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Comparative analysis of the threshold behavior of two-electron escape in electron-impact ionization of valence-1 atoms

M. S. Lubell

Department of Physics, City College of the City University of New York, New York, New York 10031

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Using spin-asymmetry data from electron-impact-ionization studies of H, He*, Li, Na, K, and Cs, we determine the energies at which dynamical influences on the inside part of the final-state wave function create detectable departures from the simple Wannier power-law dependence of the cross section on escape energy for the singlet and triplet channels separately. From a comparison of these energies for the six valence-1 systems examined, we conclude that asymmetry measurements provide a window for observing the effects of screening and core polarization on the dynamics of the highly correlated Coulomb three-body breakup.

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In the Wannier threshold theory [1] of two-electron escape, the electron ionization cross section σ is usually expressed in the form

$$\sigma(E)=aE^{\eta}\ , \tag{1}$$

where E is the combined escape energy of the two electrons, a is a proportionality constant, and η is given by

$$\eta=\frac{1}{4}\left[\left[\frac{100Z-9}{4Z-1}\right]^{1/2}-1\right]\ , \tag{2}$$

with Ze the charge of the residual ion. For systems in which $Z=1$, the exponent η has the value 1.127. Wannier's original work was classical in nature. However, semiclassical [2] and quantum [3] derivations of the power law appeared subsequently, and the subject of threshold double escape along the "Wannier ridge" has remained an active field of theoretical study to the present [4–17].

Until recently, virtually all experimental tests of the threshold behavior of double escape—the first precision measurements having been carried out for H [18] and He [19] about two decades ago—confirmed the Wannier prediction. Despite this, Temkin and his collaborators [20], continued to challenge the ansatz of the Wannier theory and advocate instead the Coulomb-dipole approach, which they had first developed approximately 30 years ago. While no experiment has been able to confirm the predictions of the Coulomb-dipole approach, the results of several recent investigations [21–23] nonetheless have raised new questions about the accuracy of the Wannier power law, as well as its general interpretation. These questions have surfaced against the backdrop of new interpretations of the characterization and classification of doubly excited states in the discrete spectrum of two-electron atoms and ions [24–26].

For the threshold region of the continuum, two issues must be confronted—the correct form of the energy dependence of the cross section describing double escape and the energy range over which the form is valid. It has been noted by a number of authors that the energy dependence of $\sigma(E)$ given by Eq. (1) actually contains only the leading term in E. As Klar and Schlecht [6] and later in more detail Klar [7], Greene and Rau [9], Feagin [10], and Peterkop [11] all showed, the cross section in general must contain a higher-order term in energy with the exponent 3η (or alternatively $3\eta+1/2$, depending upon the details of the analysis) in order to account for the angular characteristics of the final-state wave function. Macek [27] and Rau [28] have noted, however, that even with the inclusion of such a term, the resulting threshold law is inadequate, since it only treats the asymptotic part of the final-state wave function. In fact, in 1951, two years before Wannier published his classical threshold analysis, Teichmann [29] in a paper primarily devoted to effective range theory for low-energy nuclear collisions, pointed out that, as in the case of n-p scattering, Coulomb scattering also requires the inclusion of higher-order energy terms in order to describe the effects of the inside part of the final-state wave function. Whether one should interpret such effects as integral but often omitted parts of the quantum extensions of the Wannier approach, or whether one should interpret them as outside the usual confines of the ordinary asymptotic Wannier theory and hence evidence of its inadequacy is an issue that we will not even attempt to treat in this paper. Instead, we will explore the existing experimental evidence that dictates the inclusion of such higher-order terms. We will then use the relative importance of the higher-order terms for a number of valence-1 systems to probe the variation of the range of validity of the simple Wannier power law given by Eq. (1), an issue that has been raised either directly or obliquely in several theoretical papers [7,11,17].

Consistent with this approach, we first write the cross section $\sigma(E)$ in the form

$$\sigma(E)=aE^{\eta}g_1(E)+bE^{3\eta}g_3(E)\ , \tag{3}$$

where $g_1(E)$ and $g_3(E)$ are analytic functions of E that, for simplicity, we take as power series. In a recent paper [30] that examined the influence of higher-order terms on the cross section for electron impact ionization of atomic hydrogen, it was shown that the terms containing the 3η exponent do not contribute significantly over the energy range of interest. Thus we retain, as an approximation, only the lowest terms in $g_1(E)$ and write $\sigma(E)$ as

