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Threshold ionization laws for electron-hydrogen scattering and their dominant region of configuration space

Philip L. Bartlett,* Andris T. Stelbovics,† and Igor Bray‡
Centre for Atomic, Molecular and Surface Physics, Murdoch University, Perth 6150, Australia
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A fully quantal calculation of the ^1S three-body wave function is performed for the Wannier model of electron-hydrogen scattering in the near-ionization threshold region using an exterior complex scaling method. The region of configuration space of the wave function that provides the dominant contribution to the total ionization cross section is demonstrated to be $r_1 \approx r_2$ in accord with the argument of Rau [Phys. Rev. A **4**, 207 (1971)], but only in the so-called Coulomb zone. The work confirms to a high precision ($E^{1.128\pm0.004}$) the Wannier threshold law for the total ionization cross section, which is strictly valid only at the threshold. A threshold law for the ^3S total ionization cross section is determined to be $E^{3.37\pm0.02}$, in agreement with the semiclassical calculations of Peterkop [J. Phys. B **16**, L587 (1983)].

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The validity of the Wannier [1] threshold law for the electron-hydrogen total ionization cross section (TICS) has experimental [2] and theoretical support, e.g., Refs. [3–8]. However, several interesting questions remain. The semiclassical and quantal derivations of Peterkop and Liepinsh [9] and Rau [3] assume that the region of configuration space in the neighborhood of $r_1 \approx r_2$ is dominant. Temkin [10] suggested that, on the contrary, the dominant region is the one where r_1 and r_2 are highly asymmetric. This assumption leads to a quite different threshold behavior. Interestingly, no *fully quantal calculation* has established which regions of the wave function are dominant for electron-hydrogen ionization. Also, the Wannier threshold law is based on arguments concerning the form of the three-body wave function in the region $r_1 \approx r_2$. The major contributions are from those three-body partial waves that do not vanish in this region. For example, if we consider the $L=0$ partial wave, quantum mechanically the singlet scattering provides the law although ^3S ionization may occur but should be suppressed relative to the singlet on account of the Pauli principle. What is the nature of the threshold law when there is no classical analog? Finally, the experimental work of McGowan and Clarke [2] indicates that the region is less than 0.4 eV above the ionization threshold. The computations of Kato and Watanabe [7] provided some weight for this conclusion but further work is warranted.

The purpose of this paper is to investigate these questions within the Wannier model of near-threshold ionization where the two electrons escape in the opposite directions. This model provides the essential features leading to the Wannier threshold law of the full problem near the ionization threshold. Its simpler collinear nature allows us to compute the three-body wave function with great accuracy since the electron-electron potential becomes just $1/(r_1+r_2)$ to second order in the deviation of the mutual angle from π.

*Electronic address: bartlett@fizzy.murdoch.edu.au
†Electronic address: stelbovi@fizzy.murdoch.edu.au
‡Electronic address: I.Bray@murdoch.edu.au

Writing the total wave function as a sum of the incident and scattered parts, $\Psi^{(+)}=\Psi_{\mathbf{k}_0}+\Psi_{\rm sc}$, the time-independent Schrödinger equation leads to

$$(E-\hat{H})\Psi_{\rm sc}(\mathbf{r}_1,\mathbf{r}_2)=(\hat{H}-E)\Psi_{\mathbf{k}_0}(\mathbf{r}_1,\mathbf{r}_2), \tag{1}$$

where $\hat{H}$ is the total Hamiltonian

$$\hat{H}(\mathbf{r}_1,\mathbf{r}_2)=-\frac{1}{2}\nabla_1^2-\frac{1}{2}\nabla_2^2-\frac{1}{r_1}-\frac{1}{r_2}+\frac{1}{r_1+r_2}. \tag{2}$$

Wannier [1] argued that the ionization process would be dominated by the S wave. Furthermore, as discussed by Rost [11] the threshold laws for the higher partial waves scale as the singlet $L=0$ partial wave, and our calculations support this also. Thus to determine the TICS near-threshold behavior it is sufficient to study the $L=0$ case.

