Impossibility of distinguishing between identical particles in quantum collision processes

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(Received 21 May 1998)

The very successful convergent close-coupling (CCC) method [I. Bray and D. V. Fursa, Phys. Rev. A **54**, 2991 (1996)] for calculating cross sections for the ionization of atoms by electron impact is examined for the case of hydrogen ionization. The theory is recast in an operator theoretic form, allowing explicit formulas to be given for the solutions of the truncated CCC equations. The limit to which these solutions tend as ever more terms are included in the truncation is shown to be the exact transition operator for the system. This analytical result is not consistent with the numerical CCC results, which yield amplitudes that violate the symmetrization principle of quantum mechanics and which depend for success on treating the electrons as classically, rather than quantally, indistinguishable particles. This new result is also inconsistent with the ''step function hypothesis'' [I. Bray, Phys. Rev. Lett. **78**, 4721 (1997)] that was introduced to restore the consistency of the CCC calculations with the symmetrization postulate. In addition, the CCC method does not include long-range effects of the Coulomb interaction that are expected to be important in certain kinematic configurations at low energies. Consequently, it is not justified yet to interpret the numerical successes of the CCC method as confirmation of a fundamental dynamical understanding of electron-induced atomic ionization.
[S1050-2947(99)03204-7]

PACS number(s): 34.80.Bm, 34.80.Dp, 03.65.Nk

Over the past five years Igor Bray and colleagues [1–8] have performed a remarkable series of calculations of physical observables for the ionization of hydrogen and helium, as well as other atoms, by electron impact. Most recently they have treated the double photoionization of helium [9]. Their approach, which they call the convergent close-coupling (CCC) method, introduces a finite expansion for the continuum wave function of the ionized electrons. Thus, instead of solving mathematically sound but multidimensional three-body equations (e.g., Faddeev equations), they solve a more tractable set of coupled integral equations in a single variable. Due to the finite expansion, the wave functions can be accurate only in a finite volume since the square integrable basis functions do not satisfy the proper scattering conditions at large radii. Nevertheless, the results of the numerical calculations are in good agreement with experimental data, aside from an overall normalization factor for certain differential cross sections.

Presumably due to the spectacular phenomenological success of the CCC method, little attention has been paid to the fact that the calculations produce ionization amplitudes that are very different from those of the standard quantum theory of such processes [10,11]. The purpose of this Comment is to study these differences between the standard theory and the CCC results with more mathematical precision than has previously been done.

Let us concentrate on the electron-hydrogen system, the simplest for which the theory of Bray *et al.* [1,3] can be formulated. Following Refs. [1,3], let us assume that the infinitely massive proton is held fixed at the origin and that the long-range effects of the Coulomb interaction can be ignored to the extent that the scattering formalism for short-range forces can be employed. In atomic units the Hamiltonian is expressed as

$$H=-\frac{1}{2}\Delta_{\mathbf{r}_1}-\frac{1}{2}\Delta_{\mathbf{r}_2}-\frac{1}{|\mathbf{r}_1|}-\frac{1}{|\mathbf{r}_2|}+\frac{1}{|\mathbf{r}_2-\mathbf{r}_1|}, \tag{1}$$

where $\mathbf{r}_1$ and $\mathbf{r}_2$ denote the electron position vectors. The exact spin-singlet ($S=0$) and spin-triplet ($S=1$) ionization amplitudes F_S are given in partial distorted-wave form by

$$F_S(\mathbf{p},\mathbf{q};\mathbf{k})=\langle \mathbf{p},\chi_{\mathbf{q}}^{(-)}|T_S(E+i0)|\mathbf{k},\phi\rangle, \tag{2}$$

$$T_S(z)=V[1+(-1)^S P_{12}]\{I+(z-H)^{-1}V\}, \tag{3}$$

where $V=-|\mathbf{r}_1|^{-1}+|\mathbf{r}_2-\mathbf{r}_1|^{-1}$, P_{12} is the permutation operator that interchanges $\mathbf{r}_1$ and $\mathbf{r}_2$, while I denotes the identity operator. In the initial state, $|\mathbf{k},\phi\rangle$, electron 1 has momentum $\mathbf{k}$ and electron 2 is bound to the proton (with bound state wave function ϕ and bound state energy ϵ). In the final state, $|\mathbf{p},\chi_{\mathbf{q}}^{(-)}\rangle$, electron 1 is described by a plane wave with momentum $\mathbf{p}$ and electron 2 by an eigenstate of the Hamiltonian $-(1/2)\Delta_{\mathbf{r}_2}-|\mathbf{r}_2|^{-1}$ that satisfies incoming wave scattering boundary conditions corresponding to asymptotic momentum $\mathbf{q}$. The amplitudes are to be evaluated on the energy shell, $E=\epsilon+|\mathbf{k}|^2/2=(|\mathbf{p}|^2+|\mathbf{q}|^2)/2$. It is well known that the following symmetry condition, required by the symmetrization postulate [12], is a consequence of the above definitions:

$$F_S(\mathbf{q},\mathbf{p};\mathbf{k})=(-1)^S F_S(\mathbf{p},\mathbf{q};\mathbf{k}). \tag{4}$$

