Omega_q^2 = \frac{2e^2}{\pi v^2} \int_0^{\infty} \frac{dk}{k} \rho_q^2(k) \int_0^{kv} d\omega \, \omega^2 \, \mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right] , \tag{6}$$

where e is the absolute value of the electron charge, $\rho_q(k)$ is the Fourier transform of the projectile charge density for the charge state q, $\hbar k$ and $\hbar\omega$ are, respectively, the momentum and energy transferred in an inelastic process to the target, $\varepsilon(k,\omega)$ is the dielectric function and $\mathrm{Im}[-1/\varepsilon(k,\omega)]$ is the energy loss function (ELF) of the stopping material, which contains all the information about the electronic properties of the target. To the ELF of water we will devote the next section.

III. Energy loss function of liquid water

It is necessary a good description of the liquid water ELF for the entire $k-\omega$ plane, in order to obtain good results for the IMFP, the stopping power and the energy loss straggling.

Only recently experimental data of the energy loss function (ELF) of liquid water at momentum transfer different from zero have become available. These data are from

[Watanabe 1997, Hayashi 1998, 2000], where the authors studied the Compton scattering in liquid water of high energy photons, resulting from synchrotron radiation. By the use of inelastic X-ray scattering (IXS) spectroscopy, the GOS can be obtained in an absolute scale as a function of the transferred energy and momentum. This quantity is uniquely related to the dielectric response function, which determines the interaction of the material with a charged particle.

Old experimental data of ELF of liquid water in the optical limit are from Heller [Heller 1974], and in this limit new and old data from both research groups are very different. Therefore, it is very important to calculate again the stopping magnitudes of charged particles in liquid water, using reliable ELF as input data.

We present an improved description of the ELF corresponding to liquid water, obtained by an empirical fitting to available optical data and fulfilling physical constraints such as the *f*-sum rule. We use the MELF-GOS approach [Abril 1998] to describe the energy loss function of the liquid water. The electrons of the medium are divided into two groups: the inner-electrons, whose states retain an atomic character even in condensed medium and the outer-electrons, responsible of the phase state and chemical properties of the aggregate. Using this approach, the ELF is separated into two contributions:

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right]=\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right]_{\mathrm{outer}}+\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right]_{\mathrm{inner}} . \qquad (7)$$

The excitations of the outer electrons of the solid, including both collective and single-particle (electron-hole) excitations, are described by fitting the experimental optical ELF (k=0) to a sum of Mermin-type ELFs,

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(k=0,\omega)}\right]_{\mathrm{outer}}=\mathrm{Im}\left[\frac{-1}{\varepsilon(k=0,\omega)}\right]_{\mathrm{exp}}=\sum_i A_i\,\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{M}}(\omega_i,\gamma_i;k=0,\omega)}\right]_{\omega\geq\omega_{\mathrm{th},i}} , \qquad (8)$$

where ε_{M} is the Mermin dielectric function [Mermin 1970].

The fitting parameters ω_i and γ_i are related to the position and width, respectively, of the i-th Mermin-type ELF, while the coefficients A_i are the corresponding weights. $\hbar\omega_{\mathrm{th},i}$ is a threshold energy in this fitting procedure. For liquid water we fit to the experimental data from [Hayashi 2000] in the optical limit and obtain the parameters shown in the table.

In the other hand, inner-shell electrons retain their atomic character since they have large binding energies, therefore it is suitable to model their excitation spectrum in terms of generalized oscillator strengths (GOS) [Egerton 1989]. The connection between the ELF and the GOS model is given by

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right]_{\mathrm{inner}}=\frac{2\pi^2 N}{\omega}\sum_{nl}\frac{\mathrm{d}f_{nl}(k,\omega)}{\mathrm{d}\omega} , \qquad (9)$$

where $\boldsymbol{N}$ is the density of atoms in the target (for liquid water $N=4.949\cdot10^{-3}$ a.u.) and $\mathrm{d}f_{nl}(k,\omega)/\mathrm{d}\omega$ is the GOS of the (n,l) subshell. We use the hydrogenic approach to obtain the GOS because it is analytical and describes reasonably well the contribution of the K, L and M inner-shell ionization to the stopping magnitudes [Abril 2007]. For liquid water, we use the GOS of the oxygen K-shell.

