Free electrons: fundamental interactions, applications and data needs

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Abstract In spite of the important recent advances in the field of low-energy electron interactions with atoms and molecules, there still exist fundamental needs and challenges, some of which are generic in their basic and applied significance. These include measurements of cross sections for a number of fundamental low-energy electron–ground-state molecule interaction processes such as vibrational excitation (direct and indirect) and dissociation into neutral fragments, studies of low-energy electron–excited molecule interactions, investigations of low-energy electron interactions with radicals and transient species, and measurements of electron energy losses in electron–molecule collision processes. They also include studies for linking of knowledge on isolated electron–molecule interactions to that in the condensed phases of matter, and the systematic and quantitative exploration of the interactions of low-energy electrons with molecular and macromolecular biological structures.

Key words applications • data • interactions • low-energy electrons • molecules

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Received: 14 November 2002, Accepted: 24 February 2003

Introduction

One hundred and five years have past since J. J. Thomson discovered the electron, the first elementary particle of the microcosmos. Although many other particles have since been discovered, the electron – especially the low-energy electron – still remains one of the most fundamental, abundant, reactive, and challenging particles in nature. In nature, electrons are either bound or free/quasi-free. This paper deals with the latter.

The systematic recent studies of low-energy electronmolecule interactions as a function of the electron energy, the internal energy of the molecule, and the nature, density, and state of the medium in which they occur has provided a wealth of basic knowledge which has advanced our understanding of the structure of atoms and molecules, the phenomena which accompany the interaction of ionizing radiation with matter and the deposition of energy by radiation in matter, the transport of electricity in gases, the interactions of electrons in plasmas, and the behavior of electrons in the condensed phases of matter. Concomitantly, this fundamental knowledge and understanding has underpinned and is underpinning the development of many of today's advanced technologies and is branching out into the environmental and life sciences.

Important as the recent advances have been there are still fundamental needs and challenges in this field, some of which are generic in their nature and in their basic and applied significance. A number of such areas are pointed out and are briefly discussed.



Fig. 1. Cross sections for direct and indirect electron collision processes in CF₄. The symbols $\sigma_{vib,dir,t}$, $\sigma_{dis,neut,t}$, and $\sigma_{i,t}$ refer, respectively, to the total cross sections for direct vibrational excitation, dissociation into neutrals, and ionization of the CF₄ molecule. The symbols $\sigma_{vib,indir,t}$, and $\sigma_{da,t}$ refer, respectively, to the total cross sections for indirect vibrational excitation and dissociative electron attachment. The symbols σ_m , $\sigma_{e,int}$, and $\sigma_{sc,t}$ refer, respectively, to the cross sections for momentum transfer, integral elastic, and total electron scattering (from Ref. [14]).

Low-energy electron-ground-state molecule interactions

Absolute cross sections are now known for many electronmolecule collision processes for a large number of molecules [8, 15, 17, 28]. An example of the type of experimental data available is shown in Fig. 1. The data in this Figure are representative of the basic mechanisms via which energy is transferred from electrons to atoms and molecules and through which new particles are generated in matter. One of the most striking features of these cross-section data is the large magnitude of the cross sections for direct and indirect vibrational excitation of the CF_4 molecule. A consequence of such large cross sections for energy transfer from the electrons to vibrational excitation of



Fig. 2. Cross sections for electron-impact dissociation of the CF₄ molecule into neutrals. ---, $\sigma_{dis,neut,t}$, data of Nagano and Sugai as revised by Sugai *et al.* [35, 43];, $\sigma_{dis,n} - \sigma_{i,t}$, Ref. [15]; \circ , $\sigma_{dis,neut,t}$, Ref. [34]; \times , $\sigma_{dis,neut,t}$, Ref. [30]; --, suggested values, Ref. [15]; •, $\sigma_{dis,n}$, Ref. [46] ---, $\sigma_{i,t}$, Ref. [15] (from Ref. [15]).

molecules is the significant effect of vibrational excitation on the electron-slowing-down properties of gases and of the electron production and loss processes in them when they are electrically stressed, as, for instance, in low-density, low-temperature, non-equilibrium plasmas. However, accurate cross sections for vibrational excitation (direct and indirect) of molecules are very limited and so is our knowledge on the cross sections for other fundamental processes such as the electron-impact dissociation of molecules into neutral fragments. Only limited data are available on the electron-impact dissociation of molecules into neutrals [15, 17], and Fig. 2 shows an example of these data for the case of the CF_4 molecule.

