Time-dependent, lattice approach for atomic collisions

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Several recent applications of the time-dependent, lattice approach to solving the Schrodinger equation for atomic collisions are described that illustrate progress in treating fundamental collisions. Specifically, we consider here the three-dimensional, lattice treatment of inelastic processes in proton-impact of atomic hydrogen and a four-dimensional, fully-correlated, planar model of antiproton-impact ionization of helium. One of the major goals of these studies has been to augment the basic toolkit available to describe fundamental, atomic-scale, few-body dynamics utilizing this direct, time-dependent approach.

1. The time-dependent, lattice approach

Significant insight regarding the dynamics of atomic collisions, as well as results with high accuracy may be obtained by solving the time-dependent Schrodinger equation iteratively on a multidimensional lattice using a variety of discrete variable representation, finite element, or spectral techniques. These methods seek to add a new approach to the standard ways in which atomic collisions are treated by avoiding certain approximations and difficulties. Examples of these include the need for somewhat arbitrary electron translation factors in molecular orbital close coupling calculations at intermediate collision energies, the assumption of weak interactions in perturbation theories, small discretized continua in atomic or molecular orbital close coupling bases, and multi-center basis set pathologies, such as over-completeness, in some atomic orbital close coupling approaches. In particular, techniques and approaches used in other fields of physics and engineering like aerodynamics, fluid mechanics, and hydrodynamics that have been very successful and that have driven computational advances concerning similar problems can provide a fertile ground from which to learn.

Further motivation comes from the fact that in atomic physics, it has become possible to calculate and to measure certain quantities with high precision. For example, atomic structure calculations and experimental spectroscopy together are precise enough to test the very fine scale effects of quantum electrodynamics. From a theoretical point of view, such an advance has hinged on several characteristics. Such atomic scale systems are described completely by a differential equation of rather simple structure, the interactions are of a simple form, and the well-developed techniques of eigensolution via basis function expansion and perturbation theory are generally applicable. However, for atomic dynamical problems such as collisions, even for systems with only one or two electrons, such a high degree of precision in describing observables has seldom been achieved for strongly interaction systems. Inherent complications that limit accuracy include the

necessity to describe thoroughly the two-center continuum and to represent processes driven through channels involving states on both centers or involving the interaction of electrons.

Recently it has been our goal to develop robust approaches that can overcome many of the difficulties and limitations associated with these methods by solving the time-dependent Schrodinger equation (TDSE) as directly as possible on a numerical lattice, taking advantage of contemporary techniques of computational science. In this approach, the electronic wavefunction and the action of operators are represented discretely on a grid of points using finite differences, finite elements, basis-splines, Fourier or pseudospectral representation in Galerkin or collocation approaches with uniform, non-uniform, or adaptive meshing, for example. Then, evolution of the wavefunction may be followed through time propagation using a variety of schemes such as direct Taylor or Chebyshev expansion of the formal exponential propagator or split-operator methods. Finally, various auxiliary procedures such as partial lattice eigensolution, projection and filtering methods, wall masking and absorption, and multigridding can be used to aid in extracting information about the results of the collision.

Though one-dimensional solutions of the Schrodinger equation have been a cornerstone of atomic collision calculations for many years, full three-dimensional treatments had to await the development of computers with sufficient speed and memory. By the mid-1990's such calculations became readily feasible. Early applications of this approach included treatment of ionization in collisions of antiprotons with atomic hydrogen [1,2] and He^+ [3] yielding significant insight into the behavior of these processes at low collision energy. Here, we describe treatments of several aspects of the very fundamental collision system H^+ + H and our first explorations of two-electron cases using the lattice TDSE (LTDSE) approach in a four-dimensional model of antiproton-impact of He.

2. Proton-impact of atomic hydrogen

Surveys of theoretical and experimental data for inelastic transitions as recently as 1990 showed order of magnitude differences and wide variations of behaviors for even the most fundamental collision system, H^+ + H. Clearly of great importance is a full and accurate understanding of this system since it is the least complex ion-atom collision and because it serves as a prototype for a wide range of collisions in which an unstructured ion interacts with an atom with primarily only one active electron. Utilizing the LTDSE approach, we have recently studied the principal inelastic channels in H^+ + H collisions with the goals of elucidating physical mechanisms not clearly understood through application of other methods and producing results as free as possible from bias owing to the choice of theoretical approach.

