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

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1 Program Scope

This research program, begun in the last year, is intended to explore the few-body problem in a variety of circumstances including bound states, collisions, and in external fields. A major component of this program is to develop novel theoretical tools required for this effort. In the last year, special emphasis has been placed on developing a general method for treating time-dependent wave functions in scaled coordinates. Applications of this new approach and plans for other projects are also addressed.

2 Intense laser-atom interactions

2.1 Background

High harmonic generation and above threshold ionization are two of the most familiar phenomena that have emerged from studies of interactions of intense, ultrafast lasers with atoms. Both processes are inherently nonperturbative and involve one or more electrons in the continuum. As such, they are difficult to treat theoretically even after many years of study.

2.2 Recent progress

We (Sidky and Esry) have developed a representation for the time-dependent Schrödinger equation that simplifies the problem considerably. The new representation, a combination of a coordinate and a wave function transformation, removes nearly all of the effects of kinetic energy from the wave function. This "scaling" representation frees us from the necessity of absorbing boundaries, allowing essentially boundary-free propagation of the wave function. It is then only necessary to propagate the relatively smooth envelope of the continuum wavepacket. In collaboration with C.D. Lin, the scaling method has since been applied to the model problem of an intense laser interacting with a one-dimensional atom (Zhao, Esry, and Lin). It was found there that the scaling does indeed reduce the computational burden by roughly an order of magnitude on average. To simplify the wave function even further, the wave function was transformed to the acceleration gauge to account for the effects of the oscillating laser field. We were thus able to propagate the wave function to very large times, allowing us to see directly in the wave function localized continuum wavepackets corresponding to different numbers of photons absorbed, *i.e.* above threshold ionization.

2.3 Future plans

Based upon our experience with the one-dimensional problem, we have hopes that the scaling method can make two- or three-dimensional time-dependent calculations a more routine proposi-

tion than they are currently. The generalization of the method and code is reasonably straightforward, and we fully expect the same computational benefits realized in one dimension. The scaling method shows promise for essentially any time-dependent problem involving the continuum and many applications are possible. One application that we intend to explore in the near future is described in the next section.

3 Ion impact ionization of atoms

3.1 Background

After many years of investigation, the theory for ion-atom collisions is well developed and can handle bound-bound transitions quite reliably. Bound-free processes, on the other hand, are much less reliably treated, but progress is being made — largely with time-dependent methods. One problem that continues to plague current theoretical treatments is the inability to integrate the Schrödinger equation to large internuclear separations. Such long-time propagation is necessary to make the final state analysis less ambiguous, especially for low-energy collisions. The final momentum distribution of the ionized electron can then be extracted and compared with both experimental results and existing theory. It is hoped that with a cleaner asymptotic analysis we can help resolve the discrepancies remaining between them. The need to understand such a prototype process is clear, but is more fully motivated in the report by C.D. Lin.

3.2 Recent progress

This project is only in its beginning stages, but the time-dependent equations to be solved within the scaling method have been derived. We are considering the standard problem of one electron with straightline trajectories for the nuclei.

3.3 Future plans

This project is first on the list for my new postdoc. He will be charged with adapting existing three-dimensional time-dependent codes to solve the equations already obtained. The scaling method has the potential to remove the limitations on integration times, but remains to be explored for ion-atom collisions. There appear to be few obstacles to a successful implementation, but even within the scaling approach we are considering different schemes whose relative efficiency must be evaluated.

4 Helium trimer

4.1 Background

The helium trimer is an intriguing system to study because the helium dimer is so weakly bound. In fact, quantum chemistry texts often state that it is unbound based upon simple bond number analysis. It is bound, but barely so — and only for $^4He_2$; its single bound state has a binding energy of only about 1 mK. Such a weakly bound dimer implies very interesting physics for the trimer in the form of Efimov states. Efimov states are of keen interest to nuclear physicists as they are closely related to the halo states many neutron-rich nuclei possess. Our own previous calculations as well as those of several other groups confirmed that $^4He_3$ has two bound $J = 0$ states and that the excited state has Efimov characteristics. The question naturally arises whether there are any bound states with $J > 0$.

4.2 Recent progress

We have carried out calculations for the rotationally excited states of the helium trimer within the adiabatic hyperspherical approximation (Lee, Esry, Gou, and Lin). Simple estimates and calculations by two other groups had suggested that bound states might exist, but we found only completely repulsive adiabatic potential curves. Thus, no rotationally excited bound states are possible for the helium trimer. One especially interesting aspect of this work was the role of identical particle permutation symmetry. Since a helium atom is a boson, the spatial wave function of the trimer must be completely symmetric under permutations. This requirement forces the adiabatic potentials to be far more repulsive than they would be had the helium atoms been distinguishable.

4.3 Future plans

We currently have no plans to pursue similar calculations in the future, but will revisit the problem if more interesting physics presents itself.

5 Diabatization schemes

5.1 Background

The vast majority of problems of current interest in atomic physics are described by a Schrödinger equation that is not separable in any coordinate system. Adiabatic approximations, such as the Born-Oppenheimer approximation, are convenient and powerful tools for dealing with such equations. Unfortunately, at the very points where the physics gets interesting — near the avoided crossings — the numerical treatment gets difficult. Theoretically, then, a representation that does not have avoided crossings would be desirable. Such representations are generically called diabatic, but it is difficult in general to generate a diabatic representation that includes as much physics as compactly as the adiabatic representation.

5.2 Recent progress

We have recently finished developing one possible diabatization scheme (Esry and Sadeghpour). Standard, strict diabatic transformations are well known and require the nonadiabatic derivative coupling. We have dubbed our new representation the "split diabatic representation" as it is based upon the same equation as the strict diabatic transformation but with only part of the nonadiabatic coupling included. No approximations are made, however, since the remaining coupling is retained in its adiabatic form. The new representation can thus be considered a mixed representation since it is neither purely diabatic nor purely adiabatic, but the avoided crossings are eliminated. The split diabatic representation also avoids one of the problems of the strict diabatic representation, namely the unphysical behavior of the diabatic potentials at large distances. It turns out that because of long range nonadiabatic coupling — present for both the Born-Oppenheimer and the adiabatic hyperspherical representations — the strict diabatic potential curves oscillate sinusoidally at large distances. The long range portion of the nonadiabatic coupling need not be included in the definition of the split diabatic representation, though, so the asymptotic potential curves coincide precisely with the physically appealing adiabatic potentials.

5.3 Future plans

We intend to explore applications of the split diabatic representation. In particular, one advantage of a diabatic representation is the ability to selectively include or exclude individual channels. The advantage is computational since only physically important channels need be included. While the split diabatic representation produces physical potential curves and has the potential to reduce the number of channels, it is not suitable for automatic application to a large number of channels because someone must decide what part of the coupling to use in the definition. We will thus also seek diabatic representations that are suitable for large problems.

6 Publications

- "Boundary-free propagation with the time-dependent Schrödinger equation," E.Y. Sidky and B.D. Esry, Phys. Rev. Lett. **85**, 5086 (2000).
- "Boundary-free scaling calculation of the time-dependent Schrodinger equation for laser-atom interactions," Z.X. Zhao, B.D. Esry and C.D. Lin, Phys. Rev. A (submitted) (2001).
- "Helium trimer has no bound rotational excited states," T.G. Lee, B.D. Esry, B.C. Gou, and C. D. Lin, J. Phys. B **34**, L203 (2001).
- "Split diabatic representation," B.D. Esry and H.R. Sadeghpour, Phys. Rev. A (submitted) (2001).