In figure 1 we show the ELF of liquid water in the optical limit (k=0). The round symbols correspond to experimental data [Hayashi 2000] for liquid water measured by inelastic x-ray scattering spectroscopy. Dashed curves correspond to the experimental data obtained from x-ray scattering factors [Henke 1993], which permits to obtain the ELF at larger transferred energies. The solid curves correspond to our fitting according to the MELF-GOS method, including the contribution of the outer and the inner electrons. The outer electrons from liquid water were fitted by a sum of 3 Mermin-type ELFs, while the oxygen K-inner shell is calculated by the GOS method. In table I we list the fitting parameters for the outer electrons of liquid water.

Target	i	$\hbar\omega_{\mathrm{th},i}$ (eV)	$\hbar\omega_i$ (eV)	$\hbar\gamma_i$ (eV)	A_i
Liquid water	1	7	22.0	14.0	0.352
$N = 4.949\cdot10^{-3}$ a.u.	2	7	34.0	19.0	0.08366
	3	7	47.0	32.0	0.05

Table I. Parameters used to fit the outer-electron excitations of the ELF of liquid water. N is the mass density of the target.

One of the advantage of the MELF-GOS method is that the fit of the ELF in the optical limit, that is, when the momentum transfer is zero (k=0) can be analytically and automatically extended to $k \neq 0$ through the properties of the Mermin dielectric function and the GOS model [Planes 1996]. In figure 2 we show the energy loss function of liquid water for different values of the momentum transferred, k. Comparison our results with experimental data for liquid water obtaining by x-ray scattering spectroscopy [Watanabe 1997, Hayashi 2000] shows a good agreement.

The MELF-GOS method also demands that the f-sum rule must be satisfied for all wave number k. So, the number of electrons per atom that can be excited for a given transferred energy $\hbar\omega$, $N_{\mathrm{eff}}(\omega)$, which is given by

$$N_{\mathrm{eff}}(\omega) = \frac{m_e}{2\pi^2 e^2 N}\int_0^{\omega} d\omega'\,\omega'\,\mathrm{Im}\left[\frac{-1}{\varepsilon(k=0,\omega')}\right], \qquad (10)$$

should tend to the target atomic number as the transferred energy goes to infinity. Here m_e is the mass electron.

Another verification of the method consists in evaluating the mean excitation energy I, which must compare satisfactorily with experimental data (when available)

$$\ln I = \frac{\int_0^{\infty} d\omega\,\omega \ln\omega\,\mathrm{Im}[-1/\varepsilon(k=0,\omega)]}{\int_0^{\infty} d\omega\,\omega\,\mathrm{Im}[-1/\varepsilon(k=0,\omega)]}. \qquad (11)$$

We obtain I = 79.4 eV for liquid water. The contribution of the outer electrons (all the electrons except those of oxygen K-shell, give 46.7 eV as the mean excitation energy. Experimental data from Bischel [1992] gives 79 ± 0.5 eV. The recommended values of ICRU [1984] are: I = 75.0 eV for liquid water, I = 75.0 eV for ice, I = 71.6 eV and for vapour. Theoretical calculations based in the old ELF of liquid water obtain I = 81.8 eV [Dingfelder 2000].

In summary, the energy loss functions for liquid water have been modellized from experimental data in the optical limit, satisfying physical constraints such as the f-sum rule, and providing a reasonable value for the mean ionization energy. Besides, the ELF have a momentum dependence that agrees with experiments and are suitable, accurate and of easy use for energy loss calculations.

IV. Results and discussion

The following figures show the results of our calculations as well as the input data we have used. They are supposed to be self-explained through the figure caption.