$$\sigma(E)\approx aE^{\eta}+cE^{\eta+1}\ , \tag{4}$$

a form that it was further shown [30] is sufficient to explain the observed energy dependence of hydrogen ionization spin-asymmetry measurements in the threshold region. With the use of the spin-asymmetry and spin-averaged cross-section data, it was also demonstrated that the singlet σ_s and triplet σ_t cross sections taken separately were considerably more sensitive to the presence of higher-order terms than was the spin-averaged cross section $\bar{\sigma}$. In this paper, we will extend the methods of the previous work with some revisions, and determine the respective "critical energies" E_s^c and E_t^c at which the influence of the higher-order term in Eq. (4) would become visible in the singlet and triplet channels separately at the 1% level for electron-impact ionizaton of He*, Li, Na, K, and Cs, as well as H, all of which have been the subject of experimental spin-asymmetry investigations [21,31–35].

In all of the spin-dependent ionization studies, the principal quantity determined was the ionization asymmetry A defined by

$$A=(\sigma_s-\sigma_t)/(\sigma_s+3\sigma_t) . \tag{5}$$

Based upon Eq. (4), we write the threshold ($E=0$) values of the asymmetry A_0, and its slope $(dA/dE)_0$, respectively, as

$$A_0=(a_s-a_t)/(a_s+3a_t) \tag{6}$$

and

$$\left[\frac{dA}{dE}\right]_0=4(a_t c_s-a_s c_t)/(a_s+3a_t)^2 . \tag{7}$$

We now introduce the "Wannier energy range," E_W, for which the spin-averaged cross section $\bar{\sigma}$ displays extremely good agreement with the simple power-law behavior given by Eq. (1). If we assume that the agreement is exact, c_s and c_t must obey the relation

$$c_s+3c_t=0 , \tag{8}$$

and we obtain from Eqs. (6) and (7) the expressions

$$\frac{c_s}{a_s}=3\left[\frac{dA}{dE}\right]_0\Big/(1+3A_0) \tag{9}$$

and

$$\frac{c_t}{a_t}=\left[\frac{dA}{dE}\right]_0\Big/(1-A_0) . \tag{10}$$

We note that since a_s and a_t are positive quantities and since $A_0>0$ for all systems investigated, the coefficient c_s, for the case described by Eq. (8), carries the same sign as $(dA/dE)_0$, while c_t carries the opposite sign.

If we now define E_W as the energy at which the presence of the higher-order term in Eq. (4) causes the measured value of the spin-averaged cross section to deviate from the "Wannier value" $a(E_W)^\eta$, by an amount $xa(E_W)^\eta$, we must replace Eqs (8)–(10) with the respective expressions

$$c_s+3c_t=-x(a_s+3a_t)/E_W , \tag{11}$$

$$\left[\frac{c_s}{a_s}\right]_x=\frac{3}{1+3A_0}\left[\frac{dA}{dE}\right]_0-\frac{x}{E_W} , \tag{12}$$

and

$$\left[\frac{c_t}{a_t}\right]_x=\frac{1}{1-A_0}\left[\frac{dA}{dE}\right]_0-\frac{x}{E_W} , \tag{13}$$

where we have used the minus signs because the value of $\bar{\sigma}$ eventually departs toward lower values from the aE^η prediction as E increases. For purposes of the analysis that follows, we will first take x to be 2%, an assumption that is consistent with the precision of the best spin-averaged cross-section experiments reported to date [12,18,19,36]. Later, we will examine the sensitivity of our results to the assumed value of the ratio x/E_W. The final quantities that we will evaluate are the respective "critical energies" E_s^c and E_t^c at which the singlet and triplet cross sections separately show departures from the E^η prediction at the fractional level y. These energies are evidently given by

$$(E_s^c)_x^y=y|(a_s/c_s)_x| \tag{14}$$

and

$$(E_t^c)_x^y=y|(a_t/c_t)_x| . \tag{15}$$

With perhaps an overly optimistic eye toward the future, we will quote these energies for $y=1\%$, the scaling for other values of y clearly following linearly with y.