Specifically, for a given impact energy and a given impact parameter, the time evolution of the lattice, electronic wavefunction, initially in the H(1s) state, was propagated through the H^+ + H collision typically with hundreds to thousands of time steps. The overlaps of lattice eigenstates of H with the time-evolved wavefunction were computed periodically to judge the point at which reasonable convergence of the probabilities for excitation had

been obtained. Outgoing flux was absorbed at the walls through use of a complex potential with non-zero extent only near the walls, determined through empirical means to prevent reflections or other spurious effects. This procedure was then repeated for other impact parameters until a full picture of the probabilities was obtained so that integration over impact parameter accurately yielded the corresponding cross sections.

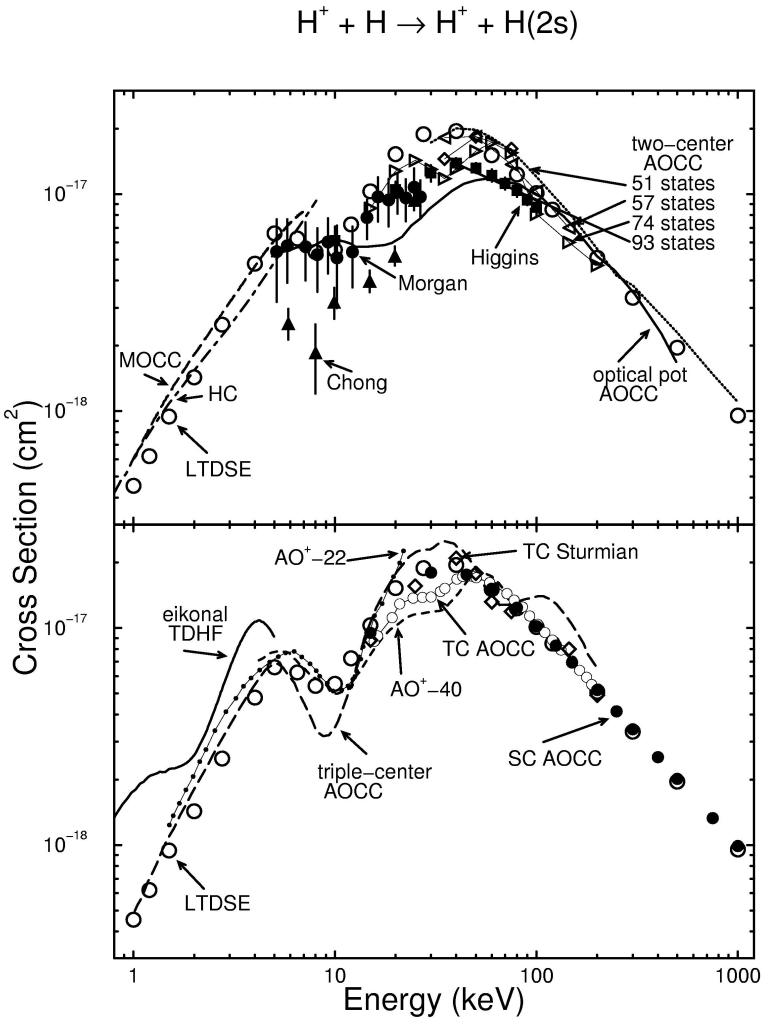

Figure 1. Comparison of the LTDSE [4] results for H(2s) excitation in proton-hydrogen collisions with a wide variety of other theoretical and with experimental results.

