Brief Reports

Brief Reports are short papers which report on completed research which, while meeting the usual **Physical Review** *standards of scientific quality, does not warrant a regular article. (Addenda to papers previously published in the* **Physical Review** *by the same authors are included in Brief Reports.) A Brief Report may be no longer than 3½ printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Electron-atom spin asymmetry and two-electron photodetachment: Addenda to the Coulomb-dipole threshold law

A. Temkin
Atomic Physics Office, Laboratory for Astronomy and Solar Physics, NASA Goddard Space Flight Center, Greenbelt, Maryland 20771
(Received 27 February 1984)

In the light of a couple of new and important experiments in threshold ionization, several aspects of the Coulomb-dipole threshold law, $\mathcal{Q} \propto E(\ln E)^{-2}M(E)$ are reexamined. The principal results are as follows. (a) The logarithmic factor has a natural cutoff $\{\ln[E\,(\mathrm{Ry})/5]\}^{-2}$ when, as indicated, E is expressed in Rydberg units. (b) The modulation factor for electron-impact ionization has the form $M \cong 1 + (D/\alpha)\sin(\alpha \ln E + \phi)$ in which the effect of a superposition of partial waves gives a factor D which is expected to be less than one. This alters the relation between frequency and amplitude that is predicted to arise in the spin-asymmetry ratio as a function of E. Finally, (c) the range of validity of the threshold law ($E < E_r$) is estimated in terms of a formula between E_r and the dipole parameter α of $M(E)$. Some implications of this, particularly for the recent two-electron photodetachment experiment of Donahue and co-workers [Phys. Rev. Lett. **48**, 1538 (1982); **52**, 164(E) (1984)] are drawn.

I. INTRODUCTION

In a recent paper[1] we have derived the ionization threshold law based on a Coulomb-dipole (CD) theory of the ionization process. The threshold law is by definition the yield of positive ions in the limit that the available energy after ionization approaches zero (lim $E \to 0$). For the purposes of this discussion we shall express that result:

$$\mathcal{Q}(E) = \frac{CEM(E)}{\{\ln[E\,(\mathrm{Ry})/5]\}^2} \ . \tag{1}$$

In comparison with the previous form,[1] the denominator here involves the energy in rydbergs E (Ry) and that energy is divided by a factor of 5. The modulation factor $M(E)$ is the given generic type but of two specific forms, $M_e(E)$ and $M_v(E)$, depending on the specific threshold process: (a) electron-atom-impact ionization and (b) two-electron photodetachment. For process (a)

$$M(E) = M_e(E) = 1 + \sum_L \frac{d_L}{\alpha_L} \sin(\alpha_L \ln E + \mu_L) \ . \tag{2a}$$

$M_e(E)$ is seen to involve a superposition of partial waves. In (2a) the dependence on α_L in the denominator is approximate. We give the exact dependence for process (b) (the word photodetachment implies that the target here is a negative ion):

$$M(E) = M_v(E) = 1 + \frac{d}{(\alpha^2+1)^{1/2}} \sin(\alpha \ln E + \mu) \ . \tag{2b}$$

Equation (2b) is clearly a special case of (2a) in which only one partial wave is involved, but in addition our derivation[1] shows the more exact form of the relation between the coefficient outside the sine term and the coefficient multiplying $\ln E$ inside the sine term. (Note only one $d_L \equiv d$ contributes. In principle $d = 1$, but the present fit has $d \neq 1$; cf. discussion below and Fig. 2 caption.)

It is our purpose in this Brief Report to discuss the origin of the specific form of the logarithmic factor in Eq. (1), which is a result particularly relevant to two-electron photodetachment, as well as a simplification of Eq. (2a) as it arises in the quantitative description of electron- (alkali or hydrogen) atom spin asymmetry. Both kinds of experiments in the neighborhood of threshold have now been initiated,[2,3] and our results will have implications for them as well as future experiments along those lines.