We solved Eq. (1) for $L=0$ using the method of exterior complex scaling (ECS), where all coordinates are rotated into the complex plane by a fixed angle θ, at a sufficiently large hyperradius R, such that convergence of the extracted cross sections is obtained. This method was first implemented by Rescigno *et al.* [12] on an analytic test case, by McCurdy, Horner and Rescigno [13] on the Temkin-Poet and collinear models, and by Baertschy *et al.* [15] for the full electron-hydrogen problem.

We used the propagation method of Poet [14], which was adapted by Jones and Stelbovics [16,17] to ionization. The method was modified to allow for the inhomogeneous term in the ECS formulation. A three-point Numerov formula was used to solve the partial differential equation on a grid symmetric in r_1 and r_2, but modified to allow for the transition from real to complex coordinates at R_0 and for arbitrary grid spacing in other regions. Details of our method will be given in a separate publication.

Convergence studies were performed to determine the optimum R_0, θ, grid spacing, and complex contour length. The calculation was insensitive to variations in θ over a large range; 0.8 rad was used in these calculations. For total energies between 0.2 a.u. and 0.04 a.u., an overall estimated error

of 0.2% in the total ionization cross sections was maintained. At 0.2 a.u. this criterion required $R_0=600$ a.u., and at 0.04 a.u. $R_0=1400$ a.u. Below 0.04 a.u., R_0 was set to 1400 a.u. due to limits on our computational resources. As a result, the estimated error of these very low-energy calculations increased to 0.5–1.0 %.

The TICS was extracted from the scattered wave function using the relationships [9,18]

$$\sigma(E)=\int_0^{E/2}\frac{16}{\pi k_0 k_1 k_2}|f(k_1,k_2,R)|^2 d\epsilon_2, \tag{3}$$

$$f(k_1,k_2,R)\sim\frac{R}{2}\int_0^{\pi/2}d\alpha\left[\hat{\phi}_0(k_1,r_1)\hat{\phi}_0(k_2,r_2)\frac{\partial}{\partial R}\psi(r_1,r_2)-\psi(r_1,r_2)\frac{\partial}{\partial R}\hat{\phi}_0(k_1,r_1)\hat{\phi}_0(k_2,r_2)\right], \tag{4}$$

where $E=k_1^2/2+k_2^2/2=\epsilon_1+\epsilon_2$, $R=\sqrt{r_1^2+r_2^2}$ and $\alpha=\tan^{-1}(r_2/r_1)$ are the hyperradius and hyperangle, and $\hat{\phi}_0(k,r)$ is the hydrogenic Coulomb function for $l=0$. Though Eq. (4) is derived in the limit $R\to\infty$, it can be used for finite but large hyperradius by choosing R_0 as described to achieve good convergence of the single-differential cross section (SDCS). This method of extracting ionization cross sections was recently employed by McCurdy, Horner, and Rescigno [13] for model hydrogen problems and by Baertschy, Rescigno, and McCurdy [19] for the full hydrogen problem.

First, we examine the regions of configuration space of the scattering wave function responsible for the threshold law. According to Wannier [1] and Rau [3], it is the region $r_1\approx r_2$ in the so-called ''Coulomb zone,'' where $RE\ll 3/\sqrt{2}$ which dominates the threshold behavior. On the other hand, Temkin [10] stated that the dominant contribution to the threshold behavior is from the region where $r_1\geqslant 2r_2$, that is, $\alpha\leqslant\alpha_T=\tan^{-1}(1/2)\approx 26°$. Since these regions are complementary, a straightforward way to examine their contribution is to let $f=f_T+f_W$, where the Temkin and Wannier amplitudes are obtained by adjusting the integration limits in Eq. (4) to $[0,\alpha_T]+[(\pi/2)-\alpha_T,(\pi/2)]$ and $[\alpha_T,(\pi/2)-\alpha_T]$, respectively. Noting that $|f|^2=|f_T|^2+|f_W|^2+2\,\mathrm{Re}(f_W^* f_T)$, the contributions to the TICS from the Wannier and Temkin regions can be derived from Eq. (3). The interference term $2\,\mathrm{Re}(f_W^* f_T)$ will also contribute to the TICS.

