Finite-Hilbert-basis-set calculations for the angular distribution of ionized electrons produced in $p+H$ impact at 20 keV

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We present a different method of extracting the angular distribution of ejected electrons in an ion-atom collision from a two-centered finite Hilbert basis-set calculation. We obtain good agreement with experiment for a $p+H$ collision at 20 keV if we include an interference between the target centered and projectile centered amplitudes.

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I. INTRODUCTION

An energetic projectile proton in collision with a hydrogen atom is a much studied fundamental system. It provides an ideal well defined laboratory wherein we may learn how to understand a dynamic quantal collision [1–8]. Our concern here is direct ionization where the electron is ejected into a continuum state; ionization also occurs through charge transfer where the electron is captured onto a bound state of the projectile proton. The cited work notwithstanding, we report here a different way of extracting differential electron cross sections from finite Hilbert basis-set (FHBS) calculations [1]; this incorporates a quantal ionization process wherein the outgoing ejected electron diffracts past the target and projectile protons. This gives rise to an interference bringing theory and experiment [9] into agreement.

Full quantal calculations of differential cross sections have been previously reported [4–8] but just a small number of direct comparisons with experiment have been made. Perhaps one reason for this is the quite remarkable successes of the classical trajectory Monte Carlo (CTMC) algorithm [2] and the continuum distorted wave (CDW) approximation [3]; they reproduce the Rudd group data over a wide range of proton energies [9].

When compared to the CTMC and CDW approximate treatments, full quantal calculations are complicated to program and expensive in computer time at convergence. However, whereas the CTMC algorithm models the initial quantum state with an ensemble of electron starting conditions, the subsequent projectile-electron dynamics is treated classically. Thus the CTMC algorithm cannot reproduce a dynamic quantal interference. The CDW approximation is fully quantal. Ionization is treated as charge transfer to a projectile continuum state, but target and projectile interference in the final state is not treated directly.

With this in mind, if we re-examine the situation, we find an anomaly: a failure of the CTMC-CDW approaches to reproduce the electron angular distribution in the $p+H$ system at 20 keV [9]. This determines our choice of projectile energy for the full quantal FHBS calculation described here.

A difficulty with the FHBS method in producing differential cross sections is the fact that the ejected electron energy continuum is modeled by a discretized set of pseudostate eigenenergies. We must re-interpret the pseudostate contributions in terms of acceptably smooth momentum amplitudes; to these we can apply the necessary Galilean transformation to move between projectile and target frames. The main theoretical thrust of this work is to describe how this may be done.

II. FINITE HILBERT BASIS-SET CALCULATIONS

A successful method of describing the reaction of a target electron to the impact of a projectile ion, presumed to be following a classical path, $\mathbf{R}(t)=(\mathbf{B},\mathbf{Z})=(\mathbf{B},\mathbf{v}t)$, has been to expand the system wave function in a *finite Hilbert basis set*. FHBS calculations typically use a single centered expansion (SCE), which has just target centered bases, or, a two centered expansion (TCE), which has both projectile and target centered bases. A third alternative which we refer to informally as a one-and-half-centered expansion (OHCE) [10] is intermediate in its flexibility; it is two centered but limits the projectile basis to contain just bound states.

The FHBS system wave function at large target-projectile separation is written as

$$\begin{aligned}\psi_{nlm} = &\Sigma\chi_{T(n'l'm')}(\mathbf{r})e^{-i\epsilon_{n'l'}t}U_{T(n'l'm'),T(nlm)}(\infty,-\infty)\\ &+\Sigma e^{i\mathbf{v}.\mathbf{r}}\chi_{p(n'l'm')}(\mathbf{r}-\mathbf{R})e^{-i\epsilon_{n'l'}t-iv Z/2}\\ &\times U_{P(n'l'm'),T(nlm)}(\infty,-\infty).\end{aligned} \quad (1)$$

Here $U_{T(n'l'm'),T(nlm)}$ are the intratarget transition amplitudes, and $U_{P(n'l'm'),(Tnlm)}$ are the target-projectile transition amplitudes. The set $\chi_{T(n'l'm')}(\mathbf{r})=\chi_{T(n'l')}(r)Y_{l'm'}(\Omega)$ are the eigenstates obtained from diagonalizing the target Hamiltonian projected onto a target centered basis, whereby we obtain the discrete energy spectrum $\epsilon_{n'l'}$. The projectile states are similarly defined, but note in Eq. (1) the various translational factors due to the motion of the projectile.