II. ENERGY SCALE OF THE LOGARITHMIC DENOMINATOR

First note that all factors other than the $(\ln E)^{-2}$ in $\mathcal{Q}(E)$ are scale independent in the sense that if one measures E in different units, then one can alter a suitable constant so as to preserve the value and form of $\mathcal{Q}(E)$. For example, if we measure E in eV, then the linear and sinusoidal factors in (1) are such that $CE(\mathrm{Ry}) = C'E(\mathrm{eV})$ and

$$\sin[\alpha \ln E\,(\mathrm{Ry}) + \mu] = \sin[\alpha \ln E\,(\mathrm{eV}) + \mu'] \ ,$$

where $C' = C/13.6$ and $\mu' = \mu - \ln(13.6) = \mu - 2.610$. However the $(\ln E)^{-2}$ factor does not have the property, although to be sure it is scale independent in the limit

$$\lim_{E\to 0} \{\ln[E\,(\mathrm{Ry})]\}^{-2} = \lim_{E\to 0} \{\ln[E\,(\mathrm{eV})]\}^{-2} \ .$$

For that limit to be numerically accurate would require a value of E far too small to be experimentally feasible.

Thus, for practical purposes one must know something about the scale of this logarithm.

In fact, the answer to the question of the appropriate cut-off was implicit in our original considerations[4] of what constitutes the CD region. Specifically in terms of the recent law[1] the Coulomb-dipole contribution to the yield can conveniently be written[5] (in Ry units)

$$\mathcal{Q}_v(E) \propto \int_0^{E/5} \left[\frac{1+\cos[\alpha(R)\ln\epsilon + C_2]}{(\ln\epsilon)^2} \right] d\epsilon \ . \tag{3}$$

The upper limit in particular assumes that the energy ϵ of the slower electron is related to its wave number k_2 via

$$\epsilon = k_2^2 \ . \tag{4}$$

Asymptotically the distances of the outgoing electrons from the nucleus at time t are given by

$$r_i = k_i t, \ i = 1,2 \ . \tag{5}$$

When this is combined with the expression for the total energy in Rydberg units,

$$k_1^2 + k_2^2 = E\,(\mathrm{Ry}) \tag{6}$$

(implying $k_1^2 = E - \epsilon$ is the energy of the faster electron) and the definition of the Coulomb-dipole region

$$(r_2/r_1) \leq 1/2 \ , \tag{7}$$

one finally arrives[4] at the differential energy range

$$0 \leq \epsilon \leq \tfrac{1}{5} E\,(\mathrm{Ry}) \tag{8}$$

and its symmetric counterpart $\frac{4}{5}E\,(\mathrm{Ry}) \leq \epsilon \leq E\,(\mathrm{Ry})$ as constituting the Coulomb-dipole region. If for example, one had used atomic units of energy (hartrees), then one would obtain the limits of the Coulomb-dipole region $0 \leq \epsilon \leq \frac{1}{10} E$ (hartree). To be sure the upper limit $\frac{1}{5}E\,(\mathrm{Ry})$ does depend on the definition of the Coulomb-dipole region as given by (7). Suppose we had used a more restrictive boundary for the CD region, $r_2/r_1 \leq \frac{1}{3}$, then a similar analysis would yield a CD energy range $0 \leq \epsilon \leq \frac{1}{10} E\,(\mathrm{Ry})$. Although this may seem to be a substantial difference from (8), it is in fact a very small difference in the logarithmic factor in the denominator of the threshold law (1), which (in eV) goes from

$$\{\ln[\tfrac{1}{5}E\,(\mathrm{Ry})]\}^{-2} = \{\ln[E\,(\mathrm{eV})] - 4.22\}^{-2} \ , \tag{9}$$

to

$$\{\ln[\tfrac{1}{10}E\,(\mathrm{Ry})]\}^{-2} = [\ln E\,(\mathrm{eV}) - 4.91]^{-2} \ .$$

The insensitivity of the cut-off parameter in the threshold law can be summarized by writing

$$\mathcal{Q}(E) = \frac{CEM(E)}{[\ln E\,(\mathrm{eV}) - X]^2} \ , \tag{10}$$

where the above arguments would suggest that $4 \leq X \leq 5$. This will be particularly pertinent to the present[2] and future photo double detachment experiments when the data are fitted to our new form, Eq. (10), of $\mathcal{Q}_v(E)$ (cf. also Sec. IV).

