Comments

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Comment on the classical-trajectory Monte Carlo method for ion-atom collisions

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It is shown that the procedure described by Olson and Salop for classical-trajectory Monte Carlo treatment of ion-atom collisions does not provide a uniform statistical distribution of all the parameters defining the initial conditions of a trajectory. Impact-ionization and charge-transfer cross sections for collisions of H^+ with H at H energies between 25 and 600 keV are recalculated eliminating this failing and compared with those obtained using the procedure of Olson and Salop and with experimental results.

In recent years, the classical-trajectory Monte Carlo (CTMC) method has been used in a number of calculations of impact-ionization and charge-transfer cross sections, as well as other characteristics, for collisions of stripped ions with the hydrogen atom. Most of these calculations have used the procedure described in detail by Olson and Salop (OS).[1] The purpose of this Comment is to point out that the procedure does not statistically distribute all of the parameters defining the initial conditions of the trajectory, as required by the Monte Carlo method.

In the CTMC approach Hamilton's classical equations of motion for the three-body system are solved numerically for a statistically large number of trajectories with initial conditions determined pseudorandomly. The dependent variables of the 12 coupled first-order differential equations in the center-of-mass frame are the coordinates and momenta of internal (electron-proton in the hydrogen atom) motion and relative (ion-atomic center-of-mass) motion. The initial relative motion is specified by the asymptotic velocity, asymptotic distance, and an impact parameter randomly distributed in the interval

$$0 \leq b^2 \leq b_{\max}^2 \ . \tag{1}$$

The orientation of the initial relative motion is arbitrary, say, in the z direction in the y-z plane, thereby completing specification of the six relative variables. The spherically symmetric ground-state hydrogen atom is represented by a microcanonical distribution.[2] This state is specified by the binding energy of the electron in the target atom and five additional parameters randomly distributed in the following ranges:

$$-\pi \leq \phi \leq \pi, \quad -1 \leq \cos\theta \leq 1, \quad -\pi \leq \eta \leq \pi \ , \quad 0 \leq \epsilon^2 \leq 1, \text{ and } 0 \leq \theta_n \leq 2\pi \ . \tag{2}$$

Here, ϵ is the eccentricity of the orbit, θ_n is a parameter of the orbit proportional to time, and ϕ, θ, and η are Euler angles. A random distribution of θ_n corresponds to equal probability of the atom having any phase in its periodic motion. The eccentric angle u is more geometrically useful than θ_n and is determined by solving Kepler's equation[3]

$$\theta_n = u - \epsilon \sin u \ . \tag{3}$$

It is in the treatment of u and ϵ that the procedure of OS errs. That something is awry can be seen from the fact that the initial conditions of OS depend only on the product $\epsilon \cos u$ and not on the two parameters independently. The reason for this loss of an initial degree of freedom will be shown by going through the initialization of the internal coordinates and momenta in detail.

The atomic initialization may be thought of as occurring in three steps: (1) placing the orbit of eccentricity ϵ in some arbitrary orientation, say, in the y-z plane, with the major axis in the z direction ; (2) locating the "particle" at the eccentric angle u on the orbit; and (3) performing the rotation specified by the Euler angles ϕ, θ, and η. Hence the initial internal coordinates and momenta are given by

$$\vec{C}^0 = \underline{A}\,\vec{C}_0^0 \quad \text{and} \quad \vec{P}^0 = \underline{A}\,\vec{P}_0^0 \ , \tag{4}$$

where

$$\vec{C}_0^0 = \begin{pmatrix} 0 \\ a(1-\epsilon^2)^{1/2}\sin u \\ a(\cos u - \epsilon) \end{pmatrix}, \quad a = Z/2U \ , \tag{5}$$

$$\vec{P}_0^0 = \begin{pmatrix} 0 \\ b(1-\epsilon^2)^{1/2}\cos u/(1-\epsilon\cos u) \\ -b\sin u/(1-\epsilon\cos u) \end{pmatrix}, \quad b = (2mU)^{1/2} \ , \tag{6}$$

and, consistent with the convention of OS,

$$\underline{A} = \begin{pmatrix} -\sin\phi\sin\eta + \cos\phi\cos\theta\cos\eta & -\sin\phi\cos\eta - \cos\phi\cos\theta\sin\eta & \cos\phi\sin\theta \\ \cos\phi\sin\eta + \sin\phi\cos\theta\cos\eta & \cos\phi\cos\eta - \sin\phi\cos\theta\sin\eta & \sin\phi\sin\theta \\ -\sin\theta\cos\eta & \sin\theta\sin\eta & \cos\theta \end{pmatrix} . \tag{7}$$

In the above, Z is the nuclear charge of the atom, and U and m are the binding energy and reduced mass of the electron in the atom.

