Origin, evolution, and imaging of vortices in proton-hydrogen collisions

D.R. Schultz¹, J.H. Macek², J.B. Sternberg², S.Yu. Ovchinnikov²,³, and T-G. Lee⁴

¹Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, ²Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37496, ³Ioffe Physical Technical Institute, St. Petersburg, Russia, ⁴Department of Physics and Astronomy, Auburn University, Auburn, AL 36849

schultzd@ornl.gov

Abstract. Using a novel computational approach, we have elucidated the origin of unexpected vortices in the electronic wavefunction during ion-atom collisions. It is shown how they could be observed in experiments and how they play a new and wide ranging role in angular momentum transfer and other atomic processes.

1. Introduction

Beginning in the mid 1990’s, we developed a direct method of solving the time-dependent Schrödinger equation (TDSE) on a numerical lattice in three or more dimensions to treat ion-atom collisions, the so-called lattice, time-dependent Schrödinger equation (LTDSE) approach [1]. This was enabled by the dramatic increases in computer speed and memory that have persisted to this day and by new computational methods. We were motivated to take this direct approach in order to circumvent shortcomings of other methods that stem from either computational or formal limitations. Since that time others have developed methods (e.g., [2]) sharing many of the same characteristics, being free from these limitations and which are demonstrably convergent with improvements such as smaller lattice spacing and time steps. Such work has led to a new range of computational methods to treat ion-, electron-, and photon-impact of atomic and molecular systems.

More recently, we applied the LTDSE method to treat not only excited states of either the projectile (ion or atom) and target (ion or atom) but also the spectrum of electrons ejected in the collision which are promoted into the two-center continuum [3], motivated by the pioneering measurements of such spectra via the “reaction microscope” or COLTRIMS method [4-8]. That work led to new insight into the formation of the σ-π distribution effects [9] observed experimentally [4] as well as their variation with impact energy. However, limits on the size of the numerical lattice prohibited reaching a satisfactorily asymptotic internuclear distance using LTDSE or similar methods [10]. To go beyond this limitation, we have very recently developed a new approach that we term the regularized LTDSE (RLTDSE) method [11], described below, in order to propagate continuum wavefunctions to essentially macroscopic distances. This has allowed us to propagate the solution to distances where we are assured that the electronic probability distribution is comparable to that which would be observed experimentally. In so doing, we have unexpectedly observed vortices in the electronic
wavefunction [12] and have begun to demonstrate their quite far reaching consequences in explaining such phenomena as angular momentum transfer in ion-atom collisions. We have also found that certain of these vortices, being born in the collision, surprisingly survive to asymptotic distances in the electronic continuum and may thus be observable in future experiments.

In fact, we note that vortices are usually associated with systems containing large numbers of particles. However, of particular interest in atomic, molecular, and optical physics are those formed within atomic-scale wavefunctions and observed in macroscopic systems such as superfluids and quantum condensates. In particular, we have shown how vortices appear in proton-hydrogen collisions, rotate around the nuclei, and interact, thereby transferring angular momentum from nuclear to electronic motions. To illustrate these phenomena here, in the following sections we briefly summarize the RLTDSE approach, its application showing vortex formation and evolution, and provide a look forward towards other atomic-scale systems that display similar phenomena. Atomic units (a.u.) are used throughout the paper unless otherwise indicated.

2. The RLTDSE method

Determining the evolution of continuum electrons in the presence of time-dependent fields as they move from atomic to macroscopic distances has been of great interest since the advent of quantum mechanics because observations are made at distances where they can be detected by devices such as particle detectors. We describe a corresponding theoretical method made possible by combining two theoretical approaches, namely, the LTDSE method and the removal of essential singularities of continuum wave packets described implicitly in the dynamical Sturmian theory [13]. This is necessary because standard numerical techniques that are successfully applied for the TDSE of bound states meet with difficulties in treating the continuum components due to highly oscillatory phase factors $\exp[ir^2/2t]$ as is evident from the free-particle propagator (where $r$ is an electronic coordinate and $t$ is time). Because these phase factors have essential singularities for large distances, both fundamental and numerical difficulties arise. This is important for physical processes where electron motion at large times, in contrast to just near the parent nuclei, plays a decisive role. For example, in ion-atom collisions, classical trajectory Monte Carlo calculations [14] show that cusps in the ejected electron spectra do not emerge until the target and projectile species are separated by more than 5,000 a.u., many times larger than the initial atomic dimensions.

