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State-to-State Photodetachment in the Negative Hydrogen Ion

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Abstract

This is the final report of a three-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL) on the fundamental physics of the negative hydrogen ion. New physical phenomena were observed for the first time in the course of this investigation, including the first direct excitation (i.e. without intermediate resonances) of a multiphoton electron detachment resonance in an atomic ion, and the first excess photon detachment of an electron from an atomic species in competition with a single photon process. These new phenomena are relevant to modeling and theory discrimination in three-body quantum mechanics.

Background and Research Objectives

"It could be fairly said that H^- is as important for the development of the quantum-mechanical three-body system as was the neutral hydrogen atom for the development of quantum mechanics itself." (Philip Harris, 1990)

The hydrogen anion, composed of two electrons and a proton, is the simplest coulombic three-body system, yet its Schroedinger equation cannot be solved. It is therefore the ideal system on which to build approximation methods for two-electron systems, as well as for the study of electron-electron correlation. It has only a single stable bound state, and when exposed to a photon of energy greater than 3/4th of an electron volt, the electron can photodetach. Despite its fundamental importance to the field, the detachment spectrum, let alone detailed dynamics, of the hydrogen anion is poorly known. The best experimental work prior to this work, carried out since 1977 at the Los Alamos Meson Physics Facility (LAMPF), measured the photodetachment cross

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section within only one order of magnitude greater than theoretical value, and the resonance widths are not resolved at all.

Ordinary photodetachment of an electron can occur when an ion absorbs a single photon of energy exceeding its electron binding energy. This report concerns the even more interesting processes by which multiple photons can be absorbed. It is known that atoms and negative ions can absorb more photons beyond the minimum number required to detach an electron necessary to detach the electron. In this event the ejected electrons carry off the excess quanta of photon energy as kinetic energy. While such processes are well known in molecules, which have an abundance of resonant states above their ionization threshold, the physical mechanism by which atomic ions do this is not agreed upon. The hydrogen system being the simplest possible negative ion, and lacking bound excited states, is thus the most essential test case for understanding this process.

In particular, two regimes of excess photon detachment (EPD) had never been observed in any negative atomic ion prior to this investigation. We discovered the first EPD process involving a direct multiphoton resonance and we observed the first non-resonant multiphoton EPD in competition with the ordinary single-photon detachment. Follow on studies will compare the deuterated ion's detachment resonances. Because the hydrogen nucleus is so light, isotopic shifts in its spectra can reveal effects due to the electron momentum correlation--such effects are usually so small that they are frequently ignored in approximations to the Schroedinger equation. Thus our studies will help determine how accurately the electron-electron correlation and the excited state dynamics are being calculated in this simplest of three-body systems.

Beyond guiding two-electron quantum theory, the negative hydrogen ion has astronomical and terrestrial importance. It is a major contributor to the opacity of stellar atmospheres and it is the ion most easily accelerated. With the largest charge-to-mass ratio, the hydrogen anion is ideal for creating neutral particle beams--it can be accelerated and then stripped of its electron either with foils or by photodetachment. Because foils can introduce unwanted aberrations, photodetachment offers the possibility of improved beam quality and consequently lower storage ring costs in future accelerator designs. For example, the narrow line width of the Feshbach photodetachment resonance can offer improved hydrogen anion beam diagnostics including a noninvasive momentum dispersion and emittance measurement device.

Los Alamos has consistently been a world leader in the experimental atomic physics of the negative hydrogen ion. In the past H^- has been studied by some rather heroic means made possible by the unique characteristic of the LAMPF 800 MeV accelerator; the ion was illuminated with a laser beam "tuned" by the Doppler shift of the

fast moving ions. The principal drawbacks of this scheme were that the electrons were unavailable for kinetic energy analysis, and the resolution was poor. A much improved approach, utilized in this study, became possible through the use of LANL's ground test accelerator. This bright source had excellent energy resolution, and offered complete access to the ion beam path so that the ejected electrons could be analyzed. Not only could we measure their kinetic energy but we could also examine the anisotropy of their ejected angular distribution.

