Threshold Law for Electron-Atom Impact Ionization

A. Temkin

Atomic Physics Office, National Aeronautics and Space Administration, Goddard Space Flight Center, Greenbelt, Maryland 20771

(Received 22 February 1982; revised manuscript received 10 June 1982)

The threshold law for electron-atom ionization is derived on the basis of the Coulomb-dipole theory. The result is a modulated quasilinear law for the yield: $\mathcal{Q} \propto E(\ln E)^{-2}[1 + C\sin(\alpha \ln E + \mu)]$. The derivation depends on a more accurate description of the dipole moment seen by the outer electron as the distance of the inner electron from the nucleus. The derivation also implies $C \cong \alpha^{-1}$, and it also suggests that α is large. The same law also applies to positron-atom impact ionization.

PACS numbers: 34.80.Dp, 32.80.Fb

Impact ionization is one of the most fundamental ongoing problems of nonrelativistic quantum mechanics. I would like to present here a derivation of the explicit form of the threshold law for electron impact ionization of atoms (i.e., neutral targets). After a long period of gestation[1] we were led to the conclusion that the threshold is dominated by the Coulomb-dipole region[2]: i.e., the region where the outgoing electrons have very different energies, corresponding to the inner (slower) electron seeing the charge of the residual ion directly whereas the outer (faster) electron sees the dipole potential formed by the residual ion and the inner electron. This led to the conjecture,[2,3] which I emphasize was heretofore not a derived result, that the threshold law (i.e., the yield of positive ions) should be of the form of a modulated linear law (E is the available energy after ionization) $\mathcal{Q} \propto EM(E)$. Very recently I have derived[4] that formula on the basis of a Coulomb-dipole *Ansatz* for the final-state wave function. The dipole function used therein[4] had a dipole moment which was taken as constant, whereas in actual fact the inner electron is not stationary; thus the dipole moment seen by the outer electron should be a function of the inner electron's coordinate. In the present Letter I shall therefore make the important generalization of using a dipole function whose moment is the dipole moment formed by inner electron and the nucleus. This will have a small but important quantitative alteration in the final result. But first a few preliminaries.

The yield of positive ions is given by the expression

$$\mathcal{Q} = \int |\mathfrak{M}|^2 \delta(E - k_1^2 - k_2^2) d^3k_1 d^3k_2. \tag{1}$$

In Eq. (2) the matrix element $\mathfrak{M}$ is to be taken in final-state form,

$$\mathfrak{M} = \langle \Psi_f | V_i | \Phi_i \rangle . \tag{2}$$

In what follows I shall confine myself to S-wave scattering in the electron-hydrogen system and specifically the two-dimensional model[2] in the dipole approximation (Rydberg units throughout),

$$V_i = -2/r_1 + 2/(r_1 + r_2) \cong -2r_2/r_1^2, \quad r_1 \geq 2r_2, \tag{3}$$

with hydrogen in its ground state in the initial-state function

$$\Phi_i = j_0(k_i r_1)\varphi_0(r_2). \tag{4}$$

[Our argument also applies to two-electron photodetachment (of H^-) in which case Φ_i is the ground-state H^- wave function, V_i would be, say, the dipole length operator, and the final state Ψ_f would consist of only one partial wave. Cf. after Eq. (19).]

None of these circumscriptions should affect the form of the threshold law.[2] That will be determined by the final-state wave Ψ_f which in principle should be an appropriate, exact solution of the Schrödinger equation. The essence of all these arguments[2,3] is that for the purposes of deriving $\mathcal{Q}(E)$ the essential part of Ψ_f is given by

$$\Psi_f = F^{(d)}(r_1)F^{(c)}(r_2), \quad r_1 \geq 2r_2, \tag{5}$$

where $F^{(c)}$ is a correctly normalized zero-energy Coulomb wave describing the inner electron:

$$\lim_{k_2 \to 0} F^{(c)} = k_2^{-1/2}[r_2^{-1/2} J_1((8r_2)^{1/2})]. \tag{6}$$

[The inequality $r_1 \geq 2r_2$ represents "authentically greater than" (i.e., larger than by about a factor 2). $\mathfrak{M}$ also contains contributions from other regions of configuration space, but I assume that they contribute less to the threshold law than what we do include.]