In comparison, excitation of H by proton impact has been treated by a wide variety of theoretical approaches and by a few key experiments. However, the results display significant disagreements for collision energies below the regime in which perturbation theory is applicable. Utilizing lattices of 135^3 and 225^3 points supporting complex exponential basis functions (Fourier collocation) we have treated excitation of H to the n=2 and 3 manifolds by proton impact at 25 energies between 1 and 1000 keV [4]. As

many as about 20 impact parameters were considered for each energy, and the time-evolution of the electronic wavefunction computed in order to extract a converged excitation probability after each time propagation and a converged cross section after integration over impact parameter. Excellent agreement was found with the best methods applicable at either high energy or low energy. Moreover, the LTDSE results spanned accurately the complete energy range without conceptual, mathematical, or numerical limitations to a sub-regime. This is illustrated in Figure 1 where the LTDSE results are compared with a large sampling of the diverse theoretical and experimental data for excitation of the 2s level. Many of the relatively recent results displayed there show large departures in magnitude and oscillatory structures from the LTDSE results for low- to intermediate-energies. Thus, the LTDSE results provide a kind of recommended, unbiased result for this fundamental collision system and inelastic channel.

We have also utilized LTDSE approaches, based on both finite-difference and Fourier collocation schemes, to treat charge transfer in H^+ + H collisions [5,6]. In the previous work on excitation, the H atom, and thus the electronic wavefunction, was initially at rest at the center of the lattice, the projectile impinged upon it, and we examined the excited density that remained associated with the target. In contrast, to study charge transfer we reversed the situation and placed the proton at the center of the lattice, allowed the H atom to impinge upon it, and then examined the excited density transferred to it after the collision. By considering a range of collision energies and impact parameters, in this way we were able to extract the n,l-dependent charge transfer total cross section. Very good agreement with the most reliable theoretical and experimental results was obtained, illustrated in part in Figure 2.

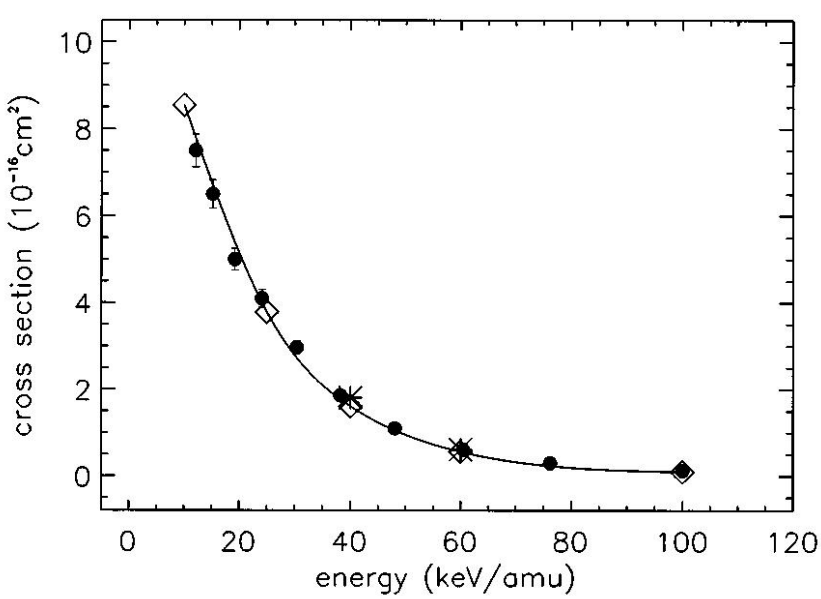

Figure 2. Comparison of the LTDSE results for charge transfer in proton-hydrogen collisions [6] with experimental measurements. Data displayed are from our finite differences (diamonds) and Fourier collocation (stars) methods and from experiments (circles).

With knowledge of both the state-selective excitation and charge transfer probabilities from these works, we were able to calculate [6] the ionization cross section by using unitarity. The result of this approach is shown in Figure 3 compared with the well-regarded experimental measurements of the Queen's University group. After careful analysis of the convergence of our results, we were not able to resolve the 10 to 35% difference between the LTDSE results and the experimental measurements near the peak of the cross section. Owing to the fundamental importance of the ionization cross section for H^+ + H, this remaining discrepancy should be resolved by further theoretical and experimental work. Very recent calculations by Sidky and Lin [7] agree well with our results.