III. THE SPIN-ASYMMETRY PARAMETER

Spin asymmetry is a quantity associated with polarized electron-atom-impact ionization. As such it measures the contribution from all partial waves, thus described by the modulation function $M_\epsilon(E)$ of Eq. (2a) in the threshold limit. Specifically, the spin-asymmetry ratio for electron-(hydrogen or alkali) atom-impact ionization is defined by (cf. Ref. 6)

$$A_I \equiv \frac{\mathcal{Q}(\uparrow\downarrow) - \mathcal{Q}(\uparrow\uparrow)}{\mathcal{Q}(\uparrow\downarrow) + \mathcal{Q}(\uparrow\uparrow)} = \frac{\mathcal{Q}_s - \mathcal{Q}_t}{\mathcal{Q}_s + 3\mathcal{Q}_t} \ . \tag{11}$$

The arrows in the argument refer to directions of the electron and target atom *before* the collision, and what is measured is the total yield of positive ions in each case. For theoretical analysis it is more convenient to convert this to singlet and triplet (s and t) yields, as is done in the right most part of Eq. (11). In principle, one can insert the threshold form of the cross section [Eq. (1)] with each set of the constants having the appropriate subscript, to find the threshold form of A_I. To a good approximation the resulting expression can be reduced to the form

$$A_I(E) = \beta + \tfrac{1}{4}(1 + 2\beta - 3\beta^2) \times \left[\frac{D_s}{\alpha_s} \sin(\alpha_s \ln E + \phi_s) - \frac{D_t}{\alpha_t} \sin(\alpha_t \ln E + \phi_t) \right] \ , \tag{12}$$

where

$$\beta = \frac{1 - (C_s/C_t)}{1 + 3(C_s/C_t)} \tag{13}$$

and ($\sigma = s$ or t)

$$D_\sigma \cos\phi_\sigma = \sum_L d_L^{(\sigma)} \cos\mu_L^{(\sigma)} \ , \tag{14a}$$

$$D_\sigma \sin\phi_\sigma = \sum_L d_L^{(\sigma)} \sin\mu_L^{(\sigma)} \ . \tag{14b}$$

The major approximation which goes into (12) is that dipole parameters for the various partial waves[1]

$$\alpha_L = [2R_L - L(L+1) - \tfrac{1}{4}]^{1/2} \tag{15}$$

are associated with minimum distances of the inner electron from the nucleus R_L which are so large that both R_L and the α_L are essentially independent of L for all important partial waves; i.e.,

$$R_L \cong R >> L(L+1) \ , \tag{16a}$$

$$\alpha_L \cong \alpha = \sqrt{2R} \ . \tag{16b}$$

The analytical result for $A_I(E)$ in Eq. (12) differs in form from the one originally derived in Ref. 5 only by the presence of the superposition constant D_σ in Eqs. (14); i.e., the formula in Ref. 5 can be recovered by setting $D_s = D_t = 1$. However, the consequence of the factors D_σ from the experimental point of view could be significant. Assuming several partial waves make sizable contributions and the associated phases $\mu_L^{(\sigma)}$ [which, in principle, result from detailed calculations and are not controlled by any dominant physical process (as far as we can see)] are randomly distributed, it is clear that a fair amount of cancellation can be expected to occur among the terms on the right-hand side (RHS) of Eqs (16). Thus, we expect

$$D < 1 \ . \tag{17a}$$

(We require that $\sum_L d_L = 1$, and the individual phases $\mu_L^{(\sigma)}$ are defined such that $d_L^{(\sigma)} \geq 0$.) In that case, the magni-

