Hyperspherical time-dependent method with semiclassical outgoing waves for double photoionization of helium

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The hyperspherical time-dependent method with semiclassical outgoing waves for study of *double photoionization* of helium is presented. It is closely related to the hyperspherical *R*-matrix method with semiclassical outgoing waves [Phys. Rev. A **65**, 032711 (2002)]: both split configuration space into two regions to solve the stationary inhomogeneous Schrödinger equation associated with the one-photon ionization problem, and both apply the same treatment to the outer region. However, the two methods differ radically in their treatments of the problem in the inner region: the most recent one applies a time-dependent approach for calculating the stationary wave function, while the previous one uses a *R*-matrix treatment. The excellent agreement observed between the triple differential cross sections obtained from these two basically different methods provides very strong support for both of them. Importantly, the very different numerical structures of both methods might make the most recent one a better candidate for investigating the near-threshold region.

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The challenge for a precise theory of the double photoionization (DPI) of He has been enhanced by the considerable experimental progress achieved in the late 1990s. The absolute values of triple differential cross sections (TDCS) of He were first measured in 1995 by a direct method that underestimated the cross section by a factor of two [1]. More recently, indirect methods, requiring the measurement of all coincidence events and renormalizing the TDCS to the known total ionization cross section have provided reliable results [2–4].

On the theoretical side, the main difficulty in the study of three-body Coulomb problems consists in imposing the outgoing wave boundary condition to the solution of the stationary Schrödinger equation. The most obvious way to circumvent this difficulty is to switch to a nonstationary approach. The time-dependent close coupling (TDCC) method was first applied to electron-impact ionization of H [5]. In 2001, the authors considered the DPI process in He [6], and later on, in a variety of many-electron and/or excited systems. In 2003, another nonstationary scheme for the study of electron-impact ionization of hydrogen has been put forward, which seems very promising [7].

This difficulty can also be circumvented within the framework of stationary approaches, as demonstrated by the exterior complex scaling (ECS) theory presented by the Livermore group in 1999 [8], and by the hyperspherical *R*-matrix with semiclassical outgoing waves (H*R*M-SOW) method that we have put forward in 2000 [9,10]. The first of these theories, based on a complex rotation technique, has been applied to electron-impact ionization of H, while the second of these theories, based on a combination of *R*-matrix and semiclassical techniques, has been applied to DPI of He.

However, the idea that some mixing of time-dependent and stationary techniques could lead to still more powerful numerical tools had emerged earlier. Namely, the basic ideas for applying a time-dependent approach to the calculation of two-electron stationary wave functions were formulated in 1997 [11,12]. Recently, a similar approach has been proposed by the Livermore group [13]. Here, we apply the ideas of Refs. [11,12] to the DPI problem.

The basic features of the H*R*M-SOW and hyperspherical time-dependent with semiclassical outgoing waves (HTD-SOW) methods are the same. Both methods formulate the one-photon ionization problem in terms of the stationary inhomogeneous Schrödinger equation

$$[\mathcal{H}(\vec{r}_1,\vec{r}_2)-E]\Phi_E(\vec{r}_1,\vec{r}_2)=\Xi_0(\vec{r}_1,\vec{r}_2), \tag{1}$$

where $(\vec{r}_1,\vec{r}_2)$ are the radius vectors of the electrons with respect to the nucleus, $\mathcal{H}$ is the two-electron Hamiltonian, E is the total energy of the system, and Φ_E is the double continuum wave function. The source term on the right-hand side reads

$$\Xi_0(\vec{r}_1,\vec{r}_2)=-\tfrac{1}{2}\vec{\mathcal{E}}_0\cdot\vec{D}\Psi_0(\vec{r}_1,\vec{r}_2), \tag{2}$$

where $\vec{\mathcal{E}}_0$ is the amplitude of the electric-field vector $\vec{\mathcal{E}}(t)=\vec{\mathcal{E}}_0\cos\omega t$, $\vec{D}$ is the dipole operator, and Ψ_0 is the ground-state wave function of He. Both methods define two spatial regions depending on the value of the hyperradius $R=\sqrt{r_1^2+r_2^2}$ compared to some critical value R_0. The region $R\leqslant R_0$ contains the singularity of the three-body interaction potential at $R=0$. In the external region $R>R_0$, by contrast, the evolution of this potential with R is smooth. These different characteristics of the two regions call for different mathematical treatments. In the external region, both methods apply the same approach, obtained as a substantial development of the extended Wannier ridge method (EWRM) [14]. Namely, by treating the *R*-motion semiclassically, the exact Schrödinger equation, which is second order over R, is transformed into an equation of first order over a *mock time*

related to R. This equation has the formal structure of a nonstationary Schrödinger equation, the initial condition of which is provided by the treatment of the inner region. It is solved using a very powerful and stable unitary algorithm that can provide the solution at arbitrary large distances with very high accuracy.