The target Hamiltonian H_T has a kinetic energy term T_T and a Coulomb potential term V_T. It is the projection of T_T onto the Hilbert basis that leads to the discretization of the continuum, replacing it with a set of positive energy states; sometimes called pseudostates.

The projection of V_T leads to an effective finite range potential. Thus there are not an infinite number of Rydberg

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states in the FHBS model, just a finite number of negative energy states. It is an intrinsic assumption of the FHBS method that this approximation of cutting off the Coulomb tail is appropriately accurate; this is a hypothesis to be tested by a convergence study of the particular cross section to be calculated. We will use this finite range assumption below.

III. A METHOD OF LIMITED ACCURACY

To obtain ionization we could examine ψ_{nlm} in a region where $\mathbf{r}$, the electron coordinate, is far from both the target and projectile, and then project onto a pure momentum state, $|\mathbf{k}\rangle = e^{i\mathbf{k}\cdot\mathbf{r}}$. But the very nature of the FHBS method is that it is not accurate in describing the electron when it is far from either center; further the result is not independent of time as all positive energy amplitudes contribute to the projection onto the system wave function, Eq. (1). A similar difficulty with lattice based calculations has been described but is not fully resolved [7,8]. Methods for working directly in momentum space so as to avoid the difficulty have been developed [4,5]. However, none of these calculations successfully addresses this angular distribution.

In this work the key observation [6] is that the true target continuum radial wave function, $\chi_{T(l)}(E,r)$, when projected on to the basis, designated by $\mathbf{P}_T$, is accurately connected to the pseudostate when $E = k^2/2 = \epsilon_{nl}$ by

$$\mathbf{P}_T \chi_{T(l)}(\epsilon_{nl}, r) = N_{nl} \chi_{T(nl)}(r). \tag{2}$$

Here $\chi_{T(nl)}(r)$ is normalized to unity; the constant N_{nl} renormalizes it for the scattering boundary conditions.

The two identities,

$$e^{i\delta_l(k)} \sin\ \delta_l(k) = 2k \int dr r^2 (\epsilon_{nl} - H_T) j_l(kr) N_{nl} \chi_{T(nl)}(r) \tag{3a}$$

$$-e^{i\delta_l(k)} \cos\ \delta_l(k) = 2k \int dr r^2 (\epsilon_{nl} - H_T) j_{l-1}(kr) \times (1 - e^{-\beta r}) N_{nl} \chi_{T(nl)}(r), \tag{3b}$$

allow the determination of the complex N_{nl}. The identities follow from examining the differential equations satisfied by $j_l(kr), j_{l-1}(kr)(1-e^{-\beta r})$, and $\chi_{T(l)}(\epsilon_{nl}, r)$ and then using Eq. (2). We are assuming that the phase shift $\delta_l(k)$ is produced by a short-range potential.

Our Hilbert basis has been described elsewhere [6], for each l value about 100 exponential radial functions are used to diagonalize the target Hamiltonian. But just some 20 eigenstates are used in the collisional calculation itself. This involves about 800 coupled equations; 400 states on each center as we include s through h states. We have made extensive global studies of convergence for the p+H system in the intermediate energy range; generally we believe our worst, usually small, total cross sections to be reliable at the 10% level.