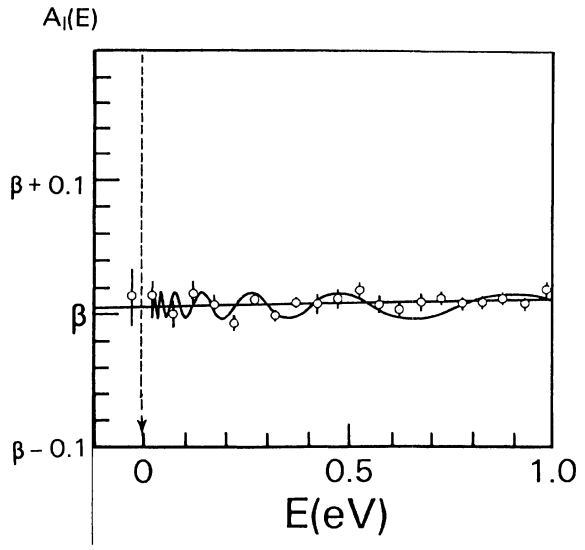

FIG. 1. Spin-asymmetry ratio A_l; oscillatory curve corresponds to the CD theory in Eq. (14), with parameters $\alpha=10$, $D=0.4$, $\phi_s=3.652$, $\phi_t=4.642$. These values can be obtained from the individual parameters [Eqs. (13) and (14)] $C_t=1$, $C_s=3.6667$; $\mu_0^{(t)}=-0.271\,739$, $\mu_1^{(t)}=3.272\,57$; $\mu_0^{(s)}=-1.261\,75$, $\mu_1^{(s)}=2.282\,54$; $\alpha_s=\alpha_t=10$, and $d_0=d_1=1$ independent of spin. The data are taken from Ref. 3. The absolute value of β in the figure is left unspecified in view of experimental discrepancies that now exist (cf. Ref. 6, and Refs. 8, 9 of Ref. 3).

tude of the modulation of A_l, which can readily be computed from Eq (12),

$$\Delta A=|A_{\max}-A_{\min}|\leq D/\alpha\ , \tag{17b}$$

(where D is a mean value of D_s and D_t) would be smaller than that derived and exhibited in Ref. 5 based on the single partial wave formula.

In Fig. 1 we have plotted $A_l(E)$ from Eq. (12) using $\alpha=10$ and $D=0.4$. It is compared to the data from the recent spin-asymmetry experiment of Kelley, Rogers, Celotta, and Mielczarek.[3] The experimental data are cut off at $E=1$ eV, although in Ref. 3 the data are given to $E=1.9$ eV. Even the former E is much beyond where we expect the threshold law to be valid for (our preferred) values of $\alpha=10$, and $\gamma=2$ (cf. Table I); nevertheless, we see that the oscillatory fit is as consistent as the straight line fit (also given), which is the prediction of the Wannier theory.[8] No concerted effort has been made to adjust the parameters of the oscillating curve (CD theory) to achieve an optimal fit. Rather, we would emphasize here that it will take a considerable improvement, particularly in energy resolution for an electron-impact spin-asymmetry experiment, to distinguish convincingly between the different predictions.

TABLE I. Expected energy range E_r (eV) of the validity of the ionization threshold law for values of α and γ [cf. Eq. (28)].

γ \ α	5	10	20	40
2	1.0	0.07	4×10^{-3}	3×10^{-4}
3	0.5	0.04	2×10^{-3}	1×10^{-4}
5	0.17	0.014	7×10^{-4}	4×10^{-5}
10	0.04	3×10^{-3}	2×10^{-4}	1×10^{-5}

IV. ESTIMATE OF α AND THE ENERGY RANGE OF VALIDITY OF THE THRESHOLD LAW: TWO-ELECTRON PHOTODETACHMENT

In Fig. 2 we give the result of the two-electron photodetachment experiment of Donahue *et al.*[2] This figure indicates several important points: first the energy range and, in particular, the resolution of the photodetachment experiment is much finer (a factor 13 to 21) than the electron-impact experiment.[3] The dashed curve represents the fit to our prior modulated linear law[4,7] $\mathcal{Y}_\nu\propto EM_\nu(E)$, and the solid curve, which is the one given in the erratum of Ref. 2, represents the theoretical fit convoluted with the experimental energy resolution[2] ($\Delta E_{\text{expt}}\cong0.007$ eV). One sees that the resolution is so good that at the upper end ($E\cong0.3$ eV) there is practically no difference between the two curves. It is also clear that the fitted[2] value of $\alpha=41.6$ was controlled by the experimental substructure in the vicinity of $E\cong0.3$ eV. The significance of the experimental oscillations, if they are real, can hardly be overestimated (see below), but we shall indicate below that the true threshold range and the true value of α are likely to be significantly smaller than the above values.