Low-energy electron-excited-state molecule interactions

Quantitative data on low-energy electron attachment to vibrationally excited molecules exist for a number of molecules [16]. However, similar knowledge for electronically excited molecules is meager and qualitative [16]. Oxygen is possibly the only molecule for which the cross sections for dissociative electron attachment to the groundstate molecule, to the vibrationally excited molecule, and



Fig. 3. (a) Comparison of the measured dissociative electron attachment cross sections for ground-state $O_2({}^{3}\Sigma_{g,\nu=0}^{-})$ (data of Ref. [37]), electronically excited (singlet) O_2^* ($a {}^{1}\Delta_{g}$)(data of Ref. [26]), and vibrationally excited $O_2^*({}^{3}\Sigma_{g,\nu=0}^{-})$ oxygen molecules (from Ref. [16]). (b) Schematic potential energy curves for $O_2({}^{3}\Sigma_{g}^{-}), O_2^*$ ($a {}^{1}\Delta_{g}$), and $O_2^* - ({}^{2}\Pi_u)$, and transitions from the ground state $O_2({}^{3}\Sigma_{g,\nu=0}^{-})$ (solid arrows), electronically excited O_2^* ($a {}^{1}\Delta_{g}$) (dotted arrow), and vibrationally excited $O_2({}^{3}\Sigma_{g,\nu=0}^{-})$ (broken arrows) species to the $O_2^* - ({}^{2}\Pi_u)$ negative ion state illustrating the processes of dissociative electron attachment to the ground state, lowest excited electronic state, and vibrationally excited states of O_2 .

to the electronically excited molecule have been accurately measured (Fig. 3). Experimental techniques are needed for quantitative studies of electron attachment to electronically excited molecules, especially since the cross sections for many electron–excited molecule collisions are very large and can have important applications [16].

The limited cross-section data on electron scattering from vibrationally excited molecules show that the cross sections for the excited species are larger than those for the ground state (Fig. 4). Similarly, the cross sections for electron-impact excitation and ionization of electronically excited atoms and molecules far exceed those for the ground state as can be seen from the data for the unexcited and excited Ar atom shown in Fig. 5. Data of this kind are limited for atoms and almost nonexistent for molecules. There is a need for experimental investigations in this area.

Low-energy electron-radical/transient species interactions

With the exemption of electron-impact ionization (e.g., see Tarnovsky *et al.* [44, 45]), there is almost complete lack of experimental data on the other electron-collision processes (e.g., electron attachment, elastic and inelastic electron scattering) for such species [15, 17]. Semiempirical calculations of electron-impact ionization (e.g., see [1, 18,



Fig. 4. Total cross section for electron scattering from the CO₂ molecule in the vibrational ground state (\circ , measurements of Ref. [5]; \Box , measurements of Ref. [23]; —, calculated values of Ref. [33]), and from vibrationally excited CO₂^{*} in the bending mode (\bullet , measurements of Ref. [5]).