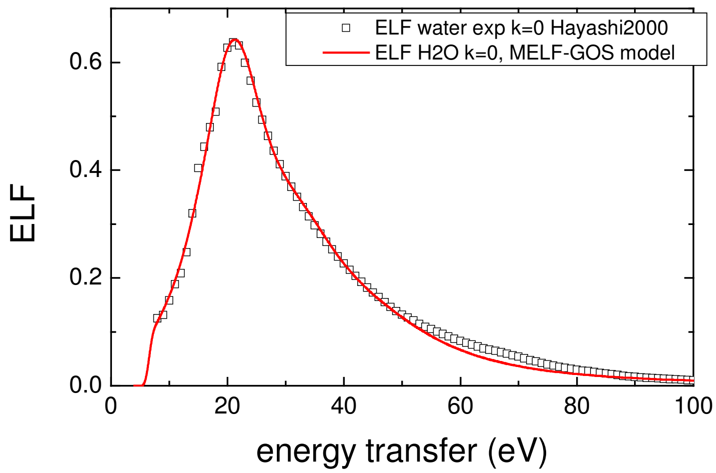

Fig. 1: ELF of liquid water in the optical limit (k=0) as a function of the transferred energy. Solid curves correspond to MELF-GOS model, while symbols represent experimental data [Hayashi 2000] for liquid water. Dashed lines are the results obtained from x-ray scattering factors [Henke1993].

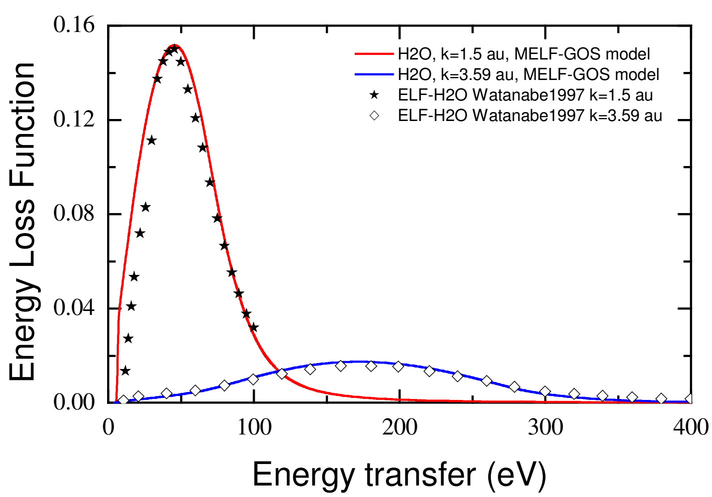

Fig. 2: ELF of liquid water as a function of the transferred energy, for different values of the wave number k.

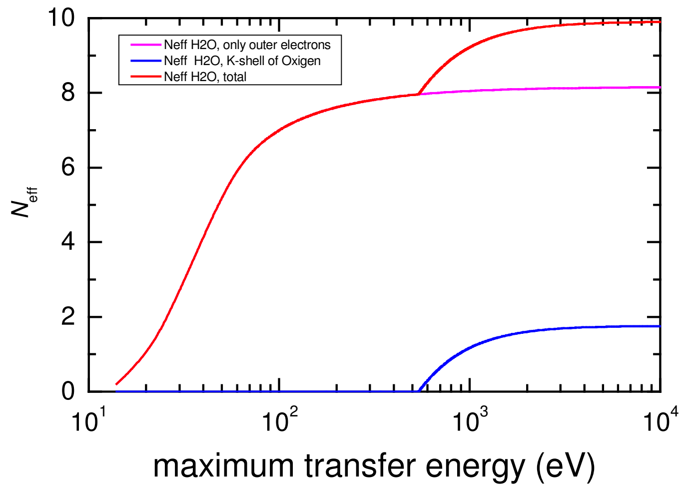

Fig 3: Effective number of electrons for liquid water as a function of the maximum transferred energy.

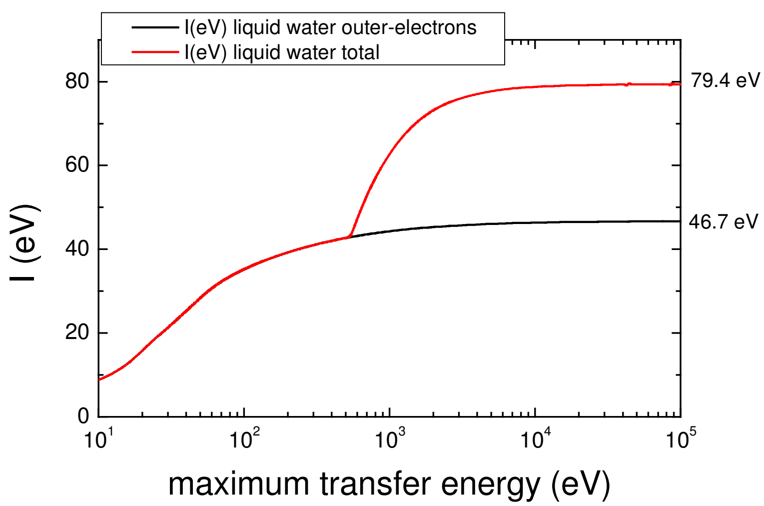

Fig. 4: Mean excitation energy of liquid water as function of the maximum transferred energy.

