New Technique Shows Promise for Analysis of Three-Body Collision Problems

For over half a century, theorists have tried to provide a complete solution to scattering in a quantum system of three charged particles, one of the most fundamental phenomena in atomic physics. Collaborators at Lawrence Berkeley National Laboratory, Lawrence Livermore National Laboratory, and the University of California—Davis have used supercomputers to obtain a complete solution of the ionization of a hydrogen atom by collision with an electron.

As reported in the December 24, 1999, issue of *Science*, the researchers employ a mathematical transformation of the Schrödinger wave equation that makes it possible to treat the outgoing particles not as if their wave functions extend to infinity—as they must be treated conventionally—but instead as if they vanish at large distances from the nucleus.

Bill McCurdy, Berkeley Lab's Associate Laboratory Director for Computing Sciences and a principal author said, "Using this transformation, we compute accurate solutions of the quantum-mechanical wave function of the outgoing particles, and from these solutions, we extract all the dynamical information of the interaction."

McCurdy and his team begin with a transformation of the Schrödinger equation called "exterior complex scaling," invented by Barry Simon of the California Institute of Technology in 1979 to prove formal theorems in scattering theory. The transformation leaves the solution unchanged in regions that correspond to physical reality, producing the correct outgoing waveform based upon the angular separation and distances of two electrons far from the nucleus.

Once the wave function has been calculated, it must be analyzed by computing the "quantum mechanical flux," a means of finding the distribution of probability densities that dates from the 1920s. This computationally intensive process can yield the probability of producing electrons at specific energies and directions from the ionized atom; because electrons are identical, there is no way to distinguish between the initially bound and initially free electron.

As with all scattering problems, the electron-ionization of a hydrogen atom begins with a particle coming in at a certain velocity. This electron interacts with the atom, and two electrons fly out at an angle to each other, leaving the proton behind. The likelihood that a specific incoming state will result in an outgoing state with the particles at specific angles and energies is the "cross section" for that result.

Cross sections of quantum-mechanical processes are derived from the system's wave function, solutions of the Schrödinger equation that yield probabilities of finding the entities involved in a certain state. In scattering problems, wave functions are not localized but extend over all space.

Moreover, said McCurdy of the electromagnetic forces between charged particles, "Coulomb interactions are forever." These infinities make it impossible to define the final state of scattering exactly. "The form of the wave function where all three particles are widely separated is so intractable that no computer-aided numerical approach has been able to incorporate it explicitly."

Previously, however, in the Proceedings of the Royal Society, Colm T. Whelan of the University of Cambridge and his colleagues published their conclusion that all such approximations perform inconsistently and that those few cases which appear to yield good agreement with

https://doi.org/10.1557/mrs2000.1 Published online by Cambridge University Press

experiment "are largely fortuitous." By contrast, the method developed by McCurdy and his team allows the calculation of a highly accurate wave function for the outgoing state that can be interrogated for details of the incoming state and interaction in the same way an experimenter would interrogate a physical system. The researchers acknowledge important advances made earlier by others such as Igor Bray and Andres Stelbovics, whose methods could give the total cross section for ionization of a scattering reaction but could not give specifics such as the directions or energies of outgoing electrons. By contrast, said Thomas Rescigno, a staff physicist at Livermore Lab, "Our work produces absolute answers at the ultimate level of detail."

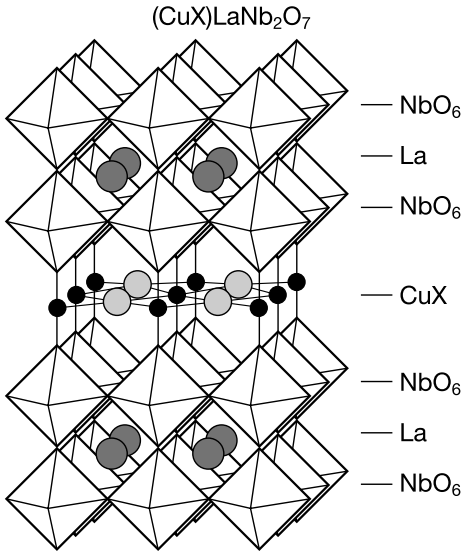
Comparisons with real scattering experiments, such as those recently published by J. Röder et al., who scattered incoming 17.6-eV electrons from hydrogen atoms and measured the angles and energies of the outgoing electrons, prove the accuracy of the new method. The experimental data points match the graph of the cross sections calculated by McCurdy's research team.