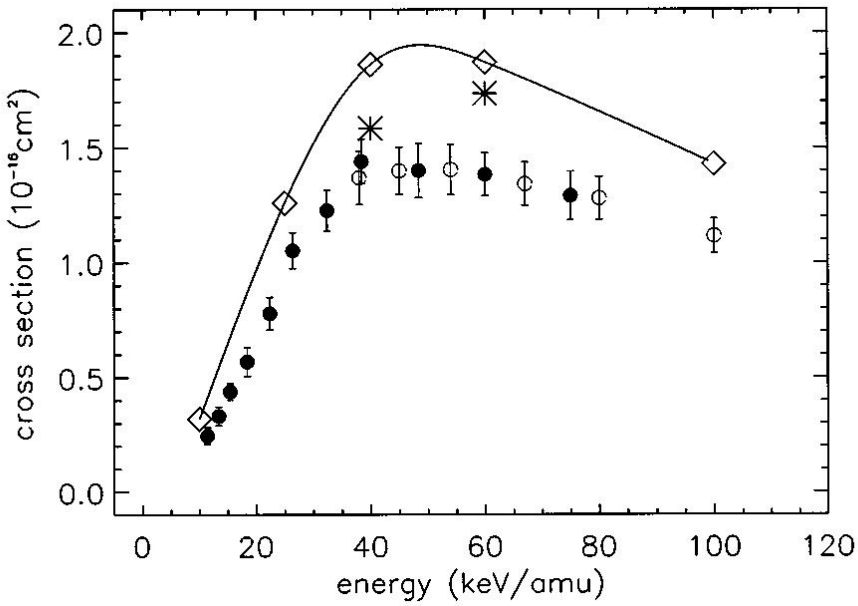

Figure 3. The same as in Figure 2 except for ionization in proton-hydrogen collisions.

A more strenuous test of theoretical methods is description of the spectrum of ejected electrons produced in ionization. Further impetus for such consideration is the result of momentum imaging experiments such as those utilizing COld Target Recoil Ion Momentum Spectroscopy (COLTRIMS) [8-10]. Those experiments have identified "sharp structures" in the electronic momentum distribution "that vary strongly with impact parameter and projectile velocity" [8] that are located near the saddle-point region, where the projectile and ionized target fields balance to create a saddle in the potential experienced by the ejected electrons. Interest has especially stemmed from the theoretical observation in the 1980's that a preponderance of the electrons emitted in slow collisions (e.g. for velocities less than about 1 a.u.) are more closely associated with the saddle region than with either the projectile or target ion [11]. The COLTRIMS measurements prompted a few theoretical models seeking to explain their results that turned out to yield at least partially contradictory conclusions. Thus, we have applied the LTDSE approach in order to try to resolve these contradictions [12].

In particular, we utilized a grid that forms a rectangular prism rather than a cube so as to accommodate a symmetric configuration of the target and projectile at the final point at which the electronic wavefunction was examined. Specifically, a grid of 135×135×270 points supported the Fourier functions was used, which covered dimensions of -26 to 26 a.u. in the x- and y-directions and -26 to 78 a.u. in the z-direction. The projectile impinged from the left and was propagated up to a final internuclear separation of 52 a.u. At that point the bound target and projectile states were projected out so that the remaining probability density represents the continuum.