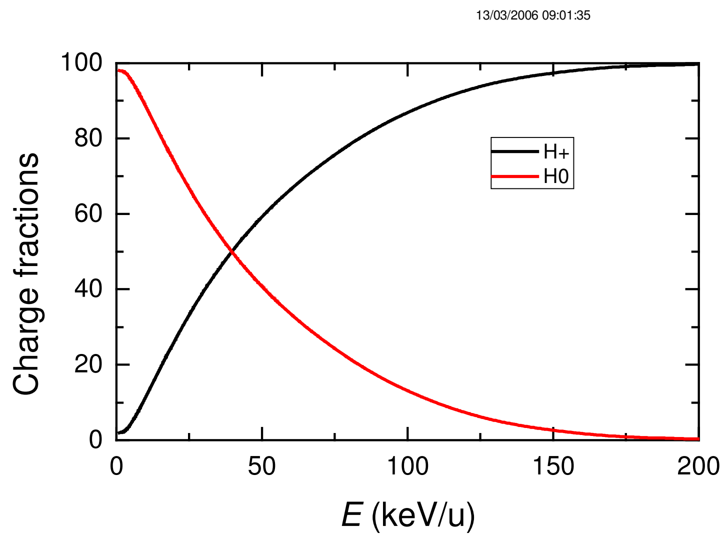

FIG. 5: Equilibrium charge state proton beams in liquid water as a function of the incident energy from the CasP code [Grande 2005].

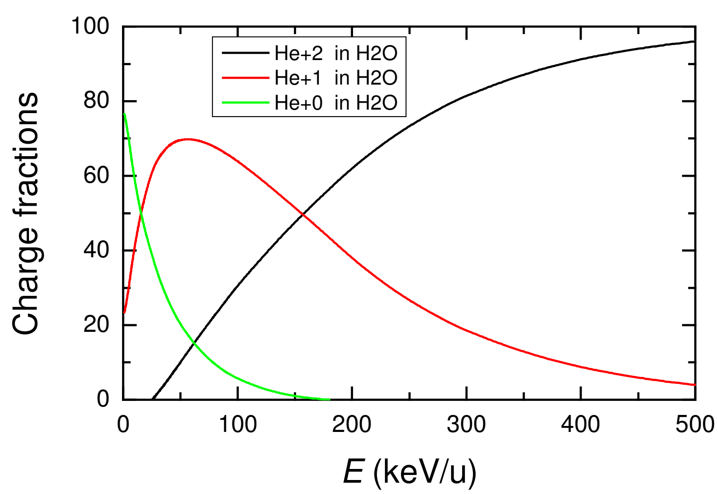

Fig. 6: Equilibrium charge state He ion beams in liquid water as a function of the incident energy from the CasP code [Grande 2005].

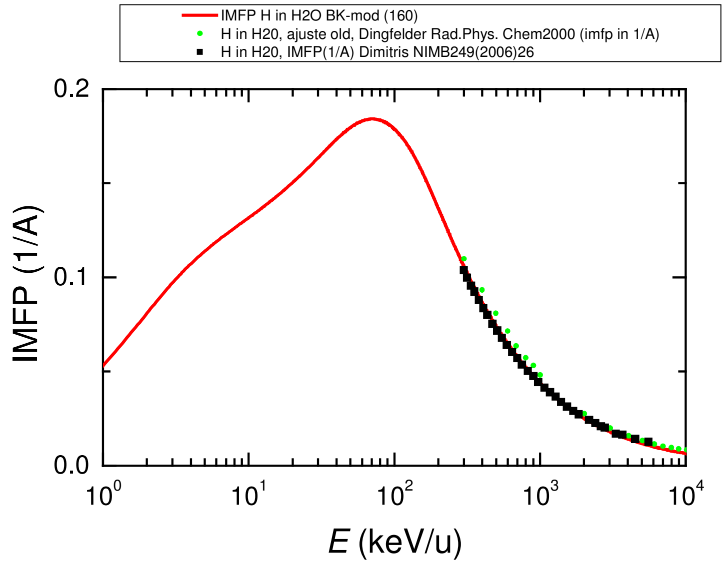

Fig. 7: IMFP of H ion beams in liquid water as a function of the incident energy. We compare with other theoretical results [Dingfelder 2000, Emfietzoglou 2006].