The triple differential ionization cross section by electron impact is given by [11]

$$\frac{d\sigma}{d\Omega_{\mathbf{p}}\,d\Omega_{\mathbf{q}}\,d\varepsilon}=(2\pi)^4\frac{|\mathbf{p}||\mathbf{q}|}{4|\mathbf{k}|}\{3|F_1|^2+|F_0|^2\}, \tag{5}$$

where $\Omega_{\mathbf{p}}$ and $\Omega_{\mathbf{q}}$ denote the differential solid angle associated with the direction of $\mathbf{p}$ and $\mathbf{q}$, respectively, and $\varepsilon = |\mathbf{q}|^2/2$.

To this point, the theory has followed the standard treatment [10,11], except for the ignoring of long-range effects of the Coulomb interaction.

In the CCC method the approximate ionization amplitudes $\tilde{F}_S^{(N)}$ have the form

$$\tilde{F}_S^{(N)}(\mathbf{p},\mathbf{q};\mathbf{k}) = \langle \mathbf{p}, \chi_{\mathbf{q}}^{(-)} | \tilde{T}_S^{(N)}(E+i0) | \mathbf{k}, \phi \rangle, \tag{6}$$

where the transition operator $\tilde{T}_S^{(N)}(z)$ is the solution of an equation of the Lippmann-Schwinger type [3],

$$\tilde{T}_S^{(N)} = \tilde{V}_S^{(N)} + \tilde{V}_S^{(N)}(E+i0-[\tilde{h}_S^{(N)}])^{-1}\tilde{T}_S^{(N)}. \tag{7}$$

Here

$$\tilde{V}_S^{(N)} \equiv I^{(N)}\{V-(-1)^S P_{12}(E-H)\}I^{(N)}, \tag{8}$$

$$\tilde{h}_S^{(N)} \equiv I^{(N)}(H-V)I^{(N)}. \tag{9}$$

The operator $I^{(N)}$ has the form

$$I^{(N)} = I_1 \otimes \sum_{n=1}^{N} |\psi_n\rangle\langle\psi_n|, \tag{10}$$

where I_1 is the identity operator for the $\mathbf{r}_1$ variable and $\Sigma_{n=1}^{N}|\psi_n\rangle\langle\psi_n|$ is a projection operator for the $\mathbf{r}_2$ variable, $|\psi_n\rangle$ being orthonormal basis vectors. The CCC method consists of choosing suitable basis vectors and subsequently calculating the amplitudes $\tilde{F}_S^{(N)}$ by solving Eq. (7) for increasing values of N until the results appear to be insensitive to the increase of N.

To what limit $\tilde{F}_S^{(\infty)}$ do the amplitudes $\tilde{F}_S^{(N)}$ converge as $N\to\infty$? It is straightforward in our operator theoretic version of the CCC theory to show that

$$\tilde{T}_S^{(N)}(z) = \tilde{V}_S^{(N)} + \tilde{V}_S^{(N)}(z-\tilde{H}_S^{(N)})^{-1}\tilde{V}_S^{(N)}, \tag{11}$$

where $\tilde{H}_S^{(N)} \equiv \tilde{h}_S^{(N)} + \tilde{V}_S^{(N)}$. That is,

$$\tilde{H}_S^{(N)} = I^{(N)}\{H-(-1)^S P_{12}(E-H)\}I^{(N)}. \tag{12}$$

Since the basis vectors $|\psi_n\rangle$ are chosen to be complete, $\lim_{N\to\infty} I^{(N)} = I$ [[3], Eq. (6) and following, and [5]]. It follows that

$$\tilde{F}_S^{(\infty)}(\mathbf{p},\mathbf{q};\mathbf{k}) = \langle \mathbf{p}, \chi_{\mathbf{q}}^{(-)} | \tilde{T}_S^{(\infty)}(E+i0) | \mathbf{k}, \phi \rangle, \tag{13}$$

where

$$\tilde{T}_S^{(\infty)}(z) = \tilde{V}_S + \tilde{V}_S(z-\tilde{H}_S)^{-1}\tilde{V}_S, \tag{14}$$