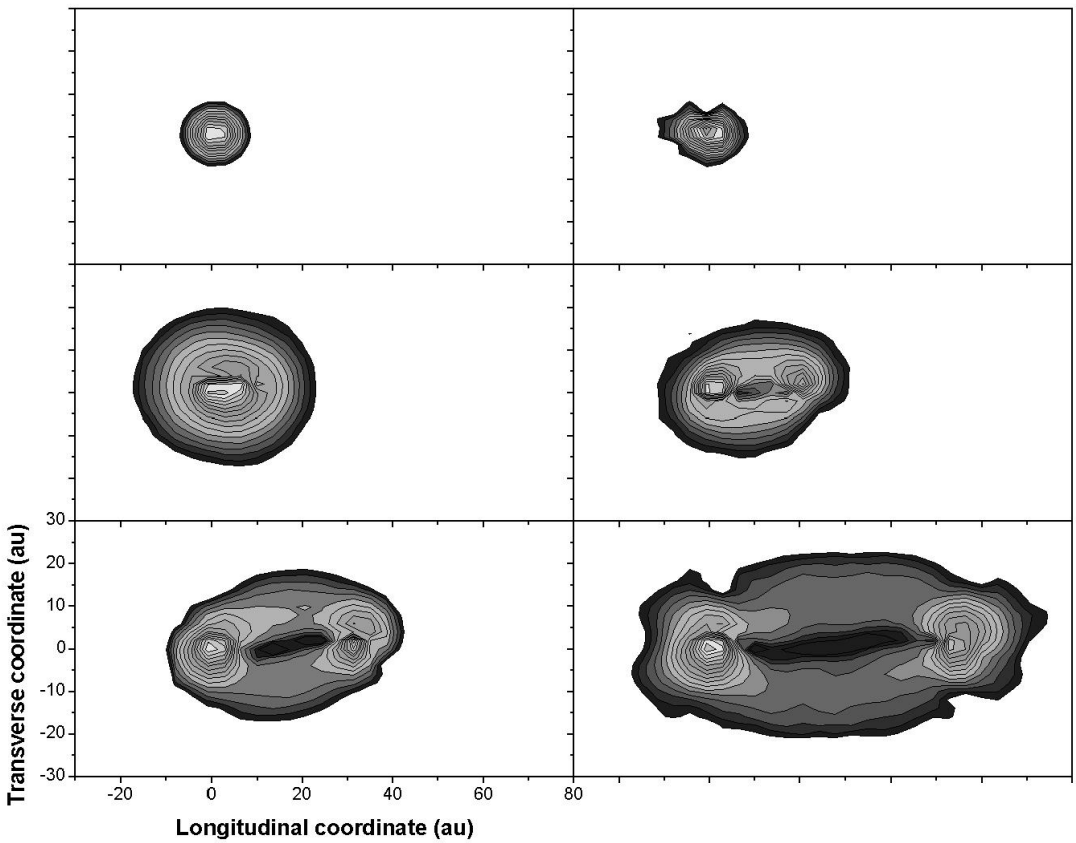

Figure 4. The time evolution of the electronic probability density in the collision plane for a 5 keV proton colliding with atomic hydrogen with an impact parameter of 0.77 a.u.

For the case of 5 keV proton impact, Figure 4 depicts the time evolution of the wavefunction at several time slices (or equivalently, at several projectile longitudinal coordinates) in the collision plane. One may readily note the bipolar character of the ejected flux for this collision energy and impact parameter. From the final wavefunction, the momentum distribution of the ejected electronic density may be extracted. Figure 5 shows the result after projecting out the target and projectile bound states and Fourier transforming the remainder to obtain the momentum distribution for the time slice in the last frame of Figure 4. We find that the asymmetry of the resulting momentum distribution, with respect to the projectile direction (z) in the collision plane converges

rather quickly at first, but reaches it asymptotic value only very slowly as the projectile and target ions separate.