Figure 1 shows the contribution to the TICS for the singlet wave function. Our computed TICS are labeled $\sigma(R_0)$. The corresponding Wannier and Temkin cross sections, extracted from our calculated wave functions, are labeled $\sigma_{W,T}(R_0)$, respectively. It is worth reiterating that in order to achieve the high accuracy stated, we used hyperradius values (R_0) ranging from 600 a.u. at $E=0.2$ a.u. $(R_0E=120)$ to 1400 a.u. at $E=0.04$ a.u. $(R_0E=56)$. At all lower energies $R_0=1400$ and at the smallest energy studied, 0.005 a.u., this gives $R_0E=7$. In all cases this places $RE\gg 3/\sqrt{2}$, so our surface integrals are probing the wave function well outside the Coulomb zone although it is being approached at the lowest energy. It appears that the dominance of σ_W is diminished as we leave this zone. In order to quantify this further, we calculated TICS for the fixed value of $RE=7$ over the whole energy range. The TICS with this setting for R have not converged, of course, to the same accuracy as previously, now having errors up to 5% at the higher energies. As is evident from Fig. 1, the dominance of the Wannier region as the Coulomb zone is neared is firmly established by our calculations. It is also evident that at constant RE the dominance of σ_W over σ_T increases from one order of magnitude at 0.2 a.u. to two orders of magnitude at 0.005 a.u., demonstrating the increasing dominance of the Wannier region as the ionization threshold is approached. Using our method for calculating the hyperradius R_0 required to achieve convergence of our results to within 0.2%, we estimate that R_0 would only fall within the Coulomb zone for energies below 10^{-5} a.u. It is only in this region that the arguments of Rau will rigorously hold and give TICS to the specified accuracy. It is also worth pointing out that the convergence of the TICS is much more rapid with R than for σ_W and σ_T separately, as evidenced from Figs. 1 and 2.

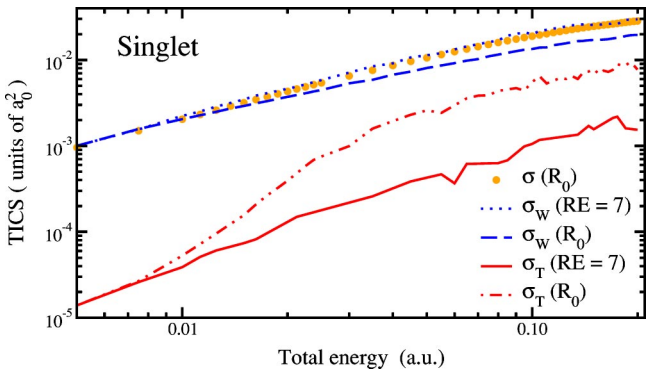

FIG. 1. ^1S TICS using the full amplitude f, the Wannier amplitude f_W, and the Temkin amplitude f_T for the collinear model with spin weighting included. σ_W and σ_T are calculated at R_0 (where convergence of σ is achieved) and also at an energy-dependent hyperradius R, such that $RE=7$, outside Rau's [3] Coulomb zone.