The outer electron is described by a dipole wave

$$F^{(d)}(r_1) = \eta(k_1) r_1^{-1/2}[A J_{i\alpha}(k_1 r_1) + B N_{i\alpha}(k_1 r_1)]. \tag{7}$$

In Eqs. (6) and (7) the J and N are Bessel and Neumann functions, and in the latter they are of imaginary order $i\alpha$, where

$$\alpha = (b - \tfrac{1}{4})^{1/2}, \tag{8a}$$

and b is the dipole moment seen by the outer electron. $F^{(d)}$ satisfies

$$\left[\frac{d^2}{dr_1^2} + \frac{b}{r_1^2} + k_1^2\right][r_1 F^{(d)}(r_1)] = 0. \tag{9}$$

Asymptotically $F^{(d)}$ has the normalization of a plane wave,

$$\lim_{r_1 \to \infty} F^{(d)}(r_1) = (k_1 r_1)^{-1} \sin(k_1 r_1 + \text{const}),$$

as is demanded by Eq. (1). As long as $k_1^2 \ll b/r^2$, $F^{(d)}$ reduces to

$$F^{(d)}(r_1) \cong \eta(k_1) r_1^{-1/2} \sin(\alpha \ln r_1 + \phi). \tag{10}$$

The phase ϕ depends logarithmically on k_1 where $k_1^2 = \epsilon$ is the energy of the outer electron:

$$\phi = \alpha \ln \epsilon^{1/2} + \text{const}. \tag{11}$$

For the purposes of this derivation, the normalization $\eta(k_1)$ of the dipole wave[2] reduces to that of a pure Coulomb wave:

$$\eta(k_1) = k_1^{-1/2}. \tag{12}$$

The essence of this present (new) derivation is to take into account the fact that the dipole moment seen by the outer electron is precisely the distance of the inner electron from the nucleus, i.e.,

$$b = r_2; \quad \alpha(r_2) = (r_2 - \tfrac{1}{4})^{1/2}. \tag{8b}$$

Under these circumstances, Ψ_f can be written

$$\Psi_f = F_{\alpha(r_2)}^{(d)}(r_1) F^{(c)}(r_2). \tag{5b}$$

$F^{(d)}$ is now a highly nonseparable function of r_1 and r_2; one can nevertheless carry out the corresponding matrix element to leading order in k and ultimately derive the threshold law explicitly. Briefly, substituting all functions into (2), with $k_i = 1$ in Eq. (4), one finds

$$\mathfrak{M} \propto (k_1 k_2)^{-1/2} \int_{2R}^{\infty} dr_1 \int_R^{r_1/2} dr_2 \sin[\alpha(r_2)\ln k r_1 + c_1] r_1^{-3/2} \sin r_1 r_2^{3/2} J_1((8r_2)^{1/2}) \varphi_0(r_2). \tag{2b}$$

R is the lower limit of the r_2 coordinate below which (5b) is not an accurate approximation to Ψ_f; I shall discuss it below. Interchanging the order of integration in (2b),

$$\int_{2R}^{\infty} dr_1 \int_R^{r_1/2} dr_2 = \int_R^{\infty} dr_2 \int_{2r_2}^{\infty} dr_1, \tag{13}$$

we find, writing $\mathfrak{M} = (k_1 k_2)^{1/2} I$,

$$I = \int_R^{\infty} e^{-r} r^{3/2} J_1((8r)^{1/2}) \cos[\alpha(r)\ln k_1] \mathcal{S}(r) dr + \int_R^{\infty} e^{-r} r^{3/2} J_1((8r)^{1/2}) \sin[\alpha(r)\ln k_1] \mathcal{C}(r) dr, \tag{14a}$$

where

$$\begin{Bmatrix} \mathcal{S}(r) \\ \mathcal{C}(r) \end{Bmatrix} = \int_{2r}^{\infty} \frac{\sin r_1}{r_1^{3/2}} \begin{Bmatrix} \sin \\ \cos \end{Bmatrix} [\alpha(r)\ln r_1 + c_1] \Bigg\} dr_1. \tag{14b}$$

The object here is to find the leading order dependence in (14a) on k_1; note that Eqs. (14b) are independent of k_1, and thus we can integrate (14a) by parts, by taking $dV = r^{-1/2} \sin(r^{1/2} \ln k_1) dr$, to obtain

$$I = (\ln k_1)^{-1} [C \sin(R^{1/2} \ln k_1) + \tilde{C} \cos(R^{1/2} \ln k_1)] + O((\ln k)^{-2}). \tag{14c}$$

Substitution of this into $\mathfrak{M}$ [above (14a)] gives

$$\mathfrak{M} = (k_1 k_2)^{-1/2} \left[C_1 \frac{\sin(R^{1/2} \ln k_1 + C_2)}{\ln k_1} + O\left(\frac{1}{(\ln k_1)^2}\right)\right], \tag{2c}$$

and substituting $\mathfrak{M}$ in Eq. (1) and carrying out the integrations over k_1 and k_2 which are straightforward, we obtain the final result, the threshold law for electron-atom impact ionization,

$$\mathfrak{L}(E) \propto [E/(\ln E)^2]\{1 + C \sin[\alpha(R)\ln E + \mu]\}. \tag{15a}$$

