Absolute Differential Cross Sections for Photo Double Ionization of Helium from the *Ab Initio* Hyperspherical *R*-Matrix Method with Semiclassical Outgoing Waves

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The hyperspherical R-matrix method with semiclassical outgoing waves introduced by us previously [Phys. Rev. A **60**, 3667 (1999)] is applied to the one-photon double ionization of the He atom. The absolute differential cross sections obtained are in excellent agreement with experiment for the very challenging dynamical situations considered. This method is the first one to compute the final double continuum state accurately over the entire configuration space, from the vicinity of the nucleus to the asymptotic region.

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The theoretical treatment of correlations in the double electronic continuum is a long-standing challenge in atomic physics. The derivation by Wannier [1] of the threshold law for the double electron escape cross section was the first success ever obtained on this problem. Since then, a lot of efforts have been devoted to the study of the simplest processes leading to a double continuum electronic state: photo double-ionization (PDI) of He, and electron-impact ionization of H. In the mid-1990s, absolute triply and singly differential cross sections (TDCS and SDCS) as well as integrated cross sections (ICS) in fair agreement with experiment were obtained using the 2SC (2 screened Coulomb) method [2,3]. This method, however, was not pushed further. By contrast, the CCC (convergent close coupling) method [4] has accumulated an extensive set of data over the last eight years [5]. The TDCS obtained agree very well with experiment in *shape* if one disregards their poor account of the well-known node for antiparallel emission at equal energy sharing. However, their agreement in magnitude follows from an *a posteriori* rescaling procedure, based on the value of the ICS, and on the assumption of a flat SDCS. An *ab initio* rescaling procedure has been proposed recently to overcome this limitation in the case of equal energy sharing [6]. Yet the unequal energy sharing case remains unsolved, and the method still fails to produce sensible SDCS, although it yields the correct ICS from the optical theorem. The reason for these drawbacks has been shown by Rescigno *et al.* [7] to lie in the extraction of the cross section, which is based on an asymptotic form implying that a single electron escapes to infinity.

Very recently, other methods have appeared, which deal with the full complexity of the double escape dynamics transparently and without introducing oversimplifying approximations. They represent an important step towards the resolution of the problem. The time-dependent method of Pindzola and Robicheaux [8] requires propagating the wave function long enough, which means far enough as well, to extract the cross section accurately. The computer power currently available has limited application of this method to either computing integrated cross sections for long range Coulomb potentials [8], or computing differential cross sections for short range potentials [9], as integrated cross sections converge faster than differential ones. Rescigno *et al.* [10], by contrast, have obtained the double continuum wave function by solving a stationary inhomogeneous equation subject to outgoing wave boundary conditions. Those authors have used exterior complex scaling that circumvents the need to formulate detailed asymptotic conditions and allows one to compute the wave function accurately within an $r_1 \times r_2$ square of a few hundreds a.u. side. The cross sections are extracted by applying an extrapolation procedure to the computed outgoing flux. The resulting method, although less demanding numerically than the time-dependent approach by Pindzola and Robicheaux [8], still relies upon intense parallel computing. It has been applied to electron-impact ionization of H with a remarkable success.

Here we report the first results obtained by applying the hyperspherical $\mathcal{R}$ matrix method with semiclassical outgoing waves (H$\mathcal{R}$M-SOW) to the PDI of He. The principle of the H$\mathcal{R}$M-SOW method being described in [11], we recall only its main features here. Taking advantage of the periodicity in time of the atom-field interaction, we use the Floquet theorem [12] to transform the time-dependent Schrödinger equation into a set of stationary coupled partial differential equations (PDE). In the weak field limit, this set reduces to the single inhomogeneous stationary equation

$$(E_0 + \omega - H_0)\Psi = V\Psi_0\,, \tag{1}$$

subject to outgoing wave conditions, with H_0 the field-free Hamiltonian, E_0 and Ψ_0 the He ground state energy and wave function, ω the photon frequency, and V the

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time-independent part of the atom-field interaction in the dipole approximation. The wave function at large time being a superposition of Ψ_0 and Ψ with the time-oscillating phase factors associated with the energies E_0 and $E_0 + \omega$, the PDI cross sections can be derived from the asymptotic behavior of Ψ. Note that a finite range approximation of this very function Ψ was involved in the 2SC approach. Note also the analogy between our formulation of the PDI problem and the formulation by Rescigno *et al.* [10] of the electron-impact ionization problem.