Let us first derive a simple relation for the local oscillation period in energy (λ_E) implicit in our (CD) threshold law. We shall confine ourselves here to the single partial wave modulation factor $M_\nu(E)$ appropriate to two-electron photodetachment. From (2b) one sees that if $M_\nu(E')=M_\nu(E)$ at two neighboring energies E and E',

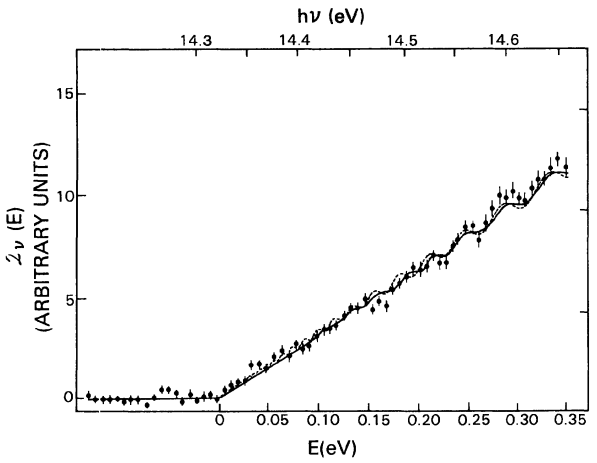

FIG. 2. Two-electron photodetachment yield $\mathcal{Y}_\nu$ vs E. Data from Fig. 3(b) of Donahue *et al.*, (Ref. 2). The dashed curve is the threshold linear modulated law (discussed at the outset of Sec. IV) using their parameters. In our notation these parameters are $C=33.5$, $\alpha=41.6$, $d=1.86$, $\mu=5.2$, and the background $B=0.69$. The photon energy (in the c.m.) is $h\nu$ and the available energy $E=(h\nu-14.321)$ eV. The solid curve, given in the erratum of Ref. 2, corresponds to the theoretical curve convoluted with the experimental energy resolution ($\Delta_{\text{expt}}=0.007$ eV). It is emphasized that the data in Ref. 2 have also been fitted, with a slightly lesser confidence factor (19% vs 25%), to the Wannier law.

$E' = E + \lambda_E$, then $\alpha \ln E' = \alpha \ln E + 2\pi$. This yields

$$\lambda_E = (e^{2\pi/\alpha} - 1)E \cong \frac{2\pi}{\alpha} E \quad . \tag{18}$$

Assuming it requires about four resolved energy points to establish a wiggle experimentally leads to an experimental energy resolution requirement:

$$\Delta E_{\text{expt}} \lesssim (\pi/2\alpha)E \quad . \tag{19}$$

One sees from Fig. 2 that Eq. (19) correctly describes where in energy the double photodetachment data[2] can and cannot follow the predicted wiggles given the fitted[2] value of $\alpha = 41.6$. The question is how reasonable is that value of α?

Let us start with the observation that in order for the quantum-mechanical (Coulomb-dipole) theory to be applicable the de Broglie wave of the outer electron, λ_d, must be substantially larger than the minimum distance (R) of the inner electron from the nucleus $\lambda_d \geqslant \gamma R$, where, in analogy to the delimitation of the Coulomb-dipole region by (7), we can limit γ by $2 \lesssim \gamma \lesssim 10$.

The origin of the inequality for λ_d is clear: the outer electron must be going slowly enough so it cannot probe the detailed structure of the dipole formed by the inner electron and the nucleus; otherwise some form of classical approximation would be appropriate, and that is the province of the Wannier theory.[8] [We have discussed the complementarity of the latter to our theory elsewhere;[4,5] suffice it here to repeat that our (CD) theory dominates the yield in the threshold limit.[1,5]] From the inequality for λ_d above and the symmetric counterpart of (8) one readily derives for the total energy E:

$$E \lesssim E_r = \tfrac{5}{4} k_f^2 = \tfrac{5}{4}\left(\frac{2\pi}{\lambda_d}\right)^2 \leqslant \frac{5\pi^2}{\gamma^2 R^2} \quad . \tag{20}$$