$$\tilde{V}_S \equiv V-(-1)^S P_{12}(E-H), \tag{15}$$

$$\tilde{H}_S \equiv H-(-1)^S P_{12}(E-H). \tag{16}$$

Due to the on-shell equalities $(H-E)|\mathbf{k},\phi\rangle = V|\mathbf{k},\phi\rangle$ and $\langle \mathbf{p},\mathbf{q}|(H-E) = \langle \mathbf{p},\mathbf{q}|V$, and because $\tilde{H}_S$ and P_{12} commute, Eq. (13) can be rewritten as

$$\tilde{F}_S^{(\infty)}(\mathbf{p},\mathbf{q};\mathbf{k}) = \langle \mathbf{p}, \chi_{\mathbf{q}}^{(-)} | \hat{T}_S(E+i0) | \mathbf{k}, \phi \rangle, \tag{17}$$

$$\hat{T}_S(z) = V[I+(-1)^S P_{12}]\{I+2(z-\tilde{H}_S)^{-1}V\}. \tag{18}$$

Introducing the notation $A^{(S)} \equiv [I+(-1)^S P_{12}]/2$ facilitates the derivation of

$$(E+i\epsilon-\tilde{H}_S)^{-1} = \frac{1}{i\epsilon}(I-A^{(S)}) + \frac{1}{2}(i\epsilon/2+E-H)^{-1}A^{(S)}. \tag{19}$$

Substituting the above expressions into Eqs. (17) and (18) yields the final result

$$\lim_{N\to\infty} \tilde{F}_S^{(N)}(\mathbf{p},\mathbf{q};\mathbf{k}) \equiv \tilde{F}_S^{(\infty)}(\mathbf{p},\mathbf{q};\mathbf{k}) = F_S(\mathbf{p},\mathbf{q};\mathbf{k}). \tag{20}$$

Equation (20) clearly implies that the approximate CCC amplitudes $\tilde{F}_S^{(N)}$ should be accurate approximations to the exact amplitudes $\tilde{F}_S$ as $N\to\infty$. However [6], the calculated complex amplitudes obtained so far are in gross violation of Eq. (4), which expresses a fundamental property of the exact amplitudes F_S. This is true of both the amplitudes $\tilde{F}_S^{(N)}$ and their absolute values $|\tilde{F}_S^{(N)}|$. The numerical CCC amplitudes have not, therefore, converged to accurate approximations of the exact amplitudes.

There is evidence that the CCC calculations not only have not converged to the standard amplitudes, they have apparently converged to something else. It was noticed [5] that the CCC amplitudes for the Temkin-Poet model [13] of electron-hydrogen ionization appear to possess a step function property when $\varepsilon = E/2$, and it was conjectured that this behavior is quite general. If the Temkin-Poet CCC amplitudes are converging to the exact, consequently correctly symmetrized amplitudes, then the integrated cross section

$$\frac{d\sigma}{d\varepsilon} = \int d\Omega_{\mathbf{p}}\, d\Omega_{\mathbf{q}}\, \frac{d\sigma}{d\Omega_{\mathbf{p}}\, d\Omega_{\mathbf{q}}\, d\varepsilon} \tag{21}$$

would vanish for $\varepsilon > E/2$. This implies that the integrand, and hence the singlet and triplet amplitudes individually, are zero whenever $\varepsilon > E/2$. Since $\varepsilon = |\mathbf{q}|^2/2$, and because the amplitudes are either antisymmetric or symmetric functions of $\mathbf{p}$ and $\mathbf{q}$, it would follow that both amplitudes would have to vanish identically on the energy shell. Clearly, the exact amplitudes, which are (even in the Temkin-Poet model) symmetrized in accord with the symmetrization postulate, cannot exhibit the step function behavior observed in the CCC calculations. It follows that the numerical limits that the CCC amplitudes apparently approach are profoundly different from the exact, correctly symmetrized amplitudes expected on the basis of Eq. (20).

Bray and co-workers have recognized that there is a conflict between the exact theory and the CCC method [5]: ‘‘. . . the CCC theory of ionization is inconsistent with formal ionization theory due to the fact that the CCC ionization cross sections are obtained from an incoherent sum of two

pairs of coherently summed direct and exchange amplitudes, which is crucial for obtaining good agreement with experimental angular profiles . . . '' They have also justified their procedure as follows [6]: ''An important, but surprising, feature of our approach is that we have a clear distinction between the primary (scattered) and the ejected (target) electrons . . . . These two theoretically distinct processes cannot be distinguished experimentally due to the indistinguishability of the detected electrons. Therefore, when comparing theory with experiment, we form an incoherent sum of the cross sections for the two theoretically distinct processes . . . . Note that this has to be done regardless of whether exchange between the projectile and target electrons has, or has not, been included in the close-coupling calculation.''