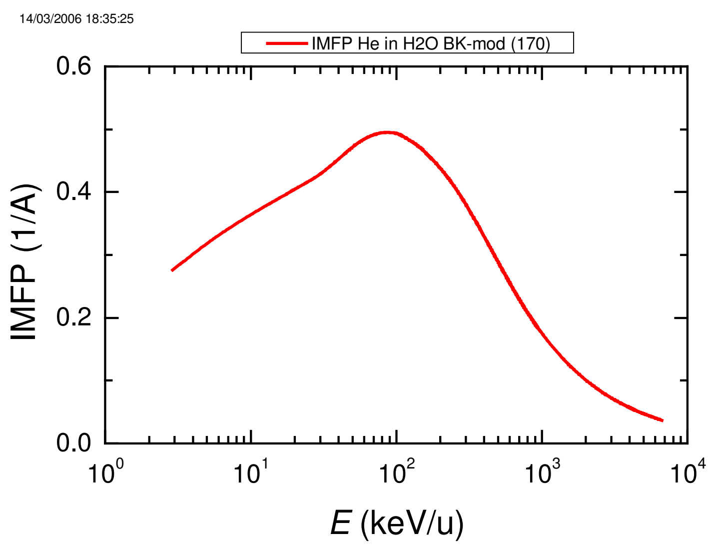

Fig. 8: IMFP of H ion beams in liquid water as a function of the incident energy.

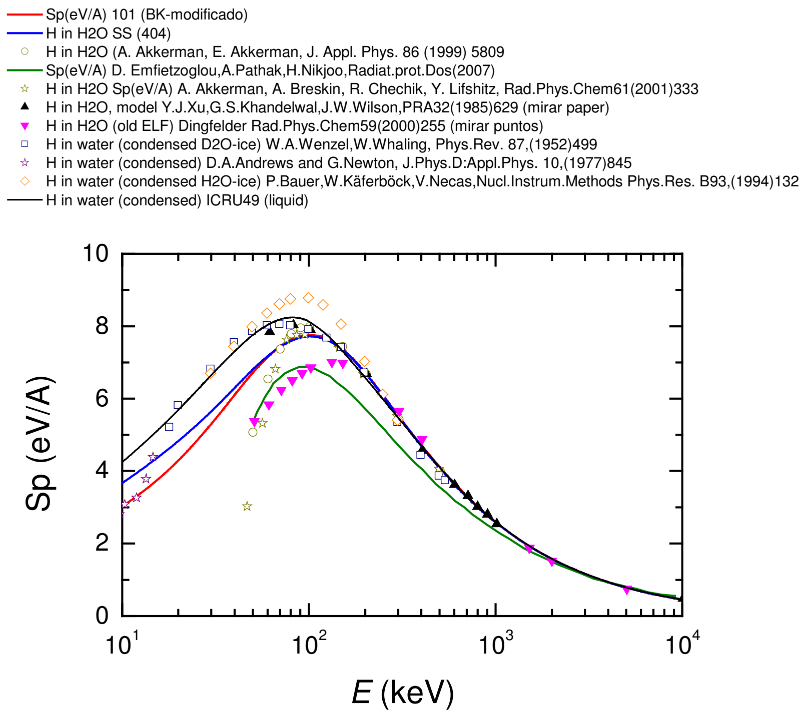

Fig. 9: Stopping power of proton beams in liquid water as a function of the incident projectile energy. The solid curve corresponds to our model, which is compared to experimental and theoretical calculations (see inset).