Our method uses hyperspherical coordinates, namely the hyperradius $R = \sqrt{r_1^2 + r_2^2}$, the angle of radial correlation $\alpha = \arctan r_1/r_2$, the angle of angular correlation $\theta_{12} = (\vec{r}_1, \vec{r}_2)$, complemented by a set of three Euler angles denoted collectively by Ω. This coordinate system, well suited to the description of a correlated motion, is numerically very convenient as only one variable is of infinite range. Projecting the $^1P^o$ wave function Ψ onto the appropriate Wigner functions $D^{1\pm}_{01}(\Omega)$ [13–15] eliminates the Euler angles from the treatment at the cost of introducing the two components $\Phi^{\pm}(R, \alpha, \theta_{12})$. In the following, we shall omit the $\pm$ labels for simplicity, and treat these two components as a unique vector.

Next, we separate configuration space into the inner region $R \leq R_0$ and the outer region $R > R_0$. From Eq. (1) and its homogeneous and Hermitian counterpart defined over the inner region by adding the usual Bloch operator [16], we can easily derive an $\mathcal{R}$ matrix relation at the boundary R_0. We express this relation in the adiabatic angular basis obtained by diagonalizing H_0 at fixed $R = R_0$. Expanding $\Phi(R, \alpha, \theta_{12})$ on the adiabatic angular eigenvectors $X_M(R_0; \alpha, \theta_{12})$ defines locally uncoupled hyperradial channel functions $F_M(R)$, the evolution of which is governed by the adiabatic potentials $E_M(R)$. These channel functions satisfy the $\mathcal{R}$ matrix condition

$$F'_M(R_0) = \sum_{M'} \mathcal{R}_{MM'} F_{M'}(R_0) + V_M(R_0), \qquad (2)$$

where the vector with components V_M is related with the inhomogeneous term in Eq. (1). Next, we approximate each $F_M(R)$ at $R_0 < R < R_0 + \epsilon$ according to the JWKB (Jeffreys, Wentzel, Kramers, and Brillouin) approximation by the product of a slowly varying function of R with a semiclassical outgoing wave associated with the momentum $p_M(R) = \sqrt{2[E - E_M(R)]}$, with E the total excess energy above threshold. A second relation between these functions and their radial derivatives follows:

$$F'_M(R_0) = \imath p_M(R_0) F_M(R_0). \qquad (3)$$

Solving Eqs. (2) and (3) yields the solution of Eq. (1) at the boundary, provided R_0 is chosen large enough for the JWKB approximation to be valid in the external region.

This solution is found to be dominated by the $M = 0$ component. This suggests for us to look for the solution for $R > R_0$ in the simpler form

$$\Phi(R, \alpha, \theta_{12}) = \frac{1}{\sqrt{p_0(R)}} e^{\imath \int_{R_0}^{R} p_0(R')\, dR'} \phi(R, \alpha, \theta_{12}), \quad (4)$$

where $\partial^2\phi/\partial R^2 \simeq 0$, so that ϕ satisfies a PDE of first order over R. This establishes a formal analogy with a time-dependent problem. However, by contrast with the time propagation of a function of two *radial* variables [8], the hyperradial propagation of a function having a twofold dependence on *angular* variables defined over a finite range is not limited by the computer resources: we are thus able to propagate the wave function up to the true asymptotic region where the cross sections are extracted directly by computing the outgoing flux.

The results we present here have been obtained at 20 eV above threshold, near the maximum of the ICS, where small variations in E hardly affect the dynamics. This allows us to compare them with experiments performed not only *at* but also *near* this energy. All cross sections have been extracted at $R = 10^5$ a.u. where they have clearly converged. They have been computed in length, velocity, and acceleration gauge: in all cases, the differences were smaller than the width of the representative lines. We emphasize that the cross sections obtained are given on an absolute scale and their calculation does not involve any adjustable parameter or *a posteriori* rescaling factor.