Within the HRM-SOW theory [9,10], we treat the internal region using an R-matrix treatment complemented by an outgoing adiabatic partial-waves condition at $R=R_0$.

Presently we consider the internal region with a radically different approach based on the fact that the solution of a stationary nonhomogeneous Schrödinger equation like Eq. (1), subjected to the outgoing wave boundary condition, can be obtained exactly as the Fourier-Laplace transform

$$\Phi_E(\vec{r}_1,\vec{r}_2)=\int_0^\infty dt\Psi(\vec{r}_1,\vec{r}_2;t)\exp(\imath Et) \tag{3}$$

of the solution of the associated Cauchy problem

$$\imath\frac{\partial}{\partial t}\Psi(\vec{r}_1,\vec{r}_2;t)=\mathcal{H}(\vec{r}_1,\vec{r}_2)\Psi(\vec{r}_1,\vec{r}_2;t), \tag{4a}$$

$$\Psi(\vec{r}_1,\vec{r}_2;0)=\imath\Xi_0(\vec{r}_1,\vec{r}_2). \tag{4b}$$

In our case, the ${}^1P^o_{M=0}$ two-electron function can be expanded over the bipolar harmonics as

$$\Psi(\vec{r}_1,\vec{r}_2;t)=\sum_{l_1,l_2=0}^{\infty} Y^{(1,0)}_{l_1,l_2}(\hat{r}_1,\hat{r}_2)\frac{F^{(1)}_{l_1,l_2}(r_1,r_2;t)}{r_1r_2}. \tag{5}$$

Note that the study of the $M=0$ case provides the basis for the treatment of any experimental situation. The symmetry of the wave function in the exchange of the two electrons is imposed on the initial wave function, as shown below. The nonstationary equations for the coefficient functions $F^{(1)}_{l_1,l_2}(r_1,r_2;t)$ read

$$\imath\frac{\partial}{\partial t}F^{(1)}_{l_1,l_2}(r_1,r_2;t)=(\mathcal{H}_{l_1}(r_1)+\mathcal{H}_{l_2}(r_2))F^{(1)}_{l_1,l_2}(r_1,r_2;t)+\sum_{l_1',l_2'}\mathcal{V}^{(1)}_{l_1,l_2,l_1',l_2'}(r_1,r_2)F^{(1)}_{l_1',l_2'}(r_1,r_2;t), \tag{6}$$

where

$$\mathcal{H}_l(r)=-\frac{1}{2}\frac{\partial^2}{\partial r^2}+\frac{l(l+1)}{2r^2}-\frac{Z}{r}, \tag{7}$$

and

$$\mathcal{V}^{(L)}_{l_1,l_2,l_1',l_2'}(r_1,r_2)=(-1)^{(L+l_2+l_2')}\sqrt{(2l_1+1)(2l_2+1)(2l_1'+1)(2l_2'+1)}\times\sum_{k=0}^{\infty}\frac{r_<^k}{r_>^{k+1}}\begin{pmatrix} l_1 k l_1' \\ 000\end{pmatrix}\begin{Bmatrix} L l_2' l_1' \\ k l_1 l_2\end{Bmatrix}\begin{pmatrix} l_2 k l_2' \\ 000\end{pmatrix}. \tag{8}$$