Note that this confirms the scaling between the threshold energy domain, E_r, and R^{-2} that was previously suggested.[9] However, the constant of proportionality ($5\pi^2/\gamma^2$) may be as large as 10, so that the threshold domain will be proportionately larger than estimated in Ref. 9. Using the relations between R and α, Eq. (16b), we obtain the desired relation between the threshold energy domain and α:

$$E_r \cong 20\pi^2/\gamma^2\alpha^4 \quad . \tag{21}$$

In Table I we present values of E_r as a function of α and γ. If the value of α were as large as 40, then the threshold domain would be less than the range of millivolts; that would be inconsistent with the value of E at which the value $\alpha = 40$ was determined in the first place[2] (cf. Fig. 2). This suggests that the experimental oscillations, if they are confirmed to be real, may correspond to higher terms (harmonics) in the CD threshold expansion. Our preferred values are $\gamma \cong 2$ and $\alpha \cong 10$; there we see from Table I that the threshold regime is limited to $E \lesssim 0.07$ eV. It is interesting to observe in Fig. 2 that there is a noticeable structure in the data at $E \cong 0.05$, and from Eq. (19) the experimental width is marginally adequate to resolve it. We recommend this region for further study. [Analytical fits to a recent $He^-(^4P^o)$ photo double detachment experiment[10] have yielded $\alpha \cong 8$ when fit to the CD law and $n \cong 1.5$ for a power law, $\mathcal{D} \propto E^n$.[11]]

Table I is also seen to contain entries for $\alpha = 5$. This is noticeably smaller than our estimate[5] of $\alpha \cong 10$, which itself, it is to be emphasized, was *a lower bound* on α derived on the basis that the region for which the relative error of the Coulomb-dipole function as an exact solution of the Schrödinger equation be less than a tenth ($\Delta\Psi/\Psi \lesssim \frac{1}{10}$). If one relaxes that condition by a factor of 2 ($\Delta\Psi/\Psi \lesssim \frac{1}{5}$), then one finds that the Coulomb-dipole region demands a minimum value of $r_2 \equiv R \cong 13.5$. This corresponds to an $\alpha \cong 5.2$, and it accounts for the added row in Table I.

There is an additional reason for taking the $\alpha = 5$ column seriously. The magnitude of the ionization matrix element coming from the Coulomb-dipole region can readily be estimated to be $\mathcal{M} \propto R^{7/4} e^{-R}$. The estimate is derived from Eqs. (14) and (15) of Ref. 1, and it does not include any k dependent or sinusoidal factors. Given the fact that $R \propto \alpha^2$, one sees that the size of the matrix element will depend very sensitively on α. This is the analytic estimate of the tunneling effect which has recently been mentioned by Feagin.[12] A value of $\alpha = 10 (R = 50)$ yields 2×10^{-19} for $\mathcal{M}$, whereas $\alpha = 5 (R = 12.5)$ gives a value 3×10^{-4}. Although this is not germane to the abstract question of the form of the threshold law, it suggests that the smaller value of α will have a profound effect on its experimental observability, particularly as regards electron-impact spin-asymmetry experiments.

[1] A. Temkin, Phys. Rev. Lett. **49**, 365 (1982).

[2] 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); **52**, 164(E) (1984).

[3] M. H. Kelley, W. T. Rogers, R. J. Celotta, and S. R. Mielczarek, Phys. Rev. Lett. **51**, 2191 (1983).

[4] A. Temkin, J. Phys. B **7**, L45 (1974).

[5] A. Temkin, in *Electronic and Atomic Collisions,* edited by J. Eichler, I. V. Hertel, and N. Stolterfoht (Elsevier, New York, 1984), p. 755.

[6] Cf. T. J. Gay, G. D. Fletcher, M. J. Alguard, V. W. Hughes, P. F. Wainwright, and M. S. Lubell, Phys. Rev. A **26**, 3664 (1982), and papers cited therein.

[7] A. Temkin and Y. Hahn, Phys. Rev. A **10**, 708 (1974).

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

[9] A. Temkin, IEEE Trans. Nucl. Sci. **30**, 1106 (1983).

[10] Y. K. Bae, M. J. Coggiola, and J. R. Peterson, Phys. Rev. A **28**, 3378 (1983).

[11] J. R. Peterson (private communication).

[12] J. Feagin, J. Phys. B **17**, 2433 (1984).