The relative TDCS of Mazeau *et al.* [17] and the absolute TDCS of Bräuning *et al.* [18] were recorded at nearby total energies, 18.6 and 20 eV, respectively, and for energy ratios $\rho = E_1/E_2$ of 8.3, 1, 0.12 and 5.67, 1, 0.18, respectively. The photon beam, directed along $\hat{k}$, was linearly polarized along the main axis $\hat{\epsilon}$. The value of the Stokes parameter S_1, 0.98 in [18], did not matter in [17]. One electron was ejected along $\hat{\epsilon}$, and the TDCS was recorded as a function of $\theta_2 = (\hat{\epsilon}, \hat{r}_2)$, with $\vec{r}_2$ scanning the $(\hat{\epsilon}, \hat{k})$ plane in [17], and the plane orthogonal to $\hat{k}$ in [18]. These equivalent geometrical configurations have been recognized as the most challenging for the theory in unequal energy sharing situations. The relative data of Mazeau *et al.* [Figs. 1(a)–1(c)] have been rescaled to our calculated TDCS: their agreement with the theory, as far as the shapes are concerned, is very satisfying at all energy sharings. The absolute data of Bräuning *et al.* [Figs. 1(d)–1(f)] show that our theory also yields the correct absolute values of the cross sections. Note, however, the difference in shape between theory and experiment in Fig. 1(e). It is worth pointing out that the data of Mazeau *et al.* have been questioned on the basis of Bräuning's results [18]. The present calculations reconcile the two sets of measurements, by proving that a single theory is compatible with both sets. They also show that the ratio between the central lobe and the two side lobes, which determines the shape of the unequal energy sharing TDCS, is a very sensitive function of the energy ratio ρ.

Before H$\mathcal{R}$M-SOW was developed, the only approach which had ever produced a full set of absolute TDCS,

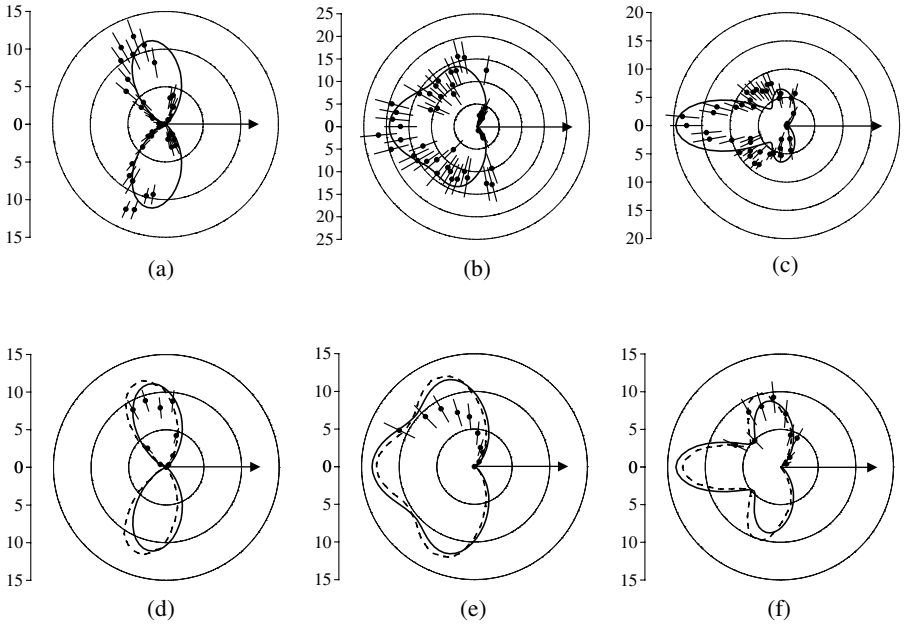

FIG. 1. Polar plots of the TDCS in $b(\mathrm{eV})^{-1}(sr)^{-2}$ versus the emission angle of the second electron. The first electron is emitted along the linear polarization axis indicated by the arrow. Points with error bars: experiments from [17] with $\rho = 1$ (a), 8.3 (b), 0.12 (c), and from [18] with $\rho = 1$ (d), 5.67 (e), and 0.18 (f). Continuous lines: present calculations (length, velocity, and acceleration superimposed); dashed lines: 2SC results [3].