Fig. 5. (a) Comparison of the cross section for electron scattering from the excited Ar $(4 {}^{3}P_{2} + 4 {}^{3}P_{2})$ argon atom, and from the unexcited Ar $(3 {}^{1}S_{0})$ argon atom. Total electron scattering cross section of the excited atom, $\sigma_{sc,t}^{*}(\varepsilon) (4 {}^{3}P_{2} + 4 {}^{3}P_{2})$: •, measurements of Ref. [6]. Total elastic electron scattering cross section of the excited atom, $\sigma_{e,t}^{*}(\varepsilon) (4 {}^{3}P_{2})$: •, calculated values of Ref. [38]. Total electron scattering cross section of the ground-state atom, $\sigma_{sc,t}(\varepsilon) (3 {}^{1}S_{0})$: measurements of Ref. [27] (•), Ref. [36] (Δ) , Ref. [22] (∇). Momentum transfer cross section for the ground-state atom, $\sigma_{m}(\varepsilon) (3 {}^{1}S_{0})$: +, measurements of Ref. [32]. (b) Cross section for electron-impact ionization of metastable Ar *, $\sigma_{i}^{*}(\varepsilon)$ (•, Ref. [19]) in comparison with the cross section, $\sigma_{i}(\varepsilon)$, for electron-impact ionization of the ground state Ar (—, measurements of Ref. [29]).

25, 44]) are rather successful in providing total ionization cross sections for radicals. An example [1] of these data is shown in Fig. 6 for a number of radicals generated by electron impact on SF_6 .

Specific processes of generic interest, energy losses, correlations

One fundamental process which needs further attention is the photodetachment of atomic and molecular negative ions of interest to technology (e.g., photodetachment cross sections and energetics for negative ions such as CF^- , CF_2^- , CF_3^- , and $C_2F_5^-$ which are important in plasma diagnostics). In this connection, the method developed by Sowada and Holroyd [41] for photodetachment from negative ions in nonpolar liquids, and the method developed more recently



Fig. 6. Calculated total electron-impact ionization cross sections for the SF_x (x = 1 to 5) radicals (Ref. [1]).



Fig. 7. (a), (b) Schematic diagram illustrating the two-laser photodetachment method of Christophorou and collaborators [11]. (c) Photodetachment cross section, σ_{pd}^{-6} , for the $C_6F_6^{-6}$ ion in gaseous tetramethylsilane (TMS) (\blacksquare , Ref. [11]), and, σ_{pd}^{-L} , in liquid TMS (\circ , Ref. [20]; \bullet , Ref. [42]).



Fig. 8. Dissociative electron attachment cross sections as a function of the electron energy for a number of molecules. The line designated by $\pi \lambda^2$ is the *s*-wave capture cross section (from Ref. [13]).

by Christophorou and collaborators [11] for the measurement of the photodetachment cross sections and the photodetachment onsets in gases and liquids may be of interest. The principle of the method of Christophorou and collaborators is shown in Fig. 7, and an example of the results obtained is given in the lower part of Fig. 7 for the case of $C_6F_6^-$ in the gaseous and in the liquid state of matter.

Furthermore, besides the need of cross-section data for the various electron collision processes, data are needed (e.g., in computational models) for the associated electron energy losses, that is, for the energy which is transferred by the electron to the molecule in each individual electron– molecule collision process.

Systematic studies are necessary to establish correlations of, and trends in, the data with predictive capability (e.g., see Fig. 8 for electron attachment).

Linking knowledge on the isolated low-energy electronmolecule interactions to that in the condensed phase

Until recently, most of the fundamental understanding on low-energy electron-molecule interactions has been derived from studies on electron interactions with isolated molecules (i.e., from studies on electron interactions with molecules in low pressure gases). Considerable progress has been made in recent years [12] in unraveling and quantifying the effects on electron-molecule interactions of the nature, density, and state of the medium in which these interactions occur. A number of methods have been devised which provide most useful information on lowenergy electron-molecule interactions in low-, high-, and very-high-pressure gases, in clusters, at surfaces, and in



Fig. 9. Schematic illustrations of electron-molecule interactions in low-, high-, and very-high-pressure gases, in clusters, at surfaces, and in condensed matter.

condensed matter (see Fig. 9 for a schematic illustration of these environments). For instance, it has been established that negative ion resonances occur in all states of matter. Figure 10 shows the 2.3 eV negative ion state of N_2 as it has been observed in solid N_2 [40]. In general, while negative



Fig. 10. Relative cross section for indirect scattering of low-energy electrons via the $N_2^{-*}(^2\Pi_g)$ negative ion state of N_2 in solid N_2 film leaving the N_2 molecule excited in the first vibrational level. The broken vertical lines indicate the positions of the peaks in the cross section as measured in the film, and the solid vertical lines indicate the corresponding positions as measured in the gas (based on data in Ref. [40]).