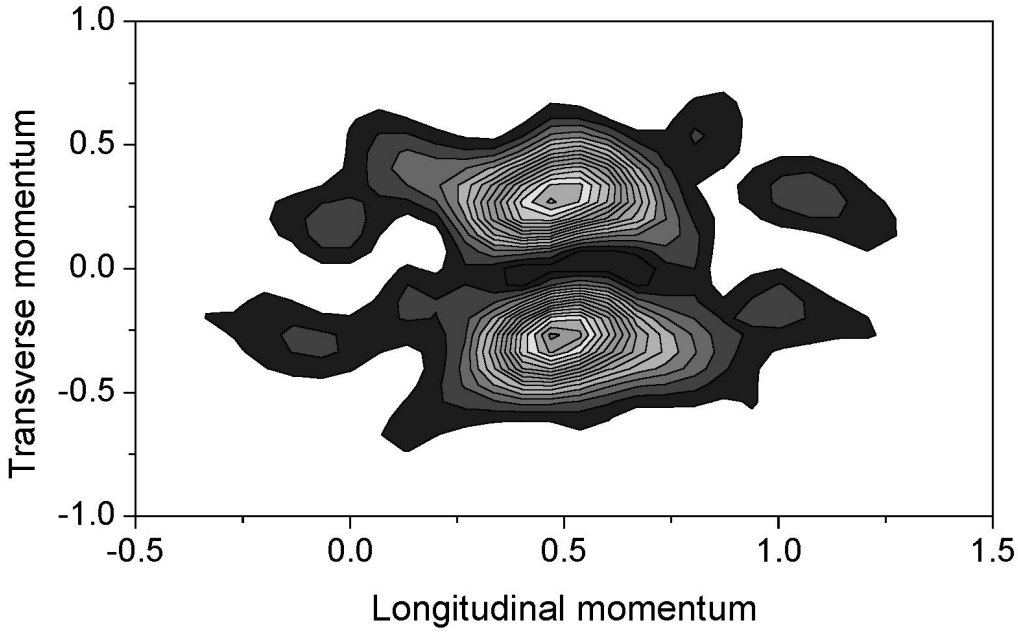

Figure 5. The continuum probability density in momentum space (in units of the projectile velocity) for the 5 keV proton-hydrogen collision depicted in the final frame of Figure 4. In these units, a longitudinal momentum of 0 corresponds to the target, 1 to the projectile, and 0.5 to the saddle point.

Our analysis of such computed momentum distributions as a function of collision energy between 1 and 25 keV supports the general picture proposed by Macek and Ovchinnikov [13] which predicted that structures in the ejected electron spectra should be expected to occur at low collision velocities because a small number of magnetic substates in the continuum are usually populated. More generally, the oscillatory behavior of the asymmetry of the spectrum provides clear evidence of coherences among magnetic substates and the changing relative transition amplitudes into these substates.

3. Fully-correlated, planar model of antiproton-helium collisions

The advent of atomic physics collision experiments using slow antiprotons produced at CERN was a significant milestone for both technical achievement and the opening up of experimental use of sign-conjugated projectiles (i.e. the antiproton has the same mass as the proton, but opposite charge, allowing comparison of collision behaviors when only a single projectile attribute is changed). A pioneering experimental measurement of the total cross section for ionization of hydrogen by antiproton impact [14] presented a challenge to theoretical understanding of the observed low energy behavior. Using the LTDSE approach and other methods [1] we were able to elucidate the proper low-energy behavior. In addition, measurements for He targets [15,16] displayed a behavior at low

energy that was not readily reproduced or supported theoretically. As a complement to theoretical work using the Forced Impulse Method (FIM) [17], the Multielectron Hidden Crossing (MEHC) method [18], and Atomic Orbital Close Coupling (CC) [19], we have sought to treat this two-electron, two-center problem using the LTDSE approach [20].

Since it is not yet feasible to represent well the full six-dimensional (6D) space that would be required to treat antiproton + He in LTDSE, we have employed a four-dimensional (4D, two-dimensions for each electron) model in which we can adjust a small, "softcore" parameter which flattens the Coulomb potential near the origin. This allows mimicking of the full 6D eigenenergy curves and thus a simulation of the full 6D dynamics in a smaller, more tractable space. We first tested this simulation procedure by mimicking antiproton + H collisions in a two-dimensional space. We were able to readily obtain eigenenergy curves for the ground and several excited states which had the same qualitative behavior as those obtained in three dimensions. Full time-dependent propagations yielded total ionization cross sections that followed the behavior of the three dimensional results obtained previously very closely apart from a constant multiplicative factor (see [20] for a more detailed description of the procedure and its results).