If $\chi_{T(l)}(\epsilon_{nl}, r)$ is well represented in the potential region then N_{nl} is independent of the choice of the parameter β! In Table I we vary β. One would be correct in inferring from this test that cross sections are going to be less reliable as the energy of the ejected electron is increased [6].

TABLE I. Values of N_{nl} for p pseudostates.

n	Energy (a.u.)	N_{nl} (β=0.5)	N_{nl} (β=1.0)
7	0.006	(−51.6, −41.1)	(−51.6, −41.1)
8	0.0298	(−9.02, 40.3)	(−9.01, 40.3)
9	0.0704	(−19.1, 19.2)	(−19.1, 19.2)
10	0.136	(−13.4, −10.8)	(−13.4, −10.8)
11	0.240	(11.3, −6.07)	(11.3, −6.06)
12	0.400	(−1.40, 8.91)	(−1.40, 8.90)
13	0.644	(−2.98, −5.54)	(−2.98, −5.54)
14	1.01	(3.80, 2.56)	(3.80, 2.56)
15	1.56	(−3.35, −0.721)	(−3.36, −0.702)
16	2.38	(2.59, −0.265)	(2.59, −0.290)
17	3.58	(−1.86, 0.663)	(−1.82, 0.711)
18	5.35	(1.31, −0.804)	(1.25, −0.833)
19	7.87	(−0.909, 0.722)	(−0.767, 0.741)

Returning to the problem at hand, using Eq. (2) to project onto an SCE ψ_{nlm} allows the connection of the differential partial wave amplitude for ionization to the pseudostate amplitude, to wit,

$$t_{T(l'm'),T(lm)}(\epsilon_{n'l'}) = N_{n'l'} U_{T(n'l'm'),T(nlm)}(\infty, -\infty). \tag{4}$$

Because of the orthogonality of the pseudostates only one state contributes and there is no time variation left after the projection.

The pseudostates for different partial waves generally speaking do not have matching sets of eigenenergies. So we cannot directly construct momentum amplitudes. We could in principle use a basis in which each set of pseudostates for different angular momentum values had the same energies. But this is unnecessary. To produce the differential cross sections it is just necessary to be able to interpolate on the smooth physical differential amplitudes, $t_{T(l'm'),T(lm)}(\epsilon_{n'l'})$, to a set of common energies. To be successful the packing density of the discrete pseudostates has to be sufficiently high, mandating the large basis sets we have used. We have previously reported on a successful application to the energy differential cross sections [6].

In Fig. 1, we demonstrate the limitation of this method in a first Born approximation for a pure Coulombic potential. At small angles when the cross section is large the FHBS method has errors up to 10%. At angles above 120° the method is quite poor. Recall that at 180° there is a $(-1)^l$ factor in any partial wave sum. The phases determined by Eq. (3) are simply not accurate enough to handle the cancelation. We conclude that the FHBS method as formulated here is unreliable in such circumstances, and should not be used at such large angles.

At 20 keV charge transfer is an important channel and the SCE method is inefficient. If an OHCE basis is used instead, the capture of the electron is directly described. And the

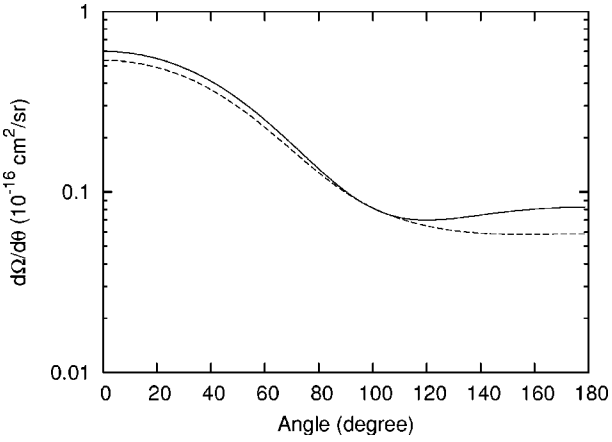

FIG. 1. Comparison of angular distribution of ionized electrons given by an exact first Born approximation (dotted line) and an FHBS Born calculation (solid line) in a $p+\mathrm{H}$ collision at 20 keV. In this result just *s* through *f* states were used for both cross sections.

same projection technique works if we limit the range of integration for the ionization to a region around the target; all projectile bound states have zero overlap with this region when the target and projectile are well separated. In Fig. 2 we plot the experimental data for the $p+\mathrm{H}$ system, and compare to CDW, CTMC, and OHCE calculations. All the theoretical methods fail to some extent.

