Theoretical studies of interactions of atoms, molecules, and surfaces: Dynamics of few-body systems

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Program Scope

This research program will explore the three- and four-body problem under a variety of circumstances ranging from bound states to collisions, both with and without an external field. A major component of this program is to develop the novel theoretical tools — both analytical and numerical — required for this effort. The ultimate goal is to understand the dynamics of these strongly coupled systems in quantum mechanical terms.

Charge exchange in α+H_2^+ collisions

Recent progress My graduate student, Shu-chun (Amy) Cheng, has recently been working on solving the time-dependent Schrödinger equation for electron capture from H_2^+ by an ion. The calculations were motivated by an experiment carried out by C.L. Cocke's group in our lab. The essential physics is that when the electron is captured from H_2^+, the molecule will dissociate. Amy is propagating the Schrödinger equation in three-dimensions for the electronic wave function. The projectile is assumed to move along a straight line trajectory, and the molecular nuclei are assumed stationary. At the collision energies relevant to the experiments, both assumptions are quite good.

To help her understand the propagation methods, Amy started working on a one-dimensional model of the system. In the model, the molecule is in line with the trajectory of the incoming ion. The approaching ion comes within some minimum distance of the molecular center of mass, then recedes along its entrance direction. We used this model to test both the propagation code and a masking function that damps all of the electron wave function save that on the molecular ion. Since we are only interested in the total electron capture, this approach is both sufficient and efficient. Amy has obtained the capture probability in this model as a function of the distance of closest approach and presented the results at a poster at DAMOP.

Future plans Amy is in the process of developing the three-dimensional code, and no problems are expected. Once the total electron capture probability is obtained for this case, the dissociation probability is known since they are the same. The angular and energy distributions of the molecular nuclei are then known as well. We can thus obtain a quantity that is directly comparable to experiment. We plan to begin generating the data soon. It will take some time, though, since the capture probability depends on five parameters: the impact parameter b, the orientation of the molecule θ and ϕ, the distance between the nuclei R, and the velocity of the incident ion v. In the end, we will integrate over b, ϕ, and R, weighting the integrals with the appropriate nuclear wave function. As long as only one electron is active, there is, in principle, no difficulty to include a polyatomic target, or even a molecular projectile. These cases will, of course, require careful modelling to reduce them to one active electron, but they could be quite interesting to study.

Ionization in ion-atom collisions

Recent progress We have worked on applying the scaled coordinate method to the time-dependent Schrödinger equation for ionization in ion-atom collisions. The advantage of the scaling method is that it analytically removes much of the known behavior of the ionized wave function, leaving only a smooth, stationary envelope to propagate. In particular, it alleviates the need for the absorbing boundaries normally used in such calculations.

The primary effort on this project came from my postdoc, Wei Guo. He arrived in August 2001 and worked to apply the scaling method to a one-dimensional model of the collision. Using this model gave us the opportunity to explore the scaling method for ion-atom collisions in an efficient manner since the one-dimensional model already contains all of the elements peculiar to the scaling method. Wei eventually got the code working and began to generate data for the one-dimensional problem, but was unable to complete the project. Wei left at the end of May, 2002; a replacement, Vladimir Roudnev, has been hired and is scheduled to arrive in October 2002.

Future plans While the numerical aspects of this method require care, there appear to be no fundamental difficulties in extending the scaling to the three-dimensional ion-atom collision problem. My plans for the near future, however, do not include this project since other efforts appear more timely.

Protonium formation in $\bar{p}$+H(1s) collisions

Recent progress The low-energy, less than roughly 10 eV, collision of $\bar{p}$ with hydrogen is one of the most asymmetric systems one can consider. The difficulty lies in the fact that $\bar{p}$ and p can form bound states with a ground state energy of approximately –459 a.u. By comparison, the energy in the incident channel is –0.5 a.u. The formation of protonium, Pn or $\bar{p}p$, occurs when the proton is captured by the antiproton, ionizing the electron in the process. Based on simple energy arguments, the most likely Pn states produced are in the n=30 manifold. The extreme change in length and energy scales — going from truly quantum mechanical in the initial state to nearly classical in the final state — makes the problem extremely difficult to treat theoretically. Not surprisingly, the experiment is also difficult to do. Nevertheless, the ASACUSA collaboration at CERN predicts that they will make preliminary measurements of Pn formation within the next year. Unfortunately for us, the target will likely be hydrogen molecules rather than atoms since the Pn formation cross section is predicted to be larger from classical calculations.

It is tempting to apply the Born-Oppenheimer approximation to this problem, and many people have. The Born-Oppenheimer curves, however, do not allow for the formation of Pn; the united atom limit is a free electron. In collaboration with Hossein Sadeghpour at ITAMP, I have instead used the adiabatic hyperspherical approach to generate potentials that do indeed correlate to the $\bar{p}$+H(1s) limit as well as all of the Pn($n\ell$)+e^- channels. Unfortunately, about 450 potential curves are required to cover the energy range of interest. This multitude of channels and the strong coupling between them is at the heart of the problems with asymmetric systems. To gain some insight into the physics, we have artificially reduced the masses of p and $\bar{p}$ and carried out the scattering calculations with these simpler systems. By examining several values of the mass, we could identify the important channels. This work has been submitted for publication and has so far resulted in two invited talks: one at an ITAMP workshop and the other at an ECT* workshop in Trento, Italy.

Future plans I want to combine the physical insight gained in our scaled-mass calculations with a diabatic representation in order to reduce the number of channels in the full-mass problem. While we have recently completed the manuscript describing our split diabatic approach, another

diabatic approach will likely be needed. The split diabatic representation requires one to identify all of the curve crossings by hand — an unreasonable task with 450 channels. We will thus seek alternative diabatization schemes that can be applied more automatically. I anticipate that any method successful for this system can be profitably applied to other collision systems, allowing quantum mechanical calculations where none are now possible.

Low-energy Ps+Ps collisions

Recent progress Roman Krems, a very capable Predoctoral Fellow at ITAMP who is soon to be a postdoc there, recently spent two weeks at Kansas State to begin implementing my formulation of an adiabatic hyperspherical approach for the low-energy collision of two Ps (e^+e^-) "atoms". The only four-body adiabatic hyperspherical treatments that have been done to date are for three electron atoms (see C.D. Lin's report). With an infinitely heavy nucleus, this system has a natural center about which to define approximately good angular momentum quantum numbers. In fact, four electrons can been treated in the adiabatic hyperspherical approach for this same reason. The equal mass problem that we are considering, however, does not have such a natural center, making the necessary expansion over partial waves more slowly converging.

Because the adiabatic equation for the four-body problem in the center-of-mass (CM) frame requires eight coordinates, we must expand the wave function using partial waves. There is one spherical harmonic associated with each of the three relative position vectors in the CM frame, accounting for six coordinates. The other two coordinates are our hyperangles. In principle, then, coupled two-dimensional partial differential equations must be solved. This route very quickly becomes too computationally demanding. Instead, we are diagonalizing the adiabatic equation for a fixed set of $\{\ell_1, \ell_2, \ell_3\}$, then coupling these together to obtain the final result. Even though the {0,0,0} curve by itself recovers more than 90% of the asymptotic Ps+Ps energy, we are finding that the last few percent require many partial waves.

Future plans The code seems to be nearly complete, but we are still struggling to achieve convergence in the partial wave expansion. Roman will continue to work on this problem as a postdoc, trying to determine whether our current difficulties are due to lingering bugs in the program or to actual physics. Successful completion of this project will be an exciting accomplishment since few other methods could similarly treat equal mass four-body systems for both bound and collisional states.

Publications

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