To begin with, the electronic wavefunction is written in the form [11]

$$\Psi(r,t) = \chi(q, \theta) \Phi(q, \theta)$$  \hspace{1cm} (1)

where $q = (rv \sin \theta)/\Omega$ and $t = (\Omega \cot \theta)/\sqrt{v}$, $\theta$ varies from 0 to $\pi$, and $v$ and $\Omega$ are arbitrary parameters in general but $v$ will represent a physical velocity and $\Omega$ will be a function of the internuclear separation, $R$, and the scaled time. The factor

$$\chi(q, \theta) = (i \sin \theta / \Omega)^{3/2} \exp[-i(\Omega q^2 \cot \theta)/2]$$  \hspace{1cm} (2)

is highly oscillatory and has an essential singularity at $1/q=0$. The regularized wavefunction $\Phi(q, \theta)$, however, has slow-varying continuum components and satisfies a regularized TDSE

$$\left[-\frac{1}{2} \nabla_q^2 + \frac{1}{2} \Omega^2 q^2 + V(q, \theta)\right] \Phi(q, \theta) = i\Omega \partial \Phi(q, \theta) / \partial \theta$$  \hspace{1cm} (3)

where $V(q, \theta)$ is a possibly time-dependent potential. Here, where the projectile is a bare ion of charge $Z_P$ (=1 for proton-impact) and the target ion has the charge $Z_T$ (=1 for hydrogen), the potential is given by

$$V(q, \theta) = \Omega / (v \sin \theta) \left[ -Z_P / |q+O(\theta)/2| - Z_T / |q-O(\theta)/2| \right]$$  \hspace{1cm} (4)
where \( \mathbf{Q}(\theta) \) can represent any trajectory, here a straightline describing the nuclear motion, i.e., \( \mathbf{Q}(\theta) = \cos \theta \mathbf{v}' + \mathbf{v} \sin \theta \mathbf{b}' / \Omega \), where \( \mathbf{v} \) is now the relative velocity, \( \mathbf{b} \) is the impact parameter, and \( \mathbf{v}' \) and \( \mathbf{b}' \) are corresponding unit vectors. The initial regularized wavefunction is

\[
\Phi_0(q, \theta_0) = \left( \Psi_0(v^{-1} q \Omega \csc \theta_0 - v^{-2} \Omega \cot \theta_0) / \chi(q, \theta_0) \right)
\]

(5)

where \( \Psi_0(r, t) \) is the full wavefunction at the initial time \( t_0 = -(\Omega \cot \theta_0) / v^2 \). At asymptotically large times as \( \theta \to \pi \), the continuum components of \( \Phi(q, \theta) \) are independent of \( \Omega \) and directly yield the ejected electron momentum distributions, i.e.,

\[
A(k) = \lim_{\theta \to \pi} \Phi(q, \theta)
\]

(6)

where \( q \to k / v \) and \( k \) is the electron wave vector. We solve Eq. (3) using the LTDSE approach in which the equation is discretized on a large, three-dimensional lattice of points and the wavefunction propagated in time using fast Fourier transforms in the split-operator approach.

In Ref. [11] we first applied the RLTDSE method to slow collisions of protons with atomic hydrogen and were able to obtain a stable and low noise solution for final internuclear separations as large as 52,000 a.u. We found that the continuum electronic momentum distribution changed significantly between 52 and 5,200 a.u., where the largest distance we had previously been able to reach without the expanding coordinates used in RLTDSE was 52 a.u. [3]. The change was small but noticeable in going from 5,200 to 52,000 a.u. As described in Ref. [11], this allowed us to trace from their origins to these large distances many features of the ejected electron spectrum including the target and projectile cusps and the S-type and T-type transition mechanism electron distributions.

3. Vortices in proton-hydrogen collisions

Enabled by these RLTDSE calculations, we examined the time-dependence of the electronic wavefunction (for proton-hydrogen collisions at 5 keV) throughout the evolution from near the collision to large internuclear separations, and noticed, unexpectedly, that deep minima existed when the nuclei were close, some of which persisted to the largest final distances considered [12]. After also examining the electronic probability current, we confirmed that these deep minima were vortices (see Figures 1 and 2), and were in fact quantized.

Vortices first form at \( R \sim 5 \) a.u. (Figure 1) and are associated with a sudden change of topology of the classically allowed region of electron motion. Initially, when the protons are far apart, the topology of the classically allowed region is that of isolated atoms. Then during the collision, the potential barrier separating the two attractive Coulomb wells drops below the atomic eigenenergy, allowing the electron to move around the two atomic nuclei so that the classically allowed region has a molecular topology. This sudden change induces 1s\( \sigma_g \rightarrow 3d\sigma_g \) transitions that give rise to a nodal surface in the time-dependent electronic wavefunction. The nodal structures are precursors to vortex formation as discussed initially by Schrödinger [15] and Dirac [16] applied later by others [17].