Importance to LANL's Science and Technology Base and National R&D Needs

The principal theoretical relevance of accurate determination of the H^- spectrum is the understanding of the electron-electron correlation in the two electron system. Our resolution should not only be sensitive to this term in the Hamiltonian but also the smaller, often neglected, momentum correlation term (the two electrons are coupled through their mutual influence on the nuclear momentum). Furthermore, angular state resolution of the ejected electrons is essential to modeling the dynamics of the detachment process.

Spin-offs from this project have potentially great impact on future accelerator and storage ring designs. Presently, beam emittances are measured by invasive techniques and as a result, the beam cannot be monitored continuously during operation. Furthermore, the momentum dispersion, a critical factor affecting the electrical energy efficiency of storage rings, can only be measured coarsely. Photodetachment is currently being considered both for beam neutralization and for noninvasive emittance measurements. The narrow Feshbach resonance can enhance these processes as well as offer a means of measuring the momentum dispersion with high precision. Technology in this area would likely have benefited the now defunct Superconducting Super-Collider, as well future accelerator programs.

Scientific Approach and Accomplishments

The research exploits a unique Los Alamos capability: the low emittance, high current hydrogen anion source built as a ground test accelerator (GTA). At the beginning of this project our foremost goals were simple: 1) determination of the cross section, line width and spectrum of the first Feshbach and Shape resonances of the negative hydrogen

ion and 2) state resolution of the hydrogen atom products. Together these measurements would yield the branching ratio into the various H atom states (1s, 2s, and 2p) at each spectral feature.

As this project progressed, we made some remarkable and unexpected discoveries that refocused our objectives and indefinitely delayed the search for the Feshbach resonance. The discovery that ions had two-photon above-threshold-resonances redirected our emphasis to the study of multiphoton detachment processes. In particular it became important to look for two-photon detachment processes that were not resonantly enhanced as well; while it is intuitively plausible that an excited resonance in the ion could somehow offer a mechanism for multiphoton absorption, it is less obvious how this process would proceed non-resonantly. In either event the process is striking because the optical field must be perturbing the ion itself since a free electron cannot absorb photons.

It took us a year of refinements to our apparatus to realize a sufficient signal-to-noise ratio, but eventually we observed nonresonant excess photon detachment. This was an extraordinarily difficult experiment, and in fact the reviewer of our physical review letter paper pointed out that this was not only the first time that multiphoton nonresonant electron detachment had been observed in an atomic ion but in any atomic species (i.e. neutrals as well) in the high energy photon regime where the single photon energy exceeded the binding energy. Past observations of multiphoton detachment had always been in the low energy regime where the photon energy was below the ions threshold for detachment.

We also succeeded in building a vacuum ultraviolet laser that could tune in the 113 nm region (invented here at Los Alamos by the first two authors). Although we ran out of time before we could return to our original pursuit of the Feshbach and Shape resonances, we could hardly say we were disappointed since our other discoveries overshadowed this.

The hydrogen anion source is derived from the ground test accelerator (GTA), as stated previously. As diagrammed in Fig. 1, we confined the self-repelling negative ions and controlled their momentum spread by accelerating an apertured 30 mA ion beam to 0.8% of the speed of light ($v/c=0.008$; 30 keV). Electrons are photodetached in the accelerated ion beam by a 11 ns laser pulse, producing neutral atoms in several possible quantum states. Several laser sources were used. For the multiphoton resonances we mixed the third harmonic of a Nd:YAG laser (355 nm) with a tunable Dye laser (~640 nm) to produce several millijoules of light near 240 nm (Fig. 1). For the nonresonant study we used an injection seeded Nd:YAG laser to produce a Joule pulse of photons at

the fundamental 1064 nm. Injection seeding was essential to forming a reproducible temporal and spatial profile from the high energy laser; spiking causes unpredictable nonlinear events in the process we were attempting to study. For the Feshbach and Shape experiments we intended to use a pulsed VUV laser based on two-photon resonant four-wave mixing in molecular hydrogen of a tunable argon fluoride laser (ArF; 193 nm) and a tunable Nd:YAG pumped dye laser (a process developed at LANL, see Optics Letters 16(15), 1192, 1991).

The detached electrons are drawn by a magnetic bottle into an electron time-of-flight energy analyzer and monitored with a multichannel plate detector. The heavy undetached ions are not influenced strongly by the magnetic fields and travel downstream on their original paths till they terminate in a Faraday cup monitoring their current.