"Even if the specific methods have changed, quantum chemistry was founded when the helium atom with two bound electrons was solved—it showed that these problems were, in principle, solvable," McCurdy said. "What we have done is analogous. The details of our method probably won't survive, but we've taken a big step toward treating ionizing collisions of electrons with more complicated atoms and molecules."

The authors conclude that the same computing power and tools necessary for investigating the complexity of increasingly larger systems are also needed "to answer a basic physics question for one of the simplest systems imaginable in physics and chemistry."

Researchers Demonstrate Template for Development of Extended Metal-Anion Arrays

Researchers at the University of New Orleans have synthesized a set of perovskite-related, layered copper-oxyhalides, $(CuX)LaNb_2L_7$, X = Cl and Br), where a copper-halide network was assembled within a double-layered perovskite host (see figure). The synthetic approach that T.A. Kodenkandath and his colleagues of J.B. Wiley's research group describe in the December 1999 issue of the *Journal of the American Chemical Society* demonstrates how host structures can be used as templates in the directed low-temperature (<350°C) assembly of extended metal-anion arrays.



An idealized structure of $(CuX)LaNb_2O_7$. The small dark balls are copper surrounding the large balls, which are X (Cl, Br).

The research team formed new layered copper-oxyhalide from ion-exchange reactions between $RbLaNb_3O_7$ and the copper-halides CuX_2. While this type of exchange usually involve only cations, in $RbLaNb_2O_7$ the copper and halide co-exchanged ions. The researchers relate this to the smaller layer charge of this host relative to other layered perovskites such as $Na_2La_2Ti_3O_{10}$: "to maintain a charge balance, $Na_2La_2Ti_3O_{10}$ would have to exchange two '$CuCl^+$' units, for which there may not be enough room in the interlayer." X-ray diffraction revealed a novel structure for $(CuX)LaNb_2L_7$ containing unusual CuO_2X_4 octahedra that corner-share with NbO_6 octahedra from the pervoskite slab and edge-share with each other along all four equatorial edges. They have successfully applied this chemistry to similar layered perovskites.

Thermal analysis showed that $(CuX)LaNb_2L_7$ decomposes below 700°C, indicating that the compounds are low-temperature phases. As such, according to the researchers, they are likely not accessible by direct reaction because the parent compound must be synthesized at temperatures >700°C.

The researchers concluded that their method of assembling metal-anion networks may lead to new rationally designed materials as applied to nonmolecular systems.

Modified BHA Crystals May Allow Combustion of Methane with Much Less Pollution

Jackie Ying, an associate professor of chemical engineering at the Massachusetts Institute of Technology, has created a barium hexaaluminate (BHA) catalyst that could make it easier to burn methane while drastically cutting emissions of pollutants from natural-gas power plants. Her research team's challenge was to create a catalyst that would allow the combustion process to start, known as "light-off," at a low temperature, but would also be stable at operating temperatures up to about 1300°C. The new BHA crystals are 30 nm in diameter, even at 1300°C, giving them a surface area ten times higher than the surface area for BHA produced by conventional processing. Light-off went down to 600°C from 700°C for BHA crystals formed by conventional processing.

As reported in the January 6 issue of *Nature*, the researchers created a BHA catalyst through a reverse microemulsion in which water droplets only nanometers in diameter are suspended in oil. When added to the water-oil mixture, the principal "ingredients" for the catalyst preferentially move from the oil into the water droplets, where they react. A final heat treatment and powder recovery complete the process.

Ying said that conventional approaches for producing BHA result in a material that is not well mixed before the heat treatment. As a result, the crystallization must be conducted at temperatures so high that particles undergo severe growth. That decreases their surface area, which in turn decreases their reactivity and limits light-off to about 700°C.

In the new process, however, the diffusion of ingredients into the water droplets creates a much more homogenous mixture, which means that the final crystallization heat treatment can be conducted at a lower temperature. That lower temperature suppresses particle growth, maintains the high BHA surface area and reactivity, and allows a lower light-off temperature.

The researchers furthermore experimented with ceria, which is an active catalyst at low temperatures and might allow the desired light-off. Above ~600°C, however, ceria crystals agglomerate, destroying the material.

The researchers' solution is to add ceria to the reverse microemulsion used to produce the BHA particles. The ceria, too, diffuses from the oil into the water droplets, resulting in BHA particles covered with discrete deposits of ceria. Because the

https://doi.org/10.1557/mrs2000.1 Published online by Cambridge University Press