The TICS for the ^3S case was calculated using a hyperradius of 800 a.u. for all energies, and is presented in Fig. 2. Near threshold they are similarly influenced by the contributions from the Wannier region, but to a lesser extent than the singlet TICS, due to the suppression of the amplitude at

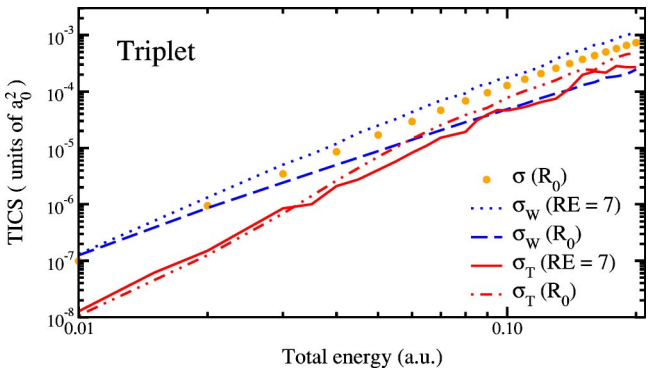

FIG. 2. ^3S TICS for the collinear model with spin weighting included. See Fig. 1 for explanation of symbols.

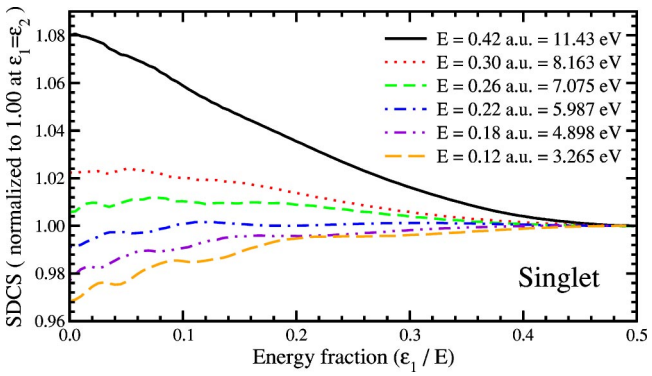

FIG. 3. Collinear ^1S SDCS, normalized to 1.00 at $\epsilon_1=\epsilon_2$.

$r_1=r_2$. Maintaining a constant energy-dependent hyperradius of $RE=7$ for the triplet case also maintains the dominance of the Wannier region at all energies considered. Like the singlet case, this gives evidence that the $r_1\approx r_2$ region, though suppressed, is also dominant within the Coulomb zone.

In Fig. 3, the shape of the SDCS at various energies is shown. At 0.22 a.u. the SDCS is flat, apart from very slight oscillations. Below this energy, there is an increasing reduction in the SDCS at unequal energy sharing as the total energy approaches threshold. At 0.12 a.u., the reduction of 3% is consistent with the semiclassical calculations of Rost [20]. However at higher energies, the SDCS remains flatter than that calculated by Rost. Our calculations were performed at similar energies to Rost to facilitate comparison with his publication. The relatively flat SDCS presented here are in line with the predictions of Rau [3].

Plotted in Fig. 4 are our singlet TICS calculations, where all datasets have been divided by $E^{1.127}$, to emphasize their threshold behavior. There is good agreement between our results and those of Kato and Watanabe [7] above 0.02 a.u., and our results showed significantly reduced energy-dependent fluctuations. Also shown are the recent ECS calculations of McCurdy, Horner, and Rescigno [13] and time-dependent calculations of Robicheaux, Pindzola, and Plante [21].

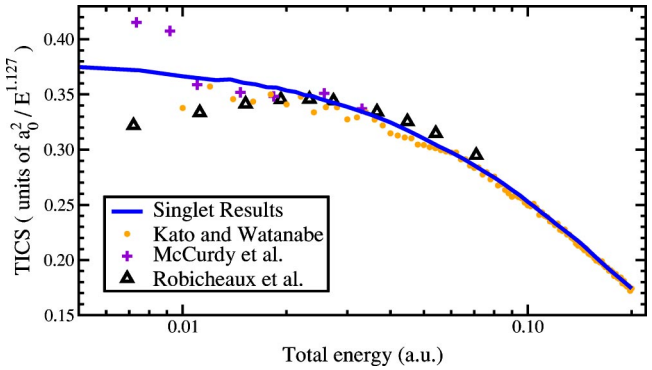

FIG. 4. Singlet $L=0$ TICS for the collinear model with spin weighting (divided by $E^{1.127}$). These results are compared with those of Kato and Watanabe [7], McCurdy, Horner, and Rescigno [13], and Robicheaux, Pindzola, and Plante [21].