A summary of the ionization asymmetry measurements obtained in various laboratories appears in Fig. 1. In some cases the uncertainties in the data points shown do not reflect the uncertainty in the absolute scale of the asymmetry. However, the uncertainties in A_0 and $(dA/dE)_0$ resulting from the applicable regression analyses are mostly large enough to accommodate such scaling errors, and where this is not the case (H, for example), the uncertainties have been adjusted accordingly. Except for the case of Cs, the values of A_0 and $(dA/dE)_0$ were obtained from linear regressions, an approach that is consistent with the low-energy behavior of the asymmetry, given the form of Eq. (4). The appropriate energy ranges for the fits were based upon the generation of acceptable reduced chi-squares χ_ν^2, ν being the number of degrees of freedom. In the case of H [21], the range was 0.4 eV, while for He* [31], it was 2 eV. For the Na and K data obtained by Baum *et al.* [32], a range of 4 eV was used, and for the Na data obtained by Kelley *et al.* [33], the full published set (ending slightly below 2 eV) was used. Since the measurements of Kelley *et al.* were published without an absolute scale, the intercept A_0 obtained from a linear fit to those data was normalized to the intercept found from a linear fit to the data of Baum *et al.* The data points of Kelley *et al.* were then scaled accordingly. The case of Li [32] was anomalous, since the data shown clearly reveal a strong departure from a linear relationship somewhere between 1 and 2 eV. The paucity of data and the extremely large uncertainties in the very low-energy measurements preclude a well-defined fit. As a result, the averages of the values of A_0 and $(dA/dE)_0$ were taken with and without the 2.11-eV data point included,

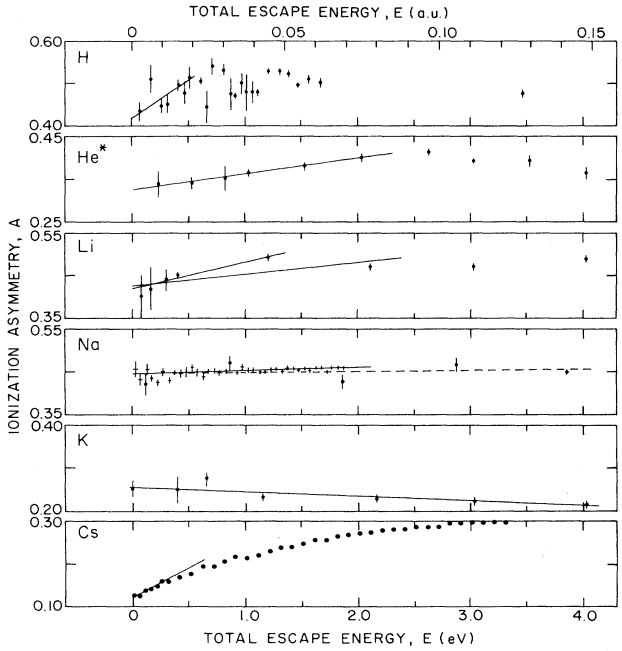

FIG. 1. Measured values of the ionization spin asymmetry A as a function of total escape energy E for H [21], He* [31], Li [32], Na (dots, Ref. [32]; crosses, Ref. [33]), K [32], and Cs [35]. Additional values for Na and K from other experiments [34] are not shown for lack of sufficient data in the energy range of interest. The straight lines are obtained from fits to the data as explained in the text. For Na, the dashed line corresponds to the dots and the solid line to the crosses.

and the uncertainties were expanded to include the fitted values as bounds. For the case of Cs [35], the data shown in Fig. 1 reflect nonlinearities at very low energies. The data set is so large, however, that it was possible to use a six-parameter nonlinear regression analysis to extract the threshold values of the asymmetry and its derivative.

Table I summarizes the results of the fitting procedures applied to the data of Fig. 1. For Na, the table contains the normalized data of Kelley *et al.*, since the data set of Baum *et al.* is much smaller and less accurate. For all fits, the values of χ^2_ν and the associated confidence levels (CL's) are quite acceptable—a CL of 50% is the ideal—especially since the uncertainty in the energy of the data points was ignored except in the case of H. Table I also contains entries for E_W that are taken from the best information available [18,36]. It should be noted that for He* the value associated with ground-state He [37] is used with the justification that the energy dependence of the ionization cross section in the threshold region is dominated by the final-state two-electron wave function and the state of the residual ionic core and, by contrast, is minimally affected by the initial state of the atomic system.

From the values of A_0, $(dA/dE)_0$, and E_W listed in the table, the "critical energies" E_s^c and E_t^c, were calculated in accordance with Eqs. (14) and (15) respectively, the results appearing as entries in the seventh and eighth columns. As the form of Eqs. (14) and (15) suggests, however, these results are potentially sensitive to the assumed values of x/E_W. We can test for this sensitivity for each atom by allowing x to vary between 1% and 5% and allowing E_W to vary between its nominal value given in Table I and half that value. The results of these tests, summarized in Fig. 2, reveal that 3 of the 12 calculated critical energies—E_s^c for Li and Na and E_t^c for K—display such a sensitivity. In spite of this drawback, we can still deduce the following conclusions with quite a high degree of confidence. First, with the atomic systems arranged in order of increasing Z, as displayed in Table I and Fig. 2, we see that the "critical energy" for singlets appears to increase monotonically until K is reached. Second, the critical energy for triplets appears to increase monotonically until Cs is reached. Finally, until K is reached, the critical energies for triplets lie lower than their counterparts for singlets.