We know that the free ionized electrons are steered by *both* the target and the projectile. To account for this feature efficiently it is necessary to use a TCE basis. Now the ionized electrons are described by two sets of pseudostates, target and projectile centered. There is no real difficulty with this *if we decide that the two contributions to ionization are incoherent*. We can easily find the projectile electron momentum distribution by the techniques described above; we then perform a Galilean transformation to obtain the distribution in the target frame which we add incoherently to the target contributions. The result is a considerable improvement in the fit to the data, Fig. 3.

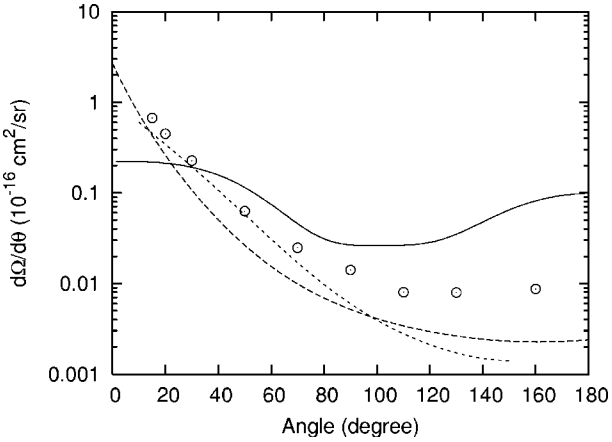

FIG. 2. Angular distribution of ionized electrons at 20-keV projectile energy $p+\mathrm{H}$ collision given by different theories: CDW (dots), CTMC (dotted line), and OHCE (solid line). The circled dots are experimental data.

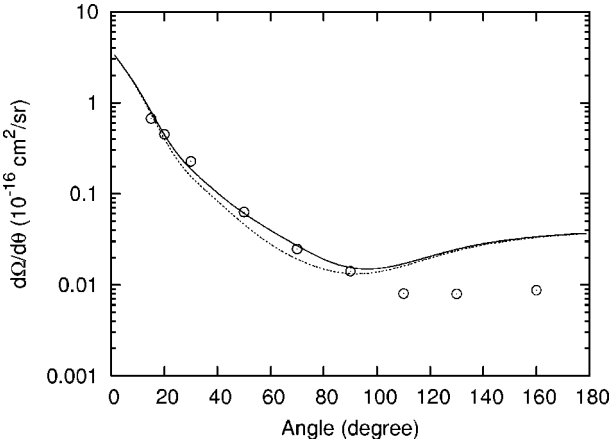

FIG. 3. Angular distribution of ionized electrons at 20-keV projectile energy $p+\mathrm{H}$ collision given by Two-Centered expansion model with interference (solid line) and without interference (dotted line). The circled dots are experimental data.

IV. COHERENCE

To include *coherently* target and projectile contributions when we use the TCE method is straightforward. We first chose to write a final ionized electron wave function as

$$\chi^{(-)}(\mathbf{k},\mathbf{r})e^{-ik^2t/2} = \chi_T^{(-)}(\mathbf{k},\mathbf{r})e^{-ik^2t/2} + e^{i\mathbf{v}\cdot\mathbf{r}+i\mathbf{k}\cdot\mathbf{B}}\chi_P^{(-)}(\mathbf{k}-\mathbf{v},\mathbf{r}-\mathbf{R}) \times e^{-i(\mathbf{k}-\mathbf{v})^2t/2-i\upsilon Z/2} - e^{i\mathbf{k}\cdot\mathbf{r}}e^{-ik^2t/2}. \quad (5)$$

We note that as $t\to\infty$, in the target region just the first term on the right of Eq. (5) survives, and in the projectile region just the second term survives. When we use this final-state wave function to project we get two contributions to the amplitude for ionization at a particular energy and angle. The first coming from the target pseudostates, the second from the projectile pseudostates.