In order to determine the near-threshold law for the TICS, we employed the same procedure as Kato and Watanabe [7], and fitted to the function $\sigma=E^{\alpha}g(E)$, where an nth-order series expansion of $g(E)$ was used. The value of α, and its error, is dependent upon both n and the error estimate given to each point. Kato and Watanabe did not provide details of their error estimating procedure. We performed a nonlinear fit based upon the estimated maximum errors from our convergence testing, and then partitioned the points into four energy regions. The standard deviation of the points in each region was used as an improved estimate for their absolute error. The fitting procedure was performed iteratively, using the latest error estimates, until convergence of the coefficients was obtained. The resultant nonlinear fit for the ^1S TICS is

$$\sigma_{S=0}=E^{(1.128\pm0.004)}[(0.386\pm0.007)-E(1.69\pm0.08) +E^2(4.1\pm0.5)-E^3(4.6\pm1.1)]. \quad (5)$$

The fit presented above was made for the energy range 0.005–0.20 a.u., which matched nonlinear fits of subsets of the data over the energy ranges 0.005–0.10 a.u. and 0.005–0.05 a.u. It also matched linear least-squares fits of the (transformed) data over these energy ranges, within their calculated standard errors. With higher-order fitting functions, the higher-order coefficients became very large, with significant errors, which indicated that the fitting procedure was overly influenced by numerical errors in our results. The leading term for the singlet TICS is $E^{1.128\pm0.004}$ which is very close to the classical Wannier [1] result of $E^{1.127}$, with a threefold improvement in the standard error compared with Kato and Watanabe's calculation. It accounts for 96% of the cross section at $E=0.01$ a.u. rising to 99% at $E=0.0025$ a.u. The conclusion of McGowan and Clarke [2] that the Wannier [1] region is within 0.4 eV of threshold is, within experimental error, consistent with our calculations.

These fitting procedures were performed, for completeness, on the other datasets shown in Fig. 4. The results for our fits are $E^{1.228\pm0.074}$ for McCurdy, Horner, and Rescigno [13] and $E^{1.197\pm0.01}$ for Robicheaux, Pindzola, and Plante [21]. The fit published by Kato and Watanabe [7] was $E^{1.124\pm0.013}$.

Due to the highly suppressed TICS of the $L=0$ triplet at low energies, linear fitting procedures were used on the transformed data in preference to nonlinear methods, giving an estimated threshold behavior of $E^{3.37\pm0.02}$, which is in excellent agreement with the semiclassical calculation by Peterkop [22] of $E^{3.38}$. The estimated error is larger than the singlet error due to the limited hyperradii used for the low-energy calculations.

In conclusion, our fully quantal study has yielded several new insights into the physics of the near-threshold ionization. We explored the regions of configuration space which are important in obtaining accurate TICS and were able to confirm that the important region that emerges from a fully quantal calculation is the $r_1\approx r_2$ one, but only while in the Coulomb zone. However, the evaluation of the surface integrals to obtain accurate amplitudes requires hyperradii well

outside the Coulomb zone. We have provided further evidence for the Wannier threshold law for ^1S scattering and its range of applicability given by Eq. (5). In particular, the nonzero linear term with E indicates that the $E^{1.127}$ functional form is only valid strictly at the threshold, which is consistent with the analysis of Rost [20]. The accuracy of our numerical methods enabled us to describe the nonclassical threshold behavior of ^3S scattering. A theoretical understanding of the emergence of a Wannier-like threshold behavior for ^3S TICS, with exponent three times the Wannier value, has been given by Peterkop [22]. Finally, we provided an independent confirmation of the ECS methodology and demonstrated that it can yield extremely accurate TICS near threshold.

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