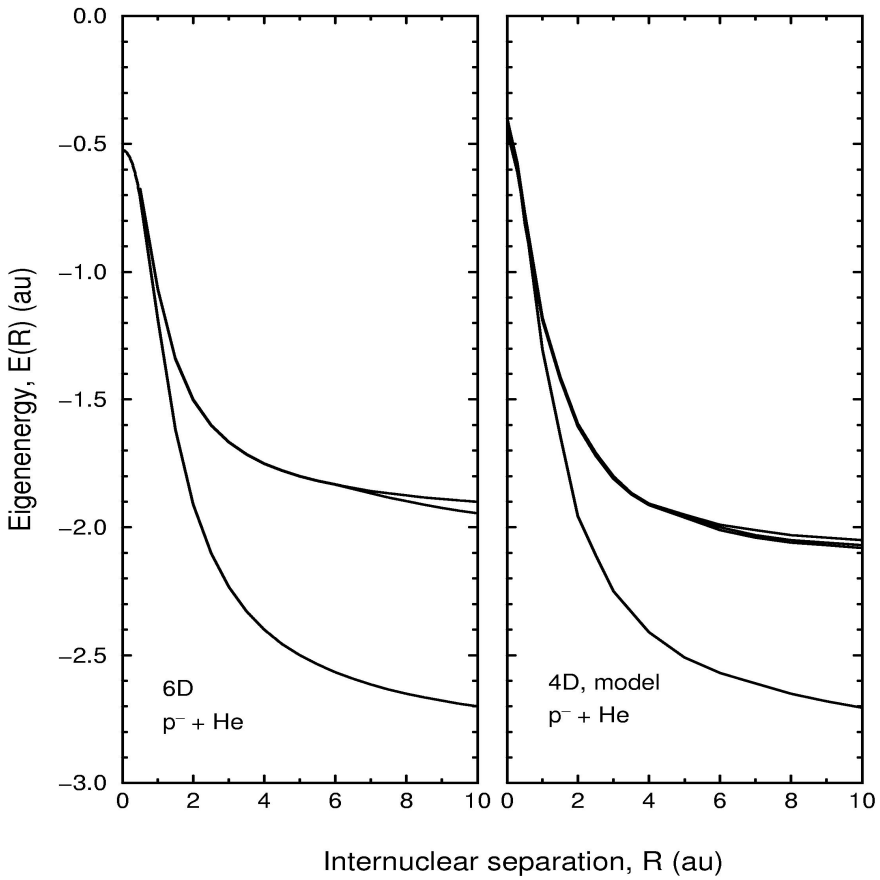

Figure 6. Comparison of the 4D, LTDSE model with the full 6D antiproton-He eigenenergies, E(R), as a function of internuclear separation, R, for the few lowest states.

The 4D eigenenergy curves obtained for the antiproton-He system as a function of internuclear separation are displayed in Figure 6 along with those from an accurate 6D quantum structure calculation. They agree reasonably well. To illustrate the evolution of the wavefunction during the collision, several time slices of the probability density are displayed in Figure 7 for 1 keV antiproton-impact. Similar to what was observed previously for antiproton + H, the early time evolution after the close approach of the antiproton to the target is an almost symmetric, s-wave outgoing distribution for impact parameters of about 1 a.u. or less.