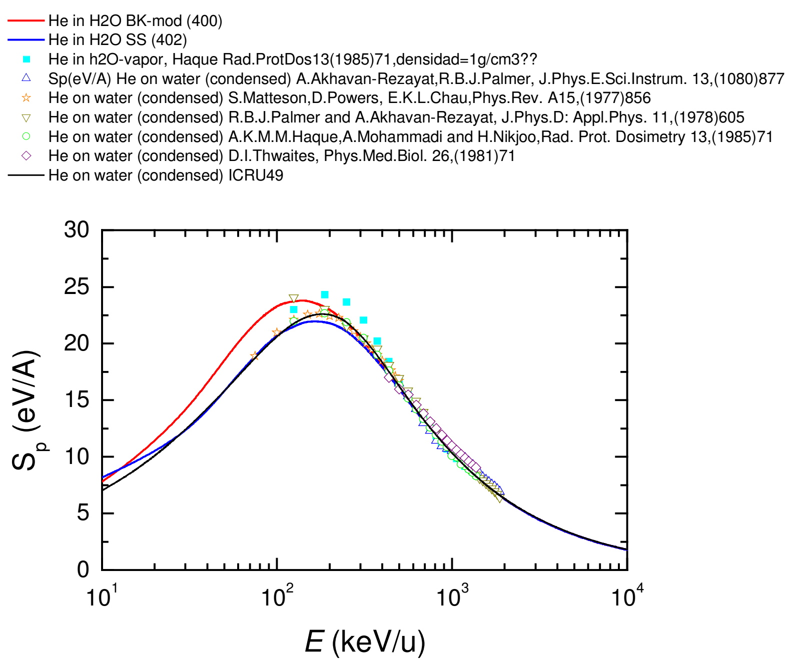

Fig. 10: Stopping power of He beams in liquid water as a function of the incident energy. The solid curve corresponds to our model, which is compared to experimental data (see inset).

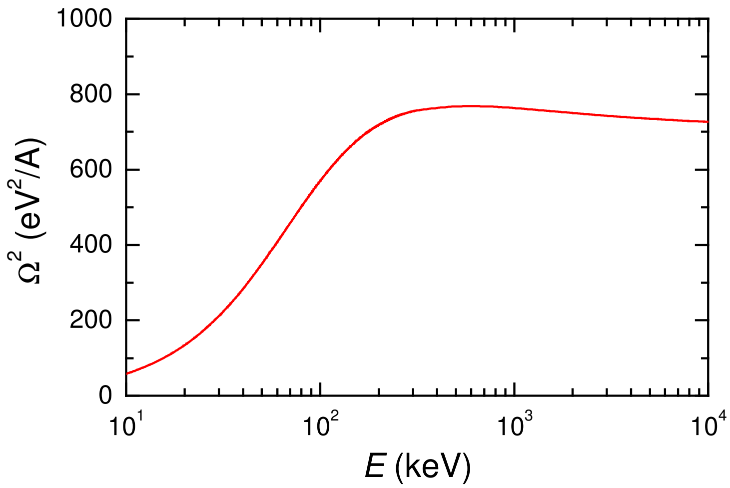

Fig. 11: Energy loss straggling of H beams in liquid water as a function of the incident energy. No experimental data is available for comparison.

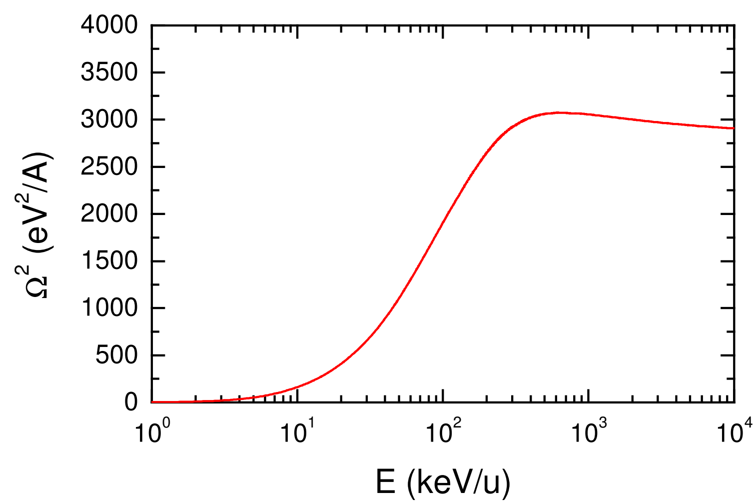

Fig. 12: Energy loss straggling of He beams in liquid water as a function of the incident energy.

Acknowledgments

This work has been financially supported by the Spanish Ministerio de Educación y Ciencia (Contract Nos. FIS2006-13309-C02-01 and FIS2006-13309-C02-02). C.D.D. thanks the Spanish Ministerio de Educación y Ciencia for support under the Ramón y Cajal Program.

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