The initial condition for the functions $F^{(1)}_{l_1,l_2}(r_1,r_2;t)$ is obtained from Eqs. (4b) and (2) by introducing the expansion of the ground-state wave function Ψ_0 of He over bipolar harmonics, the coefficient functions of which are denoted as $F^{(0)}_{l_1,l_2}(r_1,r_2)$. For a unit amplitude of the electric field and if the dipole operator is expressed in the length gauge, one obtains after some algebra

$$F^{(1)}_{l_1,l_2}(r_1,r_2;0)=\imath\sum_{l_1',l_2'}\mathcal{W}^{(1,0)}_{l_1,l_2,l_1',l_2'}(r_1,r_2)F^{(0)}_{l_1',l_2'}(r_1,r_2), \tag{9}$$

where

$$\mathcal{W}^{(LL')}_{l_1,l_2,l_1',l_2'}(r_1,r_2)=r_1\delta_{l_2,l_2'}(-1)^{l_2}\times\sqrt{(2l_1+1)(2l_1'+1)}\times\sqrt{(2L+1)(2L'+1)}\begin{pmatrix} l_1 1 l_1' \\ 000\end{pmatrix}\times\begin{Bmatrix} l_1 l_2 L \\ L' 1 l_1'\end{Bmatrix}\begin{pmatrix} L 1 L' \\ 000\end{pmatrix}+\{1\Leftrightarrow 2\}, \tag{10}$$

which ensures the symmetry of the singlet initial wave function in exchange of the two electrons.

Our numerical procedure comprises two stages: (i) determining the expansion coefficients $F^{(0)}_{l_1,l_2}(r_1,r_2)$ of the ground state Ψ_0 of the system over bipolar harmonics and (ii) solving the nonstationary Eq. (6) with the initial condition given by Eq. (9). The Laplace transform of the solution according to Eq. (3) then yields the double continuum wave function $\Phi_E(r_1,r_2)$ at any desired total energy E. Note that within our approach, it is enough to calculate $\Phi_E(r_1,r_2)$ on the sphere $R=R_0$.

To determine the He ground-state wave function, we have propagated in imaginary time the initial function taken in the form proposed by Le Sech [15]. The details of this procedure were described in Ref. [12]. Here however, the radial distances of the two electrons have been discretized on another nonuniform mesh that is defined by

$$r_i^k=\frac{1}{Z}[x_k/a-\arctan(x_k/a)]a^3, \quad i=1,2 \tag{11}$$

with $x_k=kdx$, $k=1,\ldots,200$, $dx=0.06$ a.u., and $a=2.6$ a.u. The values of the parameters dx and a were chosen so that the nonuniform mesh covers the vicinity of the

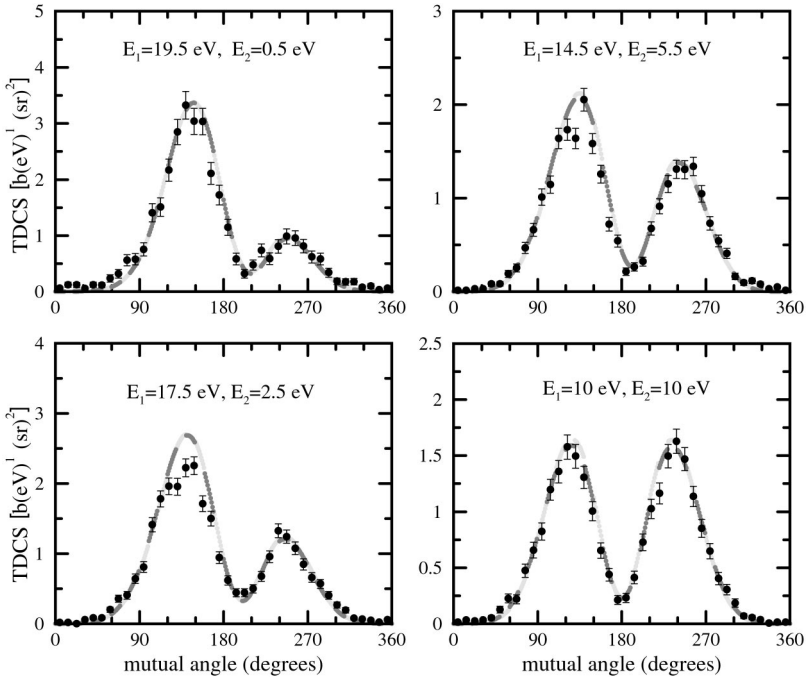

FIG. 1. (Color online) TDCS obtained at 20 eV above threshold using circular polarization in the plane perpendicular to the photon beam vs the mutual angle of the two electrons. The respective energies E_1 and E_2 of the two electrons are indicated in the figures. Dashed blue lines: H*R*M-SOW calculations; dashed red lines: HTD-SOW calculations; dots with error bars: measurements [4].

nucleus extremely densely, and the distant region more sparsely: we thus get $\delta r_i \simeq 10^{-4}$ a.u. at the origin and $\delta r_i \simeq 0.2$ a.u. at the border $r \simeq 28$ a.u. By including five angular configurations $[l,l]$, $l=0,1,2,3,4$, and taking the time propagation step as $\delta t = 0.05$ a.u., the procedure converges in 300 steps. The energy of the eigenstate obtained is equal to -2.9039 a.u., the experimental value being -2.9036 a.u. [16].