Analysis of the electron time-of-flight distributions followed the same basic principle in all of the experiments. In the ion center of mass frame the ejected electrons emerge with discrete kinetic energies separated by quanta of the photon energy. In this frame one can visualize the ejected electrons emerging in concentric spherical shells expanding from the neutrals; within each shell the electrons are monoenergetic. In the laboratory frame, some of these electrons are ejected in the ion beam direction and some were ejected in the direction opposed to the ion beam direction. While the net velocity of all the electrons is in the ion beam direction, some electrons are moving faster than others. As a result we can determine angular distribution of the ejected electrons from their arrival time distribution. And since the shells are well separated in energy we can identify how many quanta of energy an electron absorbed.

This time segregation of electrons by photon quanta is what allowed us to observe the multiphoton processes in competition with the single photon energy, as can be seen in Fig. 2. From the shape of the arrival time distribution we can determine the anisotropy of the electron distribution on the shell. For example, in the two-photon resonance case, the hydrogen ion begins in its spherically symmetric ground state and thus initially has no angular momentum. It absorbs two quanta of angular momentum from the two photons, and goes into an excited state resonance. After the electron detaches, the final neutral hydrogen is in its spherically symmetric ground state, and thus has no angular momentum. Therefore the angular momentum of the absorbed photons must be deposited in orbital angular momentum between the electrons and the neutral hydrogen. Since two photons were absorbed the net angular momentum could be either two quanta (if the photons had the same handedness) or zero (if the two photons were of opposite angular momenta). This means the orbital angular momentum must either be a D-state ($j=2$), an S-state ($j=0$), or a linear combination of these states. The orbital angular

momentum maps into the angular recoil distribution of the electrons in such a way that a D-state produces a D-orbital or "D-Wave" shaped distribution of electrons (Fig. 3).

We observed that the ejected electrons indeed emerged as a D-wave distribution, and not as an S-wave. This tells us that the symmetry of the intermediate excited-state resonance of the hydrogen ion had to have been a D-state and not an S-state.

We can tell that this is in fact a resonance by scanning the laser wavelength to map out the peak. The integrated electron yield for the two-photon process is shown as a function of wavelength in Fig. 4. The line shape is the well known Fano profile that occurs when one has a resonance embedded in a continuum background. Also shown in the figure is a prediction of the line shape using a modern approximation to the Schroedinger equation. The difference shows that this approximation technique is not adequate to predict the exact transition energy. Other competing approximation techniques (not shown) predicted the resonance energy more exactly. These differences highlight in a simple way the importance of these experimental measurements to refining 3-body quantum mechanics.

Summary of Scientific And Technical Highlights

- First observation of direct (no intermediate states) multiphoton excitation of a resonance in an ion.
- First observation of multiphoton detachment in competition with single-photon detachment (high photon energy regime).
- First electron energy analysis in H- photodetachment.
- First branching ratio determination of hydrogen intermediate states. (100% D 0% S)
- Construction of a new high power VUV laser system.

Peer Reviewed Publications

1. Phys. Rev. Lett., V75(#16), P2924 (1995).
2. Phys. Rev. Lett., V78(#9), P1565 (1997).
3. Phys. Rev. Lett., Isotopic effects in photodetachment, manuscript in preparation.

Fig 1: Experimental layout for multiphoton resonance study.

Fig 2. **Top:** Electron time-of-flight distribution after laser pulse. The shape corresponds to electrons ejected in a P-wave distribution; the early arrivals near 300 ns were ejected forward along the ion beam direction. The arrival time corresponds to the absorption of a single photon of energy. The tiny spike at $T=0$ ns is from scattered laser light. **Bottom:** Time region from 150 to 300 ns magnified 1000 times reveals existence of another peak in the arrival time distribution. This early peak corresponds to the electrons ejected along the ion beam direction that came from ions absorbing two quanta of photon energy. The shape of this peak corresponds to a D-wave ejection angle distribution. For comparison an S-wave distribution is also drawn on the figure. Prior to this work, direct two-photon excitation of an EPD resonance had not been observed.

Fig 3. D-wave angular distribution

Fig 4. The two-photon electron yield as a function of two-photon energy. The asymmetric shape of the profile is indicative of quantum interference between the resonance and continuum background. The theoretical prediction of this line shape (slightly to the right) captured this asymmetry but missed the transition energy.