At slightly later times, when \( R = 4.8 \) a.u., a vortex separates from the nodal surface. This vortex is not associated with any center of force (i.e., neither proton). Here the electronic density locally circulates around a vortex center, indicated by the arrows pointing along the electronic probability current in Figure 1. This current circulation is a physical consequence of transferring of angular momentum from relative to internal motion of the atoms. The electron density rotates about the center of the vortex and, in turn, the center of the tornado-like vortex itself rotates around the quasimolecule and transfers angular momentum to the electron.
Our computations also reveal another new topological feature, namely ring vortices, analogous to classical smoke rings, which appear and disappear during the collision. Ring vortices have been discussed by Bialynicki-Birula and collaborators [18] in the context of coherent superpositions of atomic states. The present ring vortices are associated with the superposition of united atom states. These states have a similar topology to that of atomic states where both protons are screened by a centrifugal potential and the electron perceives the protons as a single (“united”) Coulomb center. The united atom topology only exists for a short time; therefore the ring vortices also exist for the same period of time. These states are populated by recapture from continuum adiabatic states formed on the incoming phase of the collision. The distances where the ring vortices are formed coincide with distances \( R = \ell (\ell + 1)/2 \) where transitions from continuum to bound states occur. These distances are determined by the united atom orbital quantum numbers with \( \ell > 0 \).

The expanding ring vortices entangle with the rotating tornado vortex and are later absorbed by the tornado. At a post-collision distance of \( R = 50 \) a.u., the electronic wavefunction has a \( \pi \)-type distribution of \( \mathrm{H}_2^+ \) with a nodal line running between the protons. When the final atomic bound states are formed, several tornado vortices appear and some of them become pinned to atomic centers. These pinned vortices are associated with bound states that have non-zero projections of angular momentum on an axis perpendicular to the scattering plane, implying that angular momentum is ultimately transferred to circulation about one or the other proton, that is, the atomic states have non-zero orientation parameters. Such parameters have been measured for several atomic processes and provide indirect evidence of vortex motion in simple atomic systems. Further, the typical picture of collisional orientation, as summarized in propensity rules, envisions that electron distributions circulate about atomic centers without prior circulation about free vortex centers. In contrast, our computations show that the transfer of angular momentum sets up local circulation about the center of free vortices and initiates rotational motion seen as orientation of atomic states.

Even though free vortex motion is consistent with quantum mechanics, no examples of free vortices in actual physical systems have been observed. Vortices in many-particle systems are observed by direct imaging, as in the recent case of BECs, where they are visually observed by light scattering or absorption [19-21]. This is possible because the condensate is a macroscopic many-particle system.
(Manifestations of vortices in ultracold matter wave propagation through waveguides have also interestingly been suggested [22].) No similar imaging has been proposed for atomic-scale, single-particle quantum wavefunctions, yet direct observation of vortices in such systems would greatly extend our understanding of quantized vortices. The evolution of the electronic wavefunction and vortices was followed to internuclear distances of $R = 10^6$ a.u. (50 microns). Thus, the momentum distribution images the continuum wavefunction, with free vortices appearing as holes, seen in Figure 2. Observation of these structures would unambiguously confirm the importance of vortices in quantum mechanics.

![Figure 2. A logarithmic contour plot of the electron momentum distribution for $b=0.9$ a.u. in 5 keV proton-hydrogen collisions at a final internuclear separation of 52,000 a.u. The two peaks are the projectile and target continuum cusps, the connecting density is the result of the well known $\sigma-\pi$ interference, and the holes in the density near the nuclei correspond to vortices created in the near collision that survive to asymptotic distances and could be observed experimentally.](image)

4. **Vortices in other atomic systems**

We have also recently found vortices when atoms are subject to short electric field pulses. In fact, by varying the length of these pulses (on the order of several to a hundred femtoseconds), and by changing the direction of the electric field, the resulting vortices can be created in a controlled fashion, and tracked along well defined trajectories. These numerical experiments could lead to ways in which to manipulate the vortices and their interactions (such as mergers) somewhat in analogy with the way Rydberg wavepackets are manipulated using electric field pulses (see the recent review article by Dunning and collaborators [23]). We have also found that vortices exist for electron impact of atoms, and manifest themselves as deep minima in the triply differential ionization cross section. This follows, for example, calculations that revealed these deep minima (e.g., [24]) but could not associate them with any other known behavior such as interferences. Thus, it is quite likely that the formation of vortices is ubiquitous in atomic scale interactions and that in many situations they have macroscopically observable consequences, either directly as deep minima, or in other properties such as angular momentum characteristics not previously recognized as originating through vortices.

5. **Conclusions**
We note that quantized vortices are a common phenomena in BECs and in superfuilds such as liquid helium. Through recent work we see that these phenomena also occur in single-particle quantum systems described by a linear partial differential equation, namely, the Schrödinger equation with no non-linear terms as are present in the Gross-Pitaevskii equation describing BECs. Our calculations serve as a model for the dynamics of vortices in atomic systems, and show that such vortices remain when systems evolve to macroscopic distances where they can be imaged.