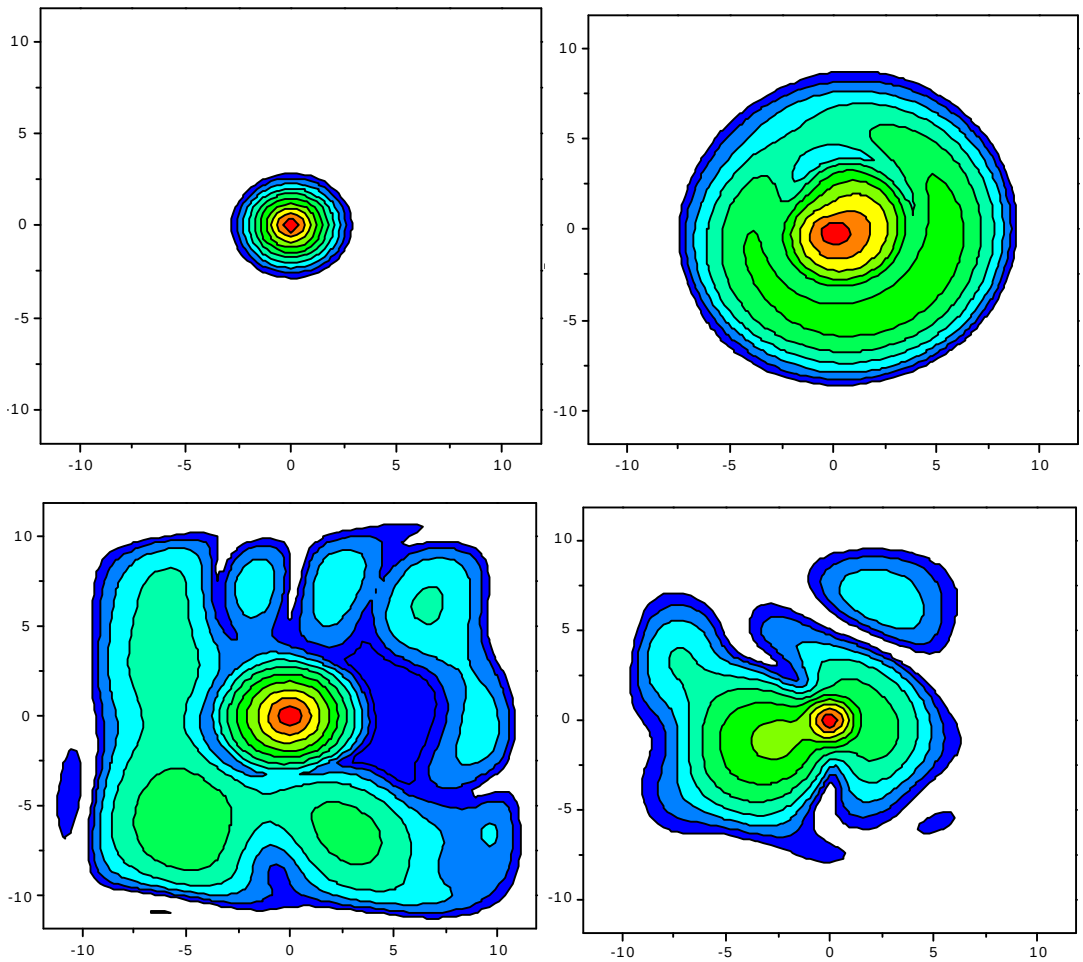

Figure 7. The electronic probability density as a function of one of the electrons' x,y coordinates in the 4D LTDSE model of antiproton-He collisions at 1 keV and an impact parameter of 0.5 a.u. for four times during the time propagation.

In Figure 8 the cross sections obtained for our 75^4 point grid employing Fourier collocation is compared with the experimental measurements and the FIM, MEHC, and CC results. Good agreement of the model results with these advanced theories was obtained, supporting their departure from the low-energy behavior of the measured cross section.

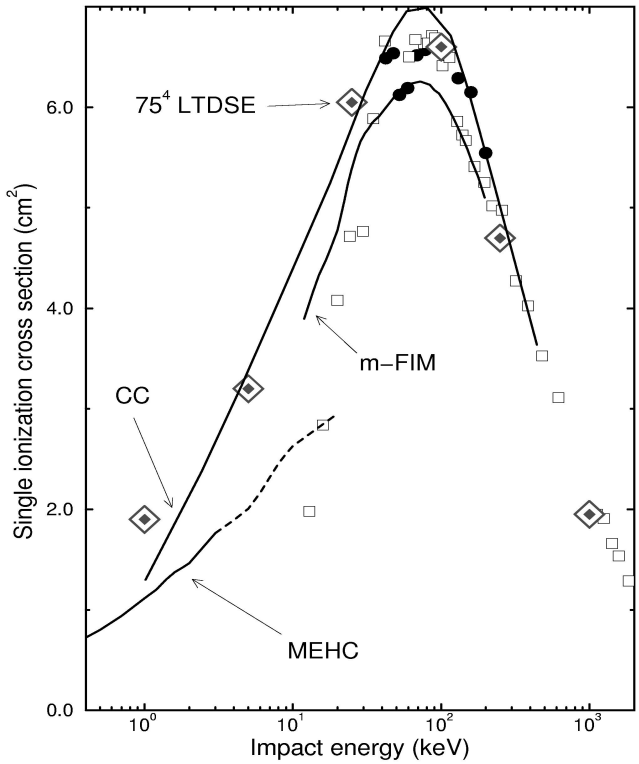

Figure 8. The total cross section for single ionization of helium by antiproton impact. Shown are pioneering experiments made by the CERN-Aarhus collaboration and various theoretical results as described in the text, compared to those of our 4D LTDSE approach [20].

4. Summary

We have treated a number of fundamental one- and two-electron collision systems utilizing a direct, numerical approach to solving the time-dependent Schrodinger equation. We have found solutions that are very accurate, bridging between and ranging over results of the best theoretical methods that are limited in the scope of their applicability. In addition, significant physical insight has been obtained aiding in the elucidation of dynamical effects in these fundamental systems.

Acknowledgments

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