

INELASTIC CROSS SECTIONS FOR ELECTRON INTERACTIONS IN LIQUID WATER*

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CONF-820998--4

DE83 009773

Our task is to develop a set of cross sections for electron inelastic processes in water in the liquid phase suitable for use in a Monte Carlo transport calculation. This has been described in much more detail elsewhere¹ and will only be summarized here. Very little of the experimental data available pertain to liquid water, and it is thus necessary to use some vapor data in arriving at consistent cross sections for the liquid. One of the basic differences between gaseous and condensed media is the existence of collective electronic modes in the latter. We take collective excitations into account implicitly by starting with $-\text{Im}(1/\epsilon) = \epsilon_2 / (\epsilon_1^2 + \epsilon_2^2)$, the so-called energy loss function for charged particles. For a gas, $-\text{Im}(1/\epsilon)$ and ϵ_2 are essentially identical. However, for the condensed phase, the $-\text{Im}(1/\epsilon)$ peak shifts to higher energies.² This is illustrated by the model calculation shown in Fig. 1.

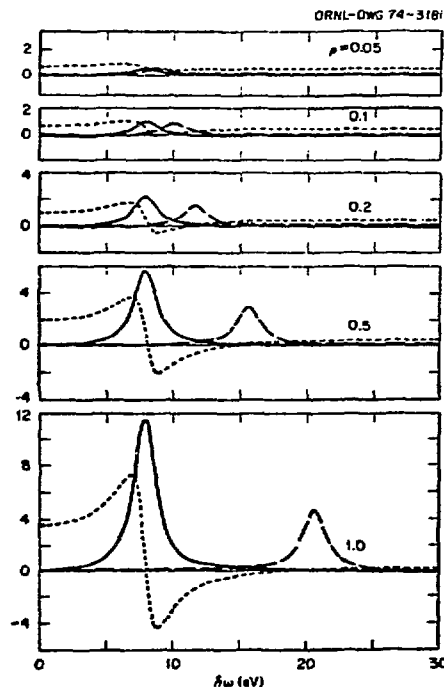


Fig. 1. Dielectric functions ϵ_1 (---) and ϵ_2 (—) and energy-loss function $-\text{Im}(1/\epsilon)$ (- - -) for a single oscillator having $\hbar\omega_1 = 8$ eV, $\hbar\gamma_1 = 2$ eV, and $f_1 = 1.0$ for various densities ρ .

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* Research sponsored by the Office of Health and Environmental Research, U.S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation.

The total differential inverse mean free path (DIMFP) for any kind of inelastic interaction may be written

$$\frac{d\mu}{d\omega} = \frac{1}{\pi E} \int_{q^-}^{q^+} \frac{dq}{q} \operatorname{Im} \left(\frac{-1}{\epsilon(q, \omega)} \right), \quad (1)$$

where E is the primary electron energy, ω the energy loss, and q^{\pm} are the kinematic limits on momentum transfer (all quantities are in atomic units).

Optical measurements on liquid water by Heller *et al.*³ have determined $\epsilon(\omega, 0)$ up to $\omega = 26$ eV. These data are combined with those on the molecule,^{4,5} constrained by sum rules on the total oscillator strength. These data are shown in Fig. 2. In order to extend ϵ into the ω - q plane, we make use of the fact that for large q the electron should behave as if initially free and at rest (the Bethe ridge). We fit the optical data using a sum of derivative Drude functions in the form

$$\epsilon_2(\omega, 0) \approx \omega_p^2 \sum_n \frac{2f_n \gamma_n^3 \omega^3}{[(E_n^2 - \omega^2)^2 + \gamma_n^2 \omega^2]^2}, \quad (2)$$

where the "resonance energies" E_n , the damping energies γ_n , and the oscillator strengths f_n are taken to be fitting parameters. The plasma energy, $\omega_p = 21.5$ eV for liquid water. We then let $E_n \rightarrow E_n + q^2/2$ for the part of ϵ_2 corresponding to ionization. We assume the part of ϵ_2 which corresponds to excitations does not disperse, but varies with q in a manner similar to the variation of the generalized oscillator strength of the water molecule.⁶ Figure 3 shows a plot of $\operatorname{Im}(-1/\epsilon)$ versus ω for several values of q . Figure 4 displays the quantity $\omega^2 d\mu/d\omega$ versus ω for various values of electron energy as computed from Eq. (1). Theory shows that for large ω , the quantity $d\mu/d\omega$ should approach $Z\omega^2/4E\omega^2$ asymptotically, corresponding to the response of nearly free electrons. For water this asymptotic value is given by $0.1556/E\omega^2$ in atomic units.