The second stage of our numerical procedure is performed with the same nonstationary routine as the first one, but with propagation in real time with $\delta t = 0.08$ a.u. using a 500 points mesh that covers the area about 88×88 a.u. Ten angular configurations, $[l,l+1]$ and $[l+1,l]$ for $l=0,\ldots,4$ are included. The stationary wave functions have been monitored on the sphere $R_0 = 60$ a.u., the largest one ever used in H*R*M-SOW theory [10]. The number of time steps needed for computing the wave function at 20 eV above threshold was 10^4.

The main difficulty in solving stationary problems with more than one dimension consists in imposing outgoing boundary condition. If these stationary problems are treated using the nonstationary scheme defined by Eqs. (3,4), this becomes rather trivial. One just adds an absorbing auxiliary potential concentrated at the box boundary, which effectively damps the wave function in that region to zero. It seems widely accepted that Ignatowsky [17] was the first to relate the outgoing boundary condition to the infinitesimal absorption. In our case the absorbing imaginary potential consists of three terms: $g(r_1,r_2) = -ig_S(r_1,r_2) - ig_a(r_1,r_2) - ig_a(r_2,r_1)$. All the terms absorb the waves outside the sphere $R_1 = R_0 + 5$ a.u.. The first term provides uniform absorption of the wave packet in that region, while the other two terms are concentrated in the vicinity of the axes and absorb the short wavelets corresponding to the singly ionized states of He.

It is important to realize that our present approach and the TDCC method [5,6] differ *essentially*. First, if both methods use *nonstationary techniques*, they apply these techniques to different formulations of the DPI problem: the *stationary* one in the HTD-SOW case and the *nonstationary* one in the TDCC case. One intrinsic advantage of the stationary formulation is that it provides a function of precisely-defined energy. On the other hand, an intrinsic difficulty of the nonstationary formulation is that it introduces an arbitrary modelization of the time envelope of the radiation pulse. Second, the techniques used are very different. The use of an absorbing potential is one great advantage of our method since it allows us to continue the wave propagation as long as we wish. On the contrary, TDCC implies terminating propagation before the waves reflected from the box boundaries alter the outgoing wave packet: this leads to a poor energy definition within the nonstationary formulation of the DPI used in this method. Additionally, we use the split propagation technique with a nonuniform spatial grid and Cranck-Nickolson [18] partial propagators. This allows us to reach at least three significant digits in the values of Φ_E on the hypersphere $R = R_0$, and to obtain the initial wave function with 10^{-4} relative precision of energy, which should be compared with the 1% accuracy accepted in Ref. [6]. The reason for these contrasted achievements of both methods is that the staggered leap-frog propagation scheme used in Refs. [5,6] is stable only if the time step remains smaller than the square of the smallest spatial step [19]. Therefore, it cannot be easily adapted to the work with dense grids.

To demonstrate the power of the present model, we compare its results with the corresponding measurements in the case of the experiment done by Achler *et al.* [4] at 20 eV above threshold. The latter is selected because it provides TDCS values on the absolute scale with a high statistics and can thus be considered a benchmark in the field. Figure 1 shows that the HTD-SOW results agree with experiment perfectly. Moreover, the HTD-SOW results coincide with the

H*R*M-SOW ones obtained from the most recent version of this theory [10]. In additional calculations, performed in the conditions of the experiment done by Seccombe *et al.* at 25 eV above threshold [10,20], the HTD-SOW and H*R*M-SOW results show mutual deviations of the order of the experimental error bars for two kinematics. Thus, we are meeting a rare case in the theoretical physics when two substantially different approaches give practically identical results. This constitutes very strong support for both methods.