Fig. 11. Electron attachment rate constant for the production of O⁻ from N₂O in gaseous, $(k_a)_G$, and in liquid Ar, $(k_a)_L$, plotted as a function of the respective, $\langle \varepsilon \rangle_G$ and $\langle \varepsilon \rangle_L$, mean electron energies (from Ref. [10], the $(k_a)_L$ data are from Ref. [3]).

ion resonances occur in all molecules and in all states of matter, the medium introduces significant changes in the energetics of their formation, as well as in other of their physical properties such as their lifetimes and decay channels [10].

Table 1. Approximate relations between the energetics of photoionization, photodetachment, and electron attachment in the gaseous and the liquid state of matter.

Process*	Approximate relation ^{+,#}
$\label{eq:main_state} \hline \\ \hline Photoionization \\ M_{\rm G} + nhv \rightarrow M_{\rm G}^+ + e_{\rm G} \\ M_{\rm L} + nhv \rightarrow M_{\rm L}^+ + e_{\rm L} \\ \hline \end{array}$	$\begin{matrix} \mathbf{I}_{\mathrm{G}} \\ \mathbf{I}_{\mathrm{L}} = \mathbf{I}_{\mathrm{G}} + \mathbf{P}^{+} + \mathbf{V}_{\mathrm{0}} \end{matrix}$
Photodetachment $M_{G}^{-} + hv \rightarrow M_{G} + e_{G}$ $M_{L}^{-} + nhv \rightarrow M_{L} + e_{L}$	$ (E_{th})_G = VDE_G = EA_G + \Delta E_G (E_{th})_L = VDE_L = EA_G + \mathbf{V}_0 - \mathbf{P}^- + \Delta E_L (EA_L \cong EA_G + \mathbf{V}_0 - \mathbf{P}^-) $
Electron attachment $M_{\rm G} + e_{\rm G} \rightarrow M_{\rm G}^{-*}$ $M_{\rm L} + e_{\rm L} \rightarrow M_{\rm L}^{-*}$	$\begin{aligned} VAE_{G} &\approx (E_{NIS})_{G} = -VEA_{G} \\ VAE_{L} &\approx (E_{NIS})_{L} \approx (E_{NIS})_{G} - (V_{0} - P_{NIS}) \end{aligned}$

^{*} The notation is as follows: e_G , M_G , M_G^- , $M_G^-^*$, and M_G^+ refer, respectively, to the electron, molecule, negative ion, transient negative ion, and positive ion in a low pressure gas. e_L , M_L , M_L^- , $M_L^-^*$, and M_L^+ refer, respectively, to the same quantities in the liquid. The symbol *nhv* represents *n* number of photons.

¹+ The notation I_G , $(E_{th})_G$, EA_G , VEA_G , VAE_G , $(E_{NIS})_G$, and VDE_G refers, respectively, to the ionization threshold energy, detachment threshold energy, adiabatic electron affinity, vertical electron affinity, vertical attachment energy, energy of the negative ion state, and vertical detachment energy in a low pressure gas. I_L , $(E_{th})_L$, EA_L , VEA_L , VAE_L , $(E_{NIS})_L$, and VDE_L refer, respectively, to the same quantities in the liquid. ΔE is a quantity which depends on the relative positions of the potential energy curves (surfaces) of M and M⁻.