SDCS, and ICS in agreement with experiment was 2SC. This is why we restrict our comparison with other theories to this single method. Theoretical 2SC results are available at $E = 18.6$ eV and $\rho = 5.2, 1, 0.19$ [3]. As the dynamics depends much more on ρ than on E, they can be compared with the results displayed at the bottom of Fig. 1. The agreement in *shape* and in absolute *magnitude* between the 2SC and H$\mathcal{R}$M-SOW theories is remarkable at all energy sharings.

Figure 2 shows the excellent agreement between our single differential cross section (SDCS) and the measurements of Wehlitz *et al.* [19]. Our very flat computed cross section is symmetrical with respect to $E/2$, as it must be, since the two electrons are treated on an equal footing throughout configuration space. It exhibits very dramatic oscillations in an energy domain of the order of $E\Delta r/R$ around 0 and 20 eV, where Δr is the range of the He^+ Rydberg states populated by the competing process of single ionization with excitation. For the large value of the hyperradius ($R = 10^5$ a.u.) where the cross sections have been extracted, these oscillations are confined within such a short range that they cannot be distinguished from the left and right vertical axis of Fig. 2. Small oscillations are superimposed on the flat pattern: they would be smeared out by further increasing the size of the α basis set used in our inner region calculations. Note that similar oscillations affect the 2SC data [2]. These 2SC results are in excellent agreement with ours, if they are corrected for a factor 2 error which has been identified by the authors later on [20].

The integrated cross section can be deduced from the SDCS of Fig. 2 provided an extrapolation is performed at the boundaries to get rid of the strong oscillations mentioned above. Clearly, the plateau at 0.9 $\mathrm{kb(eV)}^{-1}$ in the SDCS is consistent with the reference experimental 8.76 kb value of the ICS given by Samson *et al.* [21], which has been used by Bräuning *et al.* [18] to calibrate their differential data.

The H$\mathcal{R}$M-SOW method is then capable of producing a full set of differential and integrated cross sections which agree with experiment in shape as well as in magnitude in a very challenging dynamical situation. It is the first

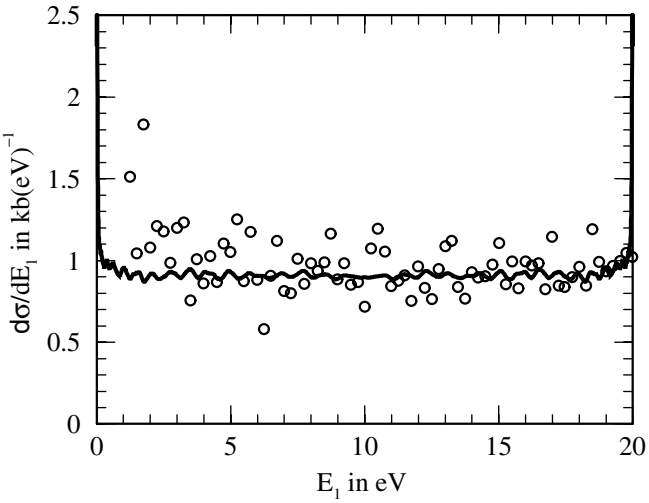

FIG. 2. SDCS in $\mathrm{kb(eV)}^{-1}$ versus the energy of one electron in eV. Continuous line: present calculations. Circles: experimental results from [19].

method which is able to compute the correct double continuum final state over the entire configuration space from the nucleus to the far asymptotic region. This allows us to demonstrate the substantial role of both the region close to the nucleus and the external region in forming the various cross sections. This specificity of our method is also the key to the success of *ab initio* studies at very low energies, where other methods fail to converge.

It is noteworthy that this method does not rely upon supercomputing facilities: the present results have been obtained on a PC running under Linux and equipped with a Pentium III 400 Mhz and 768 MB RAM. A typical exploitation requires 18 h and the full memory of the computer.

Let us emphasize, to conclude, that the power of the method results from its combined use of two theoretical tools which never met before: the $\mathcal{R}$ matrix method, which is closely linked with L^2 numerical techniques, and the JWKB approximation, which has a natural relationship with time-propagation techniques. In a forthcoming publication, we will present more extensive results, including the $a_g(E_1, E_2, \theta_{12})$ and $a_u(E_1, E_2, \theta_{12})$ parametrization functions introduced by Huetz *et al.* [22].

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