We need $\chi_T^{(-)}(\mathbf{k},\mathbf{r})$ and $\chi_P^{(-)}(\mathbf{k}-\mathbf{v},\mathbf{r}-\mathbf{R})$ projected onto the target and projectile space, respectively. But before we can directly use Eq. (2) we have to have matching energies both for the target pseudostates and the projectile pseudostates. That is, we must have target pseudostates for which there are eigenenergies for all l' such that

$$k^2/2 = \epsilon_{n',l'} \quad (5a)$$

for some choice of n' and projectile pseudostates where there are eigenenergies for all l'' such that

$$(\mathbf{k}-\mathbf{v})^2/2 = \epsilon_{n'',l''} \quad (5b)$$

for some choice of n''.

As $\mathbf{k}$ is to have an arbitrary direction we cannot guarantee that both equations can be satisfied simultaneously for any particular Hilbert basis diagonalization. But if this happy situation were obtained then our projection would be independent of time! We solve this problem not by repeated diagonalizations with different bases but by once again interpolating the quantities $N_{n'l'}U_{T(n'l'm'),T(nlm)}(\infty,-\infty)$ and $N_{n''l''}U_{P(n''l''m''),T(nlm)}(\infty,-\infty)$ in pseudostate energies to the common desired value.

The final result is plotted in Fig. 3. With the interference term included, the experimental values are well reproduced by the FHBS method, until the large angle region is reached when the method fails as expected.

V. CONCLUSION

The interference which we have uncovered in this analysis points out the importance of electrons which are strongly influenced by the target or the projectile in the final exit channel. It represents an interesting and subtle aspect of the collision. Recall that the original Young's double slit experiment was in fact performed with two pinprick holes; in this case the geometry is similar. The target and projectile play the roles of moving objects from which the electron scatters. We anticipate that this interference will become richer in information as we go to higher-order differentiation. We have yet to demonstrate that theory is capable of following the data to these deeper levels.

[1] A. L. Ford and J. F. Reading, in *Atomic, Molecular & Optical Physics Handbook*, edited by G. W. F. Drake (AIP, Woodbury, NY, 1996), p. 571.
[2] R. E. Olson, in *Atomic, Molecular & Optical Physics Handbook* (Ref. [1]), p. 664.
[3] D. S. F. Crothers, F. B. M. Copeland, and J. T. Glass, in *Atomic, Molecular & Optical Physics Handbook* (Ref. [1]), p. 589.
[4] E. Y. Sidky and C. D. Lin, J. Phys. B **31**, 2949 (1998).
[5] B. Pons, Phys. Rev. Lett. **84**, 4569 (2000).
[6] J. Fu, M. J. Fitzpatrick, J. F. Reading, and R. Gayet, J. Phys. B **34**, 15 (2001).
[7] D. R. Schultz, C. O. Reinhold, P. S. Krstic, and M. R. Strayer, Phys. Rev. A **65**, 052722 (2002).
[8] M. Chassid and M. Horbatsch, Phys. Rev. A **66**, 012714 (2002).
[9] G. W. Kerby III, M. W. Gealy, Y. Y. Hsu, M. E. Rudd, D. R. Schultz, and C. O. Reinhold, Phys. Rev. A **51**, 2256 (1995).
[10] J. F. Reading, A. L. Ford, and R. L. Becker, J. Phys. B **14**, 1995 (1981).