In accord with the viewpoint stated above, Bray and colleagues [3] use for the triple differential cross section not Eq. (5) but

$$\frac{d\tilde{\sigma}^{(N)}}{d\Omega_{\mathbf{p}}\,d\Omega_{\mathbf{q}}\,d\varepsilon}=(2\pi)^4\frac{|\mathbf{p}||\mathbf{q}|}{4|\mathbf{k}|}\{3|\tilde{F}_1^{(N)}|^2+|\tilde{F}_0^{(N)}|^2\}+(2\pi)^4\frac{|\mathbf{p}||\mathbf{q}|}{4|\mathbf{k}|}\{3|\tilde{G}_1^{(N)}|^2+|\tilde{G}_0^{(N)}|^2\}, \tag{22}$$

where $\tilde{G}_S^{(N)}(\mathbf{p},\mathbf{q})\equiv\tilde{F}_S^{(N)}(\mathbf{q},\mathbf{p})$. They justify this by an argument [3] similar to the one used by Messiah [[12], Eq. (XIV.52)] in obtaining an analog of Eq. (22) as an intermediate step in deriving an analog of Eq. (5). Messiah points out that the argument leading to that intermediate point is classical in nature, so it does not contain the essence of what distinguishes the quantum treatment of identical particles from the classical one. The subsequent quantum development by Messiah (see also [10]), which leads from the analog of Eq. (22) to that of Eq. (5), is missing from the CCC method. Indeed, assuming again that Eq. (20) is correct, Eq. (22) implies

$$\lim_{N\to\infty}\frac{d\tilde{\sigma}^{(N)}}{d\Omega_{\mathbf{p}}\,d\Omega_{\mathbf{q}}\,d\varepsilon}=2\frac{d\sigma}{d\Omega_{\mathbf{p}}\,d\Omega_{\mathbf{q}}\,d\varepsilon}, \tag{23}$$

so that the differential cross sections in the CCC method are not normalized in accord with those of standard atomic ionization theory [10].

Thus, the CCC method is a mixture of classical and quantum ideas. On the one hand, the presence of the projection operator P_{12} in the basic amplitudes defined by Eqs. (6)–(12) has its roots in the quantum indistinguishability of electrons. On the other hand, the cross sections defined in Eq. (22) are justified by arguments that are classical rather than quantum mechanical, even in the limit $N\to\infty$. The CCC method is simply not consistent in the way it handles the identity (indistinguishability) of the electrons.

Recently, Bray [5] has introduced the ''step function'' hypothesis as a way to resolve the difference between the CCC method and the standard theory. Although the hypothesis was formulated in the relatively simple context of the Temkin-Poet model [13], in the present general context it is the assertion that

$$\lim_{N\to\infty}\tilde{F}_S^{(N)}(\mathbf{p},\mathbf{q};\mathbf{k})=\tilde{H}_S^{(\mathrm{CCC})}(\mathbf{p},\mathbf{q};\mathbf{k}), \tag{24}$$

where $\tilde{H}_S^{(\mathrm{CCC})}(\mathbf{p},\mathbf{q};\mathbf{k})$ is zero for $|\mathbf{p}|<|\mathbf{q}|$. This is clearly at odds with our earlier analytical result, Eq. (20).

While it is not stated directly by Bray, let us assume that the step function hypothesis implies that the converged CCC amplitude $F_S^{(\mathrm{CCC})}$ is to be defined as

$$F_S^{(\mathrm{CCC})}(\mathbf{p},\mathbf{q};\mathbf{k})=\tilde{H}_S^{(\mathrm{CCC})}(\mathbf{p},\mathbf{q};\mathbf{k})\,\theta(|\mathbf{p}|-|\mathbf{q}|)+(-1)^S\tilde{H}_S^{(\mathrm{CCC})}(\mathbf{q},\mathbf{p};\mathbf{k})\,\theta(|\mathbf{q}|-|\mathbf{p}|), \tag{25}$$

where $\theta(x)=1$ for $x>0$, 1/2 for $x=0$, and 0 for $x<0$. This new amplitude has the symmetries required by Eq. (4), except perhaps on the surface $|\mathbf{p}|=|\mathbf{q}|$, where it is true only if $\tilde{H}_S^{(\mathrm{CCC})}$ itself satisfies Eq. (4). Moreover, using $\tilde{H}_S^{(\mathrm{CCC})}$ to calculate the right side of Eq. (22) and $F_S^{(\mathrm{CCC})}$ to calculate the right side of Eq. (5) yields identical results. Thus, this procedure simultaneously produces amplitudes that have the correct symmetries under interchange of electrons and renders Eqs. (5) and (22) compatible.