As the blank space in Fig. 2 suggests, confidence in these conclusions would be increased if ionization-asymmetry data were available for Rb. Even in the absence of such data, however, our conclusions do not seem unreasonable, since they can be expected intuitively if double escape measurements are able to sample the influence of the inside part of the final-state wave function. Under these circumstances, for example, the effects of core screening and polarizaton must become visible, and a Z dependence should be anticipated. Moreover, it is certainly plausible that, when applied globally, the Pauli exclusion principle should make triplet configurations more sensitive to changes in the probabili-

TABLE I. Summary of threshold ionization parameters (1 a.u. =27.21 eV). The Na results are based upon data from Kelley *et al.* [33] with the value of A_0 normalized to that found from data of Baum *et al.* [32]. Values in parentheses denote one standard-deviation uncertainties.

Atom	A_0	$(dA/dE)_0$ (a.u.$^{-1}$)	χ^2_ν	ν	CL (%)	E_W (eV)	$(E_s^c)^{1\%}_{2\%}$ (meV)	$(E_t^c)^{1\%}_{2\%}$ (meV)	Ref.
H	0.43(3)	4.4(1.5)	0.78	5	56	0.4	62	30	[21]
He*	0.33(1)	1.00(25)	0.07	4	99	3.6	199	167	[31]
Li	0.43(3)	0.6(1.0)	[a]	[a]	[a]	1.0	1112	169	[32]
Na	0.44(2)	0.26(4)	1.01	36	46	1.0	1306	269	[33]
K	0.259(8)	−0.33(8)	1.12	5	35	1.0	247	273	[32]
Cs	0.121(3)	3.68(2)	1.21	31	20	1.0	36	75	[35]

[a]See the text.

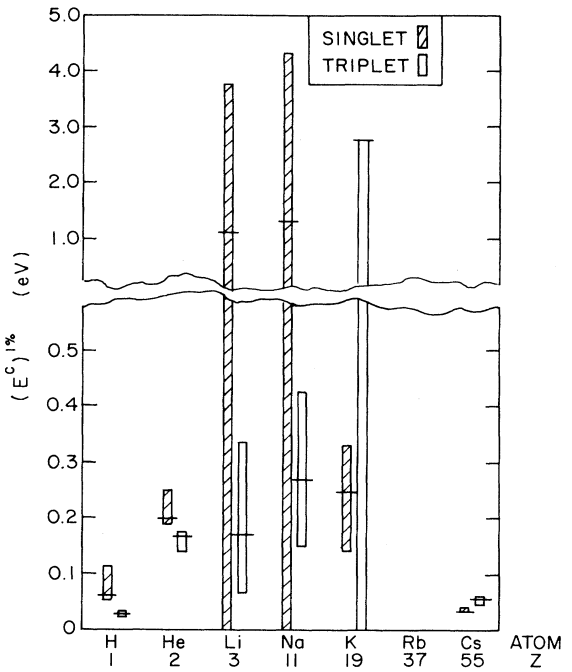

FIG. 2. Summary of "1% critical energies" for the valence-1 systems examined, with the vertical bar in each case showing the sensitivity of the result to the assumed value of x/E_W. The horizontal line through each bar represents the value of the critical energy listed in Table I.

ty distribution of the valence electron during the course of the collision. Since this distribution will vary most strongly as a function of incident energy for relatively unscreened systems, the observed pattern for E_t^c should not be unexpected for low values of Z. For high values of Z, the physics might be more complex, as the pattern reversal between K and Cs illustrates. Whether the reversal is attributable to relativistic effects or the influence of an unfilled d shell, a situation that first occurs in the case of K, or whether it is attributable to some other cause is a matter of conjecture at this point. What is certain, however, is that the inside part of the final-state wave function produces very discernible effects on the energy dependence of the cross section for double escape, and that these effects, in turn, seem to suggest a significant role for the core.

It should be noted that although some of the discussion has been rather speculative in nature, it has been intended as stimulation for future theoretical and experimental initiatives aimed at unraveling the physics of the highly correlated Coulomb three-body problem. On the basis of the analysis presented, however, several future experiments already appear to be called for—a measurement of the ionization asymmetry for Rb, a remeasurement of the ionization asymmetry for Li, considerably more accurate measurements of the spin-averaged cross sections for all the valence-1 systems, and two-electron photodetachment studies of negative ions in which the singlet or triplet channels are automatically isolated.

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