Note that this differs slightly from our previous conjecture[2,3] of a modulated linear law. Of particular interest is the fact that the derivation yields an approximate reciprocal relationship between C and $\alpha(R)$,

$$\alpha(R) = C^{-1} = R^{1/2}. \tag{16}$$

The parameter R can be estimated by assessing where Ψ_f, Eq. (5b), becomes an accurate solution of the Schrödinger equation. Defining and working out

$$[H-E]\Psi_f \equiv \delta\Psi_f,$$

we find to leading order

$$\left|\frac{\delta\Psi_f}{\Psi_f}\right| = \left|\frac{(\ln r_1)^2 - 4(2r_2)^{1/2}\ln r_1 \cot(\alpha \ln r_1)\tan[(8r_2)^{1/2} - 3\pi/4]}{4r_2}\right|. \quad (17a)$$

The condition $|\delta\Psi_f/\Psi_f| \ll 1$, away from the zeros of Ψ_f, leads to very broad limits on $(r_2)_{\min} = R$:

$$7 \lesssim R^{1/2} \lesssim 160. \quad (17b)$$

These limits encompass the value $\alpha(R) \cong R^{1/2} = 42$ found in the recent experiment of Donahue *et al.*[5] when fitted by our modulated linear law.[2,3] It will be of interest to see what happens when their data are fitted by $\mathcal{Q}$ of Eq. (15). Of more present significance is the fact that even with the modulated linear-law fit, Donahue *et al.*,[5] find $C = 0.045$, which agrees with (16) to better than a factor of 2!

For completeness we note that (15a) only applies to a single partial wave (e.g., two-electron photodetachment); in impact ionization (15a) must be generalized to

$$\mathcal{Q}_e(E) \propto [E/(\ln E)^2][1 + \textstyle\sum_L C_L \sin(\alpha_L \ln E + \mu_L)]. \quad (15b)$$

The threshold law is the same for each partial wave (L), because any finite partial wave will be eventually dominated by the attractive dipole potential

$$\lim_{r > R_L} V_d(r) = \frac{-[R_L - L(L+1)]}{r^2}. \quad (18)$$

However, this does suggest that the reciprocal relation between C_L and α_L will be somewhat altered:

$$\alpha_L = [R_L - L(L+1) - \tfrac{1}{4}]^{1/2}; \quad C_L = R_L^{-1/2}. \quad (19)$$

It is interesting that the fit of the experiment of Donahue *et al.*, which measures two-electron photodetachment ($L = 1$), deviates from reciprocity in the direction of (19).

I will discuss only briefly the implications of this result—in particular, the deviation from the Wannier law[6] or its subsequent WKB rederivations.[7,8] I have recently discussed this anew elsewhere[9] in connection with recent work of Bottcher[10] and the new experimental results of Donahue *et al.*[5] In addition to what was stated above, the latter[5] shows that the yield curve can be at least as well fitted by a modulated linear law as by $E^{1.127}$. (The confidence level takes into account the number of parameters in any assumed form of law.) Bottcher's latest time-dependent numerical calculations[11] reveal that the final state is dominated by unequal-energy events, which include ionizing collisions importantly. The dominance of such unequal-energy events is the most salient prediction of the Coulomb-dipole theory[3] beyond the form of the threshold law itself. Mention should also be made of the experiment by Cvejanovic and Read,[12] which previously provided the greatest experimental support of the Wannier theory.[6] However, as has been exhibited, most convincingly by Lineberger, Hotop, and Patterson *et al.*[13] in ordinary single-electron photodetachment, the correct (in that case Wigner) threshold law may only be valid within as little as 5 meV of threshold. In that sense the new experiment of Donahue *et al.*[5] provides an important step in lowering the experimental range.

Finally, I have argued elsewhere[14] that the Coulomb-dipole threshold law should also apply to positron-atom impact ionization. In view of the present analysis, the modulated linear law given there should be changed to the form of Eq. (15b). But the qualitative difference from the recent result of Klar[15] ($\mathcal{Q}_{\text{Klar}} \propto E^{2.65}$), derived on the basis (*mutatis mutandis*) of a Wannier approach, persists. The yield of the latter, being smaller even than phase space ($\mathcal{Q}_{\text{phase space}} \propto E^2$), provides a crucible for the underlying assumption of the Wannier theory: the validity of the quasi-ergodic hypothesis.[4,9]

I would like to thank the members of the Clinton P. Anderson Meson Physics Facility (LAMPF) collaboration (cf. Ref. 5), in particular Howard Bryant and Joey Donahue, for many informative conversations.