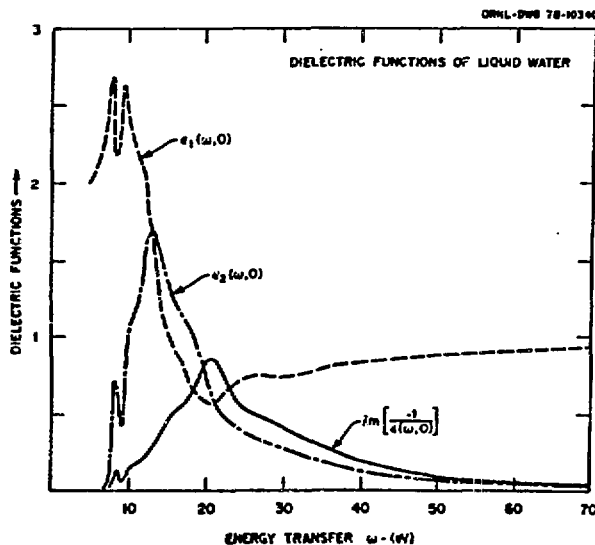


Fig. 2. Optical constants of water as a function of photon energy ω .

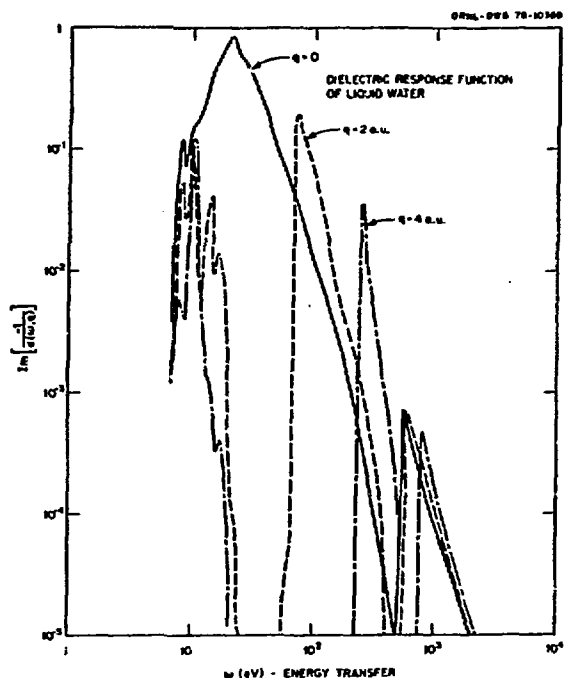


Fig. 3. The energy loss function, $\text{Im}\left[\frac{-1}{\epsilon(\omega, q)}\right]$ plotted against ω for three different values of q .

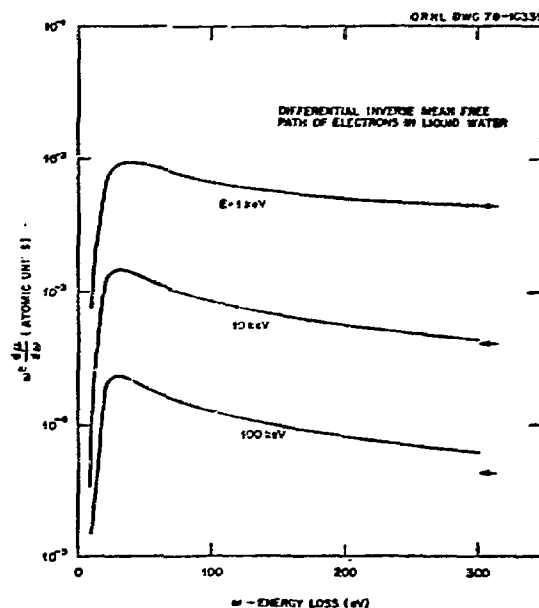


Fig. 4. Energy transfer squared times the DIMFP for all processes versus energy transfer ω for three different primary electron energies. The horizontal arrows mark the asymptotic value for each energy.