However, preliminary calculations at 1 eV above threshold, where no measurements are available yet, show more substantial deviations of HTD-SOW from H*R*M-SOW. Therefore, further work will concentrate on this region, where HTD-SOW might possibly supersede H*R*M-SOW. Going down in energy is indeed very likely to require a substantial increase of the size of the inner region that is common to the two methods. Both methods might well not face this demand with the same ease. Namely, the ''Achilles heel'' of H*R*M-SOW is that it relies upon the diagonalization of an $N \times N$ matrix the structure of which does not allow any storage saving. As the diagonalization run-time scales as N^3 [18] and storage requirements as N^2, the risk of running into the computer's limits regarding either the available memory or the allowed computing time are significant. On the other side, the storage requirements of HTD-SOW scale as N, and so do the time requirements for a single propagation step. The only difficulty that could arise with HTD-SOW is a substantial increase of the number of propagation steps required leading to a corresponding increase in the runtime. However, it seems that we are in a good position to take up the threshold region challenge in the near future, which no *ab initio* method has ever attempted so far. This makes absolute complete experiments below 5 eV excess energy all the more desirable.

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[1] O. Schwarzkopf and V. Schmidt, J. Phys. B **28**, 2847 (1995).
[2] H. Bräuning, R. Dörner, C.L. Cocke, M.H. Prior, B. Krässig, A.S. Kheifets, I. Bray, A. Bräuning-Demian, K. Carnes, S. Dreuil, V. Mergel, P. Richard, J. Ullrich, and H. Schmidt-Böcking, J. Phys. B **31**, 5149 (1998).
[3] A. Huetz and J. Mazeau, Phys. Rev. Lett. **85**, 530 (2000).
[4] M. Achler, V. Mergel, L. Speileberger, R. Dörner, Y. Azuma, and H. Schmidt-Böcking, J. Phys. B **34**, 965 (2001).
[5] M.S. Pindzola and F. Robicheaux, Phys. Rev. A **54**, 2142 (1996).
[6] J. Colgan, M.S. Pindzola, and F. Robicheaux, J. Phys. B **34**, L457 (2001); J. Colgan and M.S. Pindzola, Phys. Rev. A **65**, 032729 (2002).
[7] L. Mouret, K.M. Dunseath, M. Terao-Dunseath, and J.M. Launay, J. Phys. B **36**, L39 (2003).
[8] T.N. Rescigno, M. Baertschy, W.A. Isaacs, and C.W. McCurdy, Nature (London) **286**, 2474 (1999); M. Baertschy, T.N. Rescigno, W.A. Isaacs, X. Li, and C.W. McCurdy, Phys. Rev. A **63**, 022712 (2001); C.W. McCurdy, D.A. Horner, and T.N. Rescigno, *ibid.* **63**, 022711 (2001).
[9] L. Malegat, P. Selles, and A.K. Kazansky, Phys. Rev. Lett. **85**, 4450 (2000); P. Selles, L. Malegat and A.K. Kazansky, Phys. Rev. A **65**, 032711 (2002).
[10] P. Selles, L. Malegat, A.K. Kazansky, D.P. Seccombe, S.A. Collins, T.J. Reddish, and A. Huetz (unpublished).
[11] A.K. Kazansky, in *Proceedings of the 20th International Conference on the Physics of Electronic and Atomic Collisions, Vienna, Austria, 1997*, edited by F. Aymar and H.P. Winter (World Scientific, Singapore, 1998).
[12] A.K. Kazansky, J. Phys. B **31**, L579 (1998).
[13] C.W. McCurdy, D.A. Horner, and T.N. Rescigno, Phys. Rev. A **65**, 042714 (2002).
[14] A.K. Kazansky and V.N. Ostrovsky, J. Phys. B **27**, 447 (1994); A.K. Kazansky, V.N. Ostrovsky, and L.Yu. Sergeeva, *ibid.* **27**, 5197 (1994).
[15] C. Le Sech, J. Phys. B **30**, L47 (1997).
[16] S. Bashkin and J.O. Stoner, *Atomic Energy Levels and Grotrian Diagrams* (North-Holland, Amsterdam, 1975).
[17] W.V. Ignatowsky, Ann. Phys. **18**, 495 (1905).
[18] W.H. Press, S.A. Teulkovsky, W.T. Vettering, and B.P. Flannery, *Numerical Recipes in Fortran (The Art of Scientific Computing)* (Cambridge University Press, Cambridge, 1994).
[19] M.C. Witthoeft, M.S. Pindzola, and J. Colgan, Phys. Rev. A **67**, 032713 (2003).
[20] D.P. Seccombe, S.A. Collins, T.J. Reddish, P. Selles, L. Malegat, A.K. Kazansky, and A. Huetz, J. Phys. B **35**, 3767 (2002).