[1]A. Temkin, Phys. Rev. Lett. 16, 835 (1966), and in *Physics of the One and Two-Electron Atoms*, edited by F. Bopp and H. Kleinpoppen (North-Holland, Amster-

dam, 1969), p. 655; A. Temkin, A. K. Bhatia, and E. Sullivan, Phys. Rev. 176, 80 (1968). The dipole normalization factor was first derived in this last reference with an omission corrected in A. Temkin and Y. Hahn, Phys. Rev. A 10, 708 (1974).

[2]Temkin and Hahn, Ref. 1.

[3]A. Temkin, J. Phys. B 7, L450 (1974).

[4]A. Temkin, in Proceedings of the Study Weekend on Collision Processes, Daresbury Laboratory, 26-27 March 1982 (to be published).

[5]J. B. Donahue, P. A. M. Gram, H. V. Hynes, R. W. Hamm, C. A. Frost, H. C. Bryant, K. B. Butterfield, D. A. Clark, and W. W. Smith, Phys. Rev. Lett. 48, 1538 (1982).

[6]G. H. Wannier, Phys. Rev. 90, 817 (1953).

[7]R. Peterkop, J. Phys. B 4, 513 (1971).

[8]A. R. P. Rau, Phys. Rev. A 4, 207 (1971).

[9]A. Temkin, Comments At. Mol. Phys. 11, 287 (1982).

[10]C. Bottcher, J. Phys. B 14, L349 (1981).

[11]C. Bottcher, to be published.

[12]S. Cvejanovic and F. H. Read, J. Phys. B 7, 1841 (1974).

[13]Cf. W. C. Lineberger, H. Hotop, and T. A. Patterson, in *Electron and Photon Interactions with Atoms*, edited by H. Kleinpoppen and M. R. C. McDowell (Plenum, New York, 1976), p. 125.

[14]A. Temkin, J. Phys. B 15, L301 (1982).

[15]H. Klar, J. Phys. B 14, 4165 (1981).

Fractional Mode Numbers in Wavy Taylor Vortex Flow

Guenter Ahlers, David S. Cannell, and M. A. Dominguez Lerma
Department of Physics, University of California, Santa Barbara, California 93106
(Received 3 May 1982)

Spectral power and phase measurements are presented which demonstrate the existence of a wavy Taylor vortex-flow state with a mode number $m = \frac{3}{2}$. The mode is periodically continued around the annulus by a phase anomaly which is stationary in the laboratory frame. It is also shown that wavy Taylor vortex flow results in an increase (decrease) of the Taylor vortex-pair width near the center (ends) of the apparatus.

PACS numbers: 47.20.+m, 47.35.+i

Fluid flow experiments often reveal that boundary conditions result in spatially periodic velocity fields with an integer number of wavelengths filling the container. The most extensive example is perhaps based on the study by Coles[1] of the Couette-Taylor system of a fluid contained between two concentric cylinders with the inner one rotating. In that case, the flow could be described by the (integer) number p of Taylor vortices filling the system in the axial direction and the (integer) number m of wavelengths of a wavy mode traveling azimuthally. We report in this Letter measurements on a wavy Taylor vortex-flow (WVF) mode with *non*integer mode number $m = \frac{3}{2}$. The existence of fractional mode numbers should be of importance in classifying the various routes to nonperiodic, or turbulent, flow. The classification of WVF states and of modulated wavy vortex-flow (MWVF) states by Gorman, Swinney, and Rand[2] and by Rand[3] does not include low-symmetry modes such as ours.

Our apparatus consists of two concentric cylinders, with the inner one rotating and the outer one stationary. The inner radius was $r_i = 3.118$ cm, and the radius ratio $\eta \equiv r_i/r_0$ was 0.893. The upper and lower boundaries were rigid and nonrotating. We studied the aspect ratio $L \equiv H/d = 53.9$ (H = column height and $d = r_0 - r_i$). The temperature was constant and uniform to $\pm 5 \times 10^{-3}$ °C, and the cylinder speed was controlled accurately by a frequency synthesizer. The fluid was a 30% solution of glycerol in water by volume, with 0.6% by volume of a "Kalliroscope" suspension added for flow visualization. Two independent light reflectance probes were used to study the time dependence of the visualized flow. Each probe consisted of a normally incident 2-mW He-Ne laser beam and a photovoltaic detector. The signals were sampled simultaneously and both probes could be translated vertically with a resolution of 13 μm.

Upon increase of the angular speed Ω of the inner cylinder, and thus the Reynolds number R, a transition occurred at $R \equiv R_c$ from azimuthal flow (except near the ends) to a primary-mode[4] Taylor vortex-flow (TVF) state consisting of toroidal vortices with an axial wavelength $\lambda \simeq 2.0$. (All lengths will be reduced by the gap d.) For