Finally, the present work has revealed that vortex motion is a heretofore unrecognized characteristic of atomic-scale dynamics and that vortices may play roles in correlated dynamics that have been difficult to interpret with conventional pictures. It shows that vortices in single-electron wavefunctions can be directly observed experimentally, elucidates the origin of vortices in dynamic systems, and shows that they play an important role in angular momentum transfer. Thus, the work summarized here opens up a path towards a new perspective on a very broad range of atomic-scale physics.

Acknowledgements

This research was sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy, under Contract No. DE-AC05-96-OR22464 with Oak Ridge National Laboratory, which is managed by UT-Battelle, LLC, and by Grant No. DE-FG02-02ER15283 to the University of Tennessee.

References

[1] D.R. Schultz, M.R. Strayer, and J.C. Wells, Phys. Rev. Lett. 82, 3976 (1999).
[2] H.J. Lüde, S. Henne, T. Kirchner, and R.M. Dreizler, J. Phys. B 29, 4423 (1996); O.J. Kroneisen, H.J. Lüde, T. Kirchner, and R.M. Dreizler, J. Phys. A 32, 2141 (1999).
[3] D.R. Schultz, C.O. Reinhold, P.S. Krstic, and M.R. Strayer, Phys. Rev. A 65, 052722 (2002).
[4] R. Dörner, H. Kehrmichle, M. H. Prior, C. L. Cocke, J. A. Gary, R. E. Olson, V. Mergel, J. Ullrich, and H. Schmidt-Böcking, Phys. Rev. Lett. 77, 4520 (1996).
[5] M. A. Abdallah, C. L. Cocke, W. Wolff, H. Wolf, S. D. Kravis, M. Stöckli, and E. Kamber, Phys. Rev. Lett. 81, 3627 (1998).
[6] R. Moshammer, A. Perumal, M. Schulz, V. D. Rodriguez, H. Kollmus, R. Mann, S. Hagmann, and J. Ullrich, Phys. Rev. Lett. 87, 223201 (2001).
[7] M. Schulz, R. Moshammer, D. Fischer, H. Kollmus, D. H. Madison, S. Jones and J. Ullrich, Nature (London) 433, 48 (2003).
[8] J. Ullrich, R. Moshammer, A. Dorn, R. Dörner, L. Ph. H. Schmidt, and H. Schmidt-Böcking, Rep. Prog. Phys. 66, 1463 (2003).
[9] J.H. Macek and S.Yu. Ovchinnikov, Phys. Rev. Lett. 80, 2298 (1998).
[10] E.Y. Sidky and C.D. Lin, Phys. Rev. A 60, 377 (1999).
[11] T.G. Lee, S.Yu. Ovchinnikov, J. Sternberg, V. Chupryna, D.R. Schultz, and J.H. Macek, Phys. Rev. A 76, 050701(R) (2007).
[12] J.H. Macek, J.B. Sternberg, S.Yu. Ovchinnikov, T-G. Lee, and D.R. Schultz, Phys. Rev. Lett. 102, 143201 (2009).
[13] J.H. Macek and S.Yu. Ovchinnikov, Phys. Rev. Lett. 80, 2298 (1998); S.Yu. Ovchinnikov, G.N. Ogurtsov, J.H. Macek, Yu.S. Gordeev, Physics Reports 389, 119 (2004).
[14] C.O. Reinhold and R.E. Olson, Phys. Rev. A 39, 3861 (1989).
[15] E. Schrödinger, Phys. Rev. 28, 1049 (1926).
[16] P.A.M. Dirac, Proc. R. Soc. A 133, 50 (1931).
[17] J.O. Hirschfelder, C.J. Goebel, and L.W. Bruch, J. Chem. Phys. 61, 5456 (1974).
[18] I. Bialynicki-Birula, Z. Białynicka-Birula, and C. Sliwa, Phys. Rev. A 61, 032110 (2000).
[19] K.W. Madison et al., Phys. Rev. Lett. 84, 806 (2000).
[20] J.R. Ab-Shaeeer, Science 292, 476 (2002).
[21] M.W. Zwerlein, Nature 435, 1047 (2005).
[22] M.W.J. Bromley and B.D. Esry, Phys. Rev. A 70, 013605 (2004).
[23] F.B. Dunning, J.J. Mestayer, C.O. Reinhold, S. Yoshida, and J. Burgdörfer, J. Phys. B 42, 022001 (2009).
[24] J. Berakar and J.S. Briggs, J. Phys. B 27, 4271 (1994).