While the step function hypothesis, as stated above, produces amplitudes that have desirable properties, it is *ad hoc* in that it has no discernible dynamical roots. If the CCC method does not yield the result, Eq. (20), expected from mathematical analysis, how does one know how the numerical limit $\tilde{H}^{(\mathrm{CCC})}$ in the region $|\mathbf{q}|<|\mathbf{p}|$ is related to the theory? The CCC answer so far has been that the calculated amplitudes yield cross sections that agree so well with experimental data that

$$F_S^{(\mathrm{CCC})}=F_S\,. \tag{26}$$

This answer is completely unsatisfactory precisely because it does not prove but *assumes* Eq. (26), making it impossible within the CCC method to decide whether or not the data are actually in disagreement with the dynamics represented by the Hamiltonian H.

The partly classical and partly improvised reasoning of the CCC formalism is made even more puzzling by the fact that the mathematical scattering theory for quantum systems containing identical particles is well developed. A general algebraic treatment of identical particles in quantum scattering processes was fully elaborated almost two decades ago for particles with short range [14,15] and with Coulomb [16] interactions. For approximate techniques, such as the DWBA method, the nontrivial question of the definition and treatment of reaction mechanisms for systems containing identical particles has also been worked out, including a number of important combinatorial results regarding practical problems as the relative normalization of the various coherent amplitudes [17,18].

Despite the above objections, however, one may argue that the shape of the numerical differential cross section is in such good agreement with experimental data, even though the absolute normalization is wrong, that there must be some morsel of truth in the empirical CCC process. The main issue is whether the CCC calculations have truly converged to the reaction amplitudes dictated by the fundamental Hamiltonian H. The arguments in favor of convergence are numerical in

nature. The argument against is based on the fact that, until the step function hypothesis is grafted onto the theory, the CCC amplitudes do not have the symmetries demanded by the symmetrization postulate and by Eq. (20). What is missing is an understanding, rooted in the fundamental dynamics, of why the CCC calculations produce the behavior of Eq. (24), rather than the theoretically expected behavior of Eq. (20), as well as why the crucial Eq. (26) should be true.

Let us now turn to the treatment of the long-range effects of the Coulomb interaction, in which the CCC method as applied is deficient in two ways. The various methods of including Coulomb effects have been reviewed by several authors [10,19,20]. One practical approach is to screen the Coulomb potentials and then let the screening radius approach infinity. Practical calculations for proton-deuteron scattering [21–23] have shown that surprisingly large screening radii are needed for converged results, which has adverse implications for the size of the numerical mesh of the calculation. In another formalism Jones, Madison, and Konovalov [24] studied various approximations to the three-body Coulomb wave function to calculate cross sections for electron-induced ionization of hydrogen. This study shows that the amplitudes are rather sensitive to the way in which the long-range Coulomb effects are approximated.

The first deficiency of the CCC method is that it includes the long-range Coulomb effects only in the asymptotic Coulomb wave function $\chi^{(-)}$. The complicated three-body asymptotic logarithmic phase structure that is known to be essential [10,24] is strikingly absent. Thus, the method must be considered as one in which the Coulomb interactions are screened. Unfortunately, there have been no calculations in which the implicit screening has been studied nor any explorations of the sensitivity to the cutoff radius, which the work of Jones *et al.* [24] suggests might be considerable.

Second, the CCC method also makes use of an expansion of a Coulomb scattering amplitude in terms of eigenfunctions of the angular momentum operators. It has been known for years [25,26] that such expansions have very bad convergence properties in the case of two-particle Coulomb scattering. There are no mathematical studies of convergence in analogous three-body problems, raising the possibility that the CCC expansion does not converge at all.

On the basis of the above considerations we can only conclude that the basic CCC results presented so far are incompatible with the symmetrization postulate and with known long-range effects of the Coulomb interaction. The step function hypothesis used to restore the CCC method to compatibility with the symmetrization postulate has no known dynamical justification. Consequently, the dynamical foundations of the CCC method as presented in the publications cited are flawed, and any dynamical interpretation of the numerical successes of the method is not yet soundly justified.

We thank many colleagues for several discussions, particularly I. Bray, I. E. McCarthy, and F. S. Levin. We are grateful for support from the Hungarian Scientific Research Foundation (OTKA Grant No. T014897) (Gy.B.) and the National Science Foundation (Grant Nos. Int-9222354 and PHY-9505615) (C.C.).

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