The partitioning of ϵ_2 into the $\epsilon_2^{(j)}(\omega, 0)$ fractions is accomplished by using data on the molecule⁴ together with information on the way in which threshold energies for various excitations and ionizations shift from those for the isolated molecule to those for the condensed state. Vacuum UV photoemission experiments with amorphous ice⁷ and with H₂O multilayers adsorbed on gold surfaces at low temperatures⁸ indicate that the threshold energy for exciting an electron from the least tightly bound state to the vacuum level is 8.7 ± 1 eV, corresponding to a band gap of approximately 8 eV. This corresponds well with measured ESCA spectra from ice,⁹ which show a broad threshold at $\sim 7-8$ eV. For the present purposes we take the lowest energy required to ionize electrons in water to be ~ 8 eV. We take partitioning of the ionization probability among the various interband transitions in the liquid to be similar to measured partitioning between the four molecular orbitals in the molecule⁴ which merge to make up the valence bands of the liquid.

Figure 5 shows the partial IMFPs, $\mu^{(j)}(E)$, which are obtained from Eq. (1) by replacing $\epsilon_2(\omega, q)$ by $\epsilon_2^{(j)}(\omega, q)$ in the numerator of the integrand and then by integrating the resultant $d\mu^{(j)}/d\omega$ over ω after making approximate allowance for exchange and relativistic effects. In obtaining the breakdown shown in Fig. 5, we initially made the assumption that

fragmentation is similar to that which occurs in the molecule. At present we assume that all the ionization channels give H_2O^+ , since in the condensed phase the vibrational energy is shared with neighboring molecules before dissociation can occur.

Figure 6 shows the total IMFP, $\mu(E)$, plotted as a function of E . On the same plot we have shown the stopping power of water, $-dE/dx$, as a function of primary electron energy. The open squares show values of $-dE/dx$ recommended by the ICRU.¹⁰ The cross sections developed here, which obviously contain many uncertainties, are subject to continued revision as new data become available.

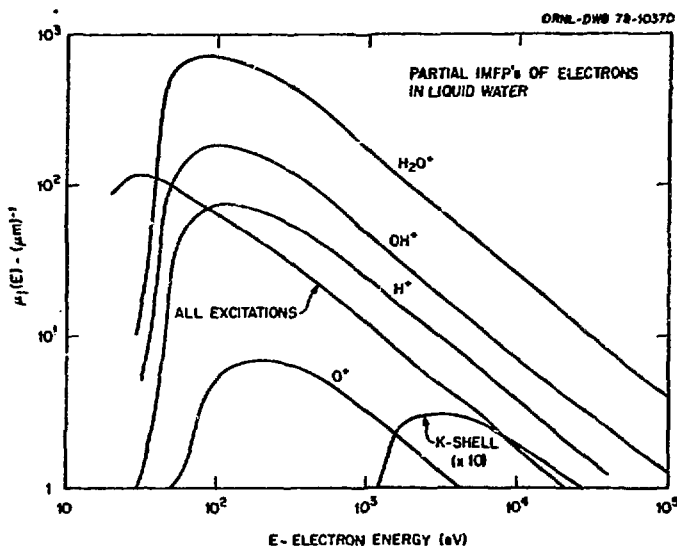


Fig. 5. Partial IMFPs versus electron energy.

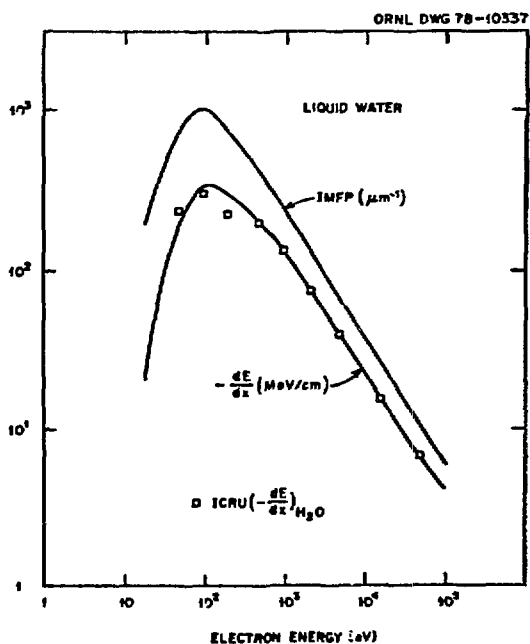


Fig. 6. The curve marked IMFP shows the variation in the total IMFP with primary electron energy. The other curve shows our results for the stopping power of water for electrons of energy E . The points shown represent values recommended in ICRU Report 16 for the stopping power of water.

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