In effect, instead of Eqs. (5) and (6), OS use

$$(\vec{C}_0^0)_{OS} = \begin{pmatrix} 0 \\ 0 \\ |\vec{C}_0^0| \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ a(1-\epsilon\cos u) \end{pmatrix} \tag{8}$$

and

$$(\vec{P}_0^0)_{OS} = \begin{pmatrix} 0 \\ |\vec{P}_0^0| \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ b(1-\epsilon^2\cos^2 u)^{1/2}/(1-\epsilon\cos u) \\ 0 \end{pmatrix} . \tag{9}$$

Applying the rotation (7) to (8) and (9) yields Eqs. (6) and (7) of Ref. 1. Comparison of (5) and (6) with (8) and (9) shows that the eccentricity of the orbit specified by Eqs. (8) and (9) is not ϵ but is actually

$$\epsilon_{OS} = \epsilon|\cos u| , \tag{10}$$

and that the particle always starts at an extremum of the orbit, the perigee ($u_{OS}=0$) if $\cos u > 0$ and the apogee ($u_{OS}=\pi$) if $\cos u < 0$. Equation (3) [Eq. (8) in Ref. 1] has no other effect in the OS procedure. Hence the eccentricity distribution is skewed in favor of circular (i.e., higher angular momentum) orbitals and all points except the orbital extrema are excluded as initial conditions.

The question now arises as to the consequence of this nonrandom bias. To elucidate this question, cross sections have been computed for collisions of H^+ with H using both procedures. For $E_H \leq 100$ keV, 4000 trajectories were run, and for $E_H > 100$ keV, 6000 trajectories were run. These numbers yield ionization cross sections with standard statistical errors of 4–6%. The results are shown in Fig. 1. The OS procedure yields a somewhat too large impact-ionization cross section at intermediate energies, having an anomalous energy dependence in the range $150 \lesssim E \lesssim 400$ keV. This peculiarity is not particularly noticeable in the figures of Ref. 1 since those calculations stopped at 200 keV. The ionization cross section obtained with the procedure utilizing Eqs. (5) and (6) exhibits the classical $1/E$ dependence at $E_H \gtrsim 150$ keV, whereas the cross section obtained with the OS procedure only exhibits this dependence at considerably higher energies where the two procedures yield similar results. In the case of charge transfer in collisions of H^+ with H, the cross section obtained with the OS procedure appears to be relatively too large at the lowest calculated energies

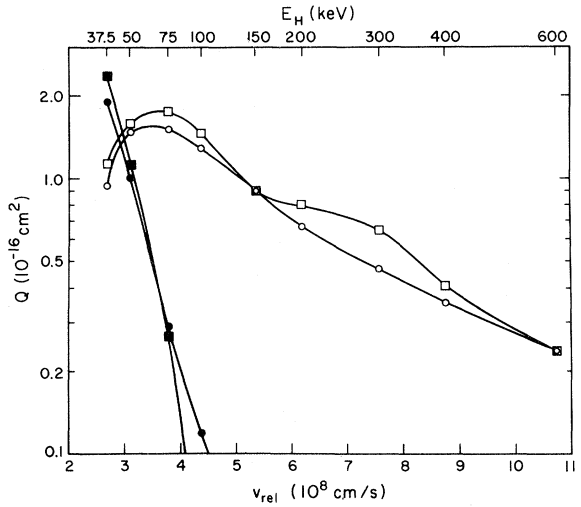

FIG. 1. Impact-ionization cross sections (open circles are present procedure; open squares are procedure of Ref. 1) and charge-transfer cross sections (solid circles are present procedure; solid squares are procedure of Ref. 1) for collisions of H^+ with H.

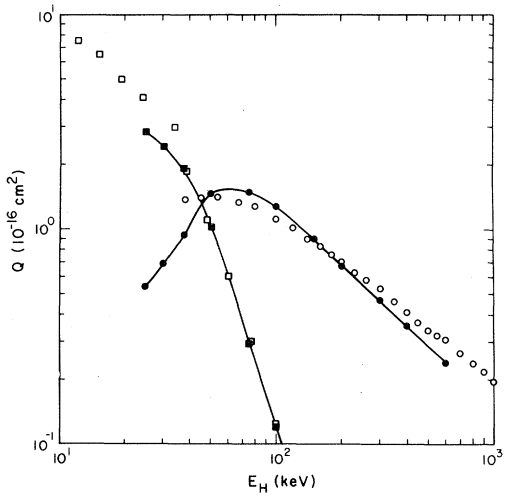

FIG. 2. Comparison of CTMC impact-ionization cross section (solid circles) with experimental results of Shah and Gilbody (open circles), and comparison of CTMC charge-transfer cross section (solid squares) with experimental results of McClure (open squares).

but to fall off too rapidly at higher energies. Comparison of the corrected CTMC results[4] with experimental results is made in Fig. 2. The CTMC impact-ionization cross section is within about 15% of the recent experimental determination[5] at energies $50 \lesssim E_H \lesssim 500$ keV; at higher energies the classical $1/E$ dependence obtains and falls off faster than the quantum-mechanical $(\log E)/E$ dependence. The CTMC charge-transfer cross section is in excellent agreement with experiment[6] at energies $E_H \gtrsim 37.5$ keV; the agreement in the high-energy tail appears to be significantly better than in Ref. 1.

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[3]E.g., L. D. Landau and E. M. Lifshitz, *Mechanics,* 3rd ed. (Pergamon, Oxford, 1976), pp. 35–39.
[4]The present results agree with D. Banks, K. S. Barnes, and J. McB Wilson, J. Phys. B 9, L141 (1976).
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