[#] V_0 is the ground-state energy of the electron in the medium and P^{+,-} is the polarization energy of the positive or negative ion in the medium and is normally approximated by the Born expression P^{+,-} = $-e^2/[(2R)(1 - 1/\varepsilon_{opt})]$, where *R* is the effective radius of the ion cavity and ε_{opt} is the optical dielectric constant of the medium.



Fig. 12. (a) Electron drift velocity w(E/N) in liquid [31] and gaseous [24] Xe (from Ref. [10]). The inset compares the elastic momentum transfer cross section in gaseous ($\sigma_{m,G}(\varepsilon)$) and in liquid ($\sigma_{m,L}(\varepsilon)$) xenon; $\sigma_{0,L}(\varepsilon)$ is the cross section for elastic energy loss [39]. (b) Electron drift velocity w(E/N) in liquid and gaseous TMS (from Refs. [10, 21]).

E/N (10⁻¹⁷ V cm²)

With regard to the energetics of electron interactions in various media, the critical role of the polarization energy has been demonstrated from studies on the energetics of electron attachment, photodetachment, and photoionization. Figure 11 shows one such example for the case of dissociative electron attachment to N₂O producing O⁻. The



Fig. 13. Mean electron energy in gaseous, $\langle \epsilon \rangle_G$, and liquid, $\langle \epsilon \rangle_L$, Ar (from Ref. [9]).



Fig. 14. (Upper part) Measured yields, per incident electron, for the induction of (a) double-strand breaks (DSBs), (b) single-strand breaks (SSBs), and (c) loss of supercoiled DNA, in DNA solid films by low-energy electron irradiation, as a function of the incident electron energy. (Lower part) Electron damage to condensed films of (A) thymine, (B) water, and (C) tetrahydrofurfuryl alcohol exemplified by the yield of H⁻ desorption as a function of the incident electron energy (from Ref. [4]).

cross section for the production of O⁻ by dissociative electron attachment to N₂O is much larger when the reaction takes place in liquid argon as opposed to gaseous Ar. The shift of the resonance to lower energy is due to the polarization energy of the medium and is common for other reactions involving negative ion states, and chargeseparated and quasi-charge-separated states. This has been indicated by studies on molecular clusters and by comparisons of measurements in the gaseous, the condensed, and the interphase states of matter.

Table 1 shows the approximate energetic relations that have been indicated from studies of the energetics of electron attachment, photodetachment, and photoionization of molecules in gases and liquids.

In addition, it has clearly been shown that electrons can have high mobilities in nonpolar liquids with negative V_0 values. Figure 12 shows this for liquid Xe and liquid TMS (tetramethylsilane). Furthermore, as in gases, quasi-free electrons in electrically stressed rare gas liquids can have energies considerably in excess of thermal (Fig. 13).

In spite of this progress, this field remains a virtually unexplored territory of utmost importance. Knowledge of the behavior of slow electrons in all states of matter and the region between the phases (interphase physics/clusters) will allow us to link the abundant knowledge in the gaseous phase with the scarce knowledge in the condensed phases of matter. Such unification of our knowledge on the fundamental electron-molecule interactions in all states of matter is of basic and applied significance and a prerequisite of bringing physics closer to biology.

Low-energy electron interactions with molecular and macromolecular structures of biological interest

There is a need to develop a physicochemical foundation of biological action. To accomplish this goal, low-energy electron and ion physics must be brought closer to biological materials and environments.

Efforts along these lines trace many years. For instance, in my 1971 book [7] I have indicated the possibility of establishing a toxicity index for molecules based on their electron-capture rate constants, which later Bakale and collaborators [2] have shown possible. The role of slow electrons in radiation damage has been conjectured during at least the last 50 years, and has recently been indicated by experiments [4] (see Fig. 14). The field is however virgin and the physics of life sciences is wide open and challenging. The low-energy electron and its interactions with molecules lie at the very basis of the physicochemical foundation of life sciences, which is yet to be developed.

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