Observation of Two-Electron Photoionization of the H^- Ion near Threshold

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Photoejection of both electrons from the H^- ion by a single incident photon has been observed with use of a crossed-relativistic-beam technique. The dipole selection rules for a single photon pick out the $^1P^o$ final state. The relative cross section was measured over a 0.30-eV range of photon energies near threshold in steps of 0.007 eV with a resolution of 0.007 eV full width at half maximum. The energy dependence of the cross section in this region was consistent with a Wannier threshold law.

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We present the first results from our study of the threshold behavior of two-electron photoionization of the H^- ion. Our experimental technique exploits the large Doppler shift of the photon energy which results when a laser beam is crossed with a relativistic ion beam. By varying the angle α between the two beams, the photon energy in the rest frame of the ions can be continuously tuned according to the relation

$$E = \gamma E_{\mathrm{lab}}(1+\beta\cos\alpha), \tag{1}$$

where E_{lab} is the energy of the photons in the laboratory frame of reference, $\beta = v/c$ for the ions, and $\gamma = (1-\beta^2)^{-1/2}$. The 800-MeV H^- ion beam at the Clinton P. Anderson Meson Physics Facility (LAMPF) has $\beta = 0.842$ and $\gamma = 1.854$. Using a frequency-quadrupled neodymium-doped yttrium aluminum garnet laser with $E_{\mathrm{lab}} = 4.65$ eV, we can tune the photon energy from 1.63 to 15.6 eV in the rest frame of the ions.

The threshold behavior for three separated charged particles at positive energy has been the subject of considerable experimental and theoretical interest. In order for both electrons to break free of the proton near threshold, their motion must be highly correlated. Such strongly correlated motion is not adequately described by the independent-particle model which is our major tool for understanding atomic, molecular, and nuclear problems.

The theoretical literature on this subject includes both classical[1] and quantum mechanical[2,3] treatments. The theories usually predict a power law for the threshold energy dependence of the cross section,

$$\sigma(E) \propto (E - E_t)^m, \tag{2}$$

where E is the energy, E_t is the threshold energy, and m is an exponent which ranges from 1.0 to 1.5 depending upon the degree of correlation which the theory assumes. For example, the theory of Wannier[1] predicts $m = 1.1268\ldots$ for a $Z = 1$ ion in a state having $^1S^e$ spin, orbital angular momentum, and parity. There are also theories which predict more complicated behavior such as a modulated linear law.[2]

The experimental problems associated with the threshold behavior of hydrogen, the simplest atomic system, are formidable. In the case of two-electron photoionization of H^-, the photon energies required are in the vacuum ultraviolet region. Indeed, we know of no previous observation of this fundamental process. In the case of electron scattering from the neutral atom, it is difficult to make sufficiently monochromatic electron and atomic hydrogen beams. Most previous experimental work has therefore been done by using electron scattering on helium with energy resolutions in the range of 0.06 to 0.10 eV. The electron scattering experiment of McGowan and Clarke[4] on hydrogen and the later helium experiments[5-7] have found exponents consistent with the Wannier law. The interpretation of these experiments is complicated by the broad energy resolutions and the unknown quantum numbers of the final state. A recent experiment[8] and more re-

cent theories[9,10] have addressed the latter problem.

Our photoionization experiment has a factor-of-10 better energy resolution than the electron scattering experiments and so presents an opportunity to look for structure. The systematic effects are also much different since the ions have a high kinetic energy in the laboratory reference frame. Our experiment also picks out the $^1P^o$ final state because of the dipole selection rules for the absorption of a single photon by the $^1S^e$ ground state of the H^- ion.

A schematic of the experiment is shown in Fig. 1. The protons (H^+) produced in the two-electron photoionization reaction

$$\gamma + H^-(1S) \rightarrow H^+ + e^- + e^- \quad (3)$$

and the neutral hydrogen atoms (H^0) produced in the one-electron photodetachment reaction

$$\gamma + H^-(1S) \rightarrow H^0(np) + e^- \quad (4)$$

are separated from the H^- beam and directed into their respective detectors by the magnet. The symbol $H^-(1S)$ refers to an H^- ion in the ground state while the symbol $H^0(np)$ refers to a hydrogen atom in a p state having principal quantum number n. A fast ($\Delta t = 50$ ns) time coincidence is formed between the laser flash and the arrival of the ions. The data are reported as relative cross sections since uncertainties in the temporal and spatial overlap of the two beams prevented us from making absolute measurements. The technique used in this experiment is an extension of that which was used in our one-electron photodetachment experiments.[11,12] The details of that apparatus are discussed in a forthcoming review article[13] and the details of the present apparatus are available in a report.[14]

In addition to the signals from Reactions (3) and (4), there were also backgrounds from gas stripping and field ionization. The gas stripping was caused by residual gas molecules in the vacuum system and appeared as a constant background in both the H^+ and H^0 detectors. It was studied by observing the detector signals with a long time delay in the coincidence and it was minimized by a good vacuum and a fast time coincidence.

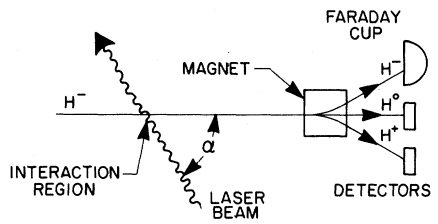

FIG. 1. Schematic view of the crossed-beam apparatus used in the two-electron photoionization studies. The deflections of the beams are greatly exaggerated in the drawing; in reality they are approximately 2 mrad.

The field-ionization background resulted from a two-step process in which the highly excited atoms produced in Reaction (4) were pulled apart by the motional electric field in the magnet. The two-step process was studied by looking at each of the steps separately. The total cross section for Reaction (4) was directly observed in our H^0 detector and was constant with energy near 14.35 eV. The contribution to the cross section of the various excited states was studied by making the magnetic field progressively weaker: 3600, 2200, 780, 380, 22, and 13.6 G. The signal in the H^+ detector, in excess of that from Reaction (3), corresponded to atoms with $n \geq n_c$, where n_c is the principal quantum number at which ionization occurs. At the highest field only the states with $n \geq 4$ were observed and we were able to observe new H^- resonances[15] which had been masked by the lower states. Since the cross section for Reaction (4) was dominated by the lower n values, the field-ionization contribution to the cross section decreased as the field was lowered and the data at the lower fields showed no structure. The lowest field of 13.6 G corresponded to a motional electric field of 6.4 kV/cm and could ionize only states with $n \geq 15$.

The second step of the field-ionization process was studied by forming an $H^0(1s)$ atomic beam out of the H^- beam, photoexciting the atoms to $H^0(np)$, and then allowing them to ionize in the magnet. The cross section which we measured by using a motional electric field of 820 kV/cm is shown in Fig. 2. Near the ionization threshold at 13.6 eV, the photoexcitation cross section approaches a constant and has no structure other than statistical fluctuations. This is because the density of states is increasing like n^3 while the oscillator strength is decreasing like n^{-3}. The figure illustrates that the ionization efficiency of the magnet is constant at high n and that the energy separation between the states is smaller than our energy resolution in the threshold region.

These studies indicate that most of the background has been eliminated and that what remains is probably without structure since the magnet has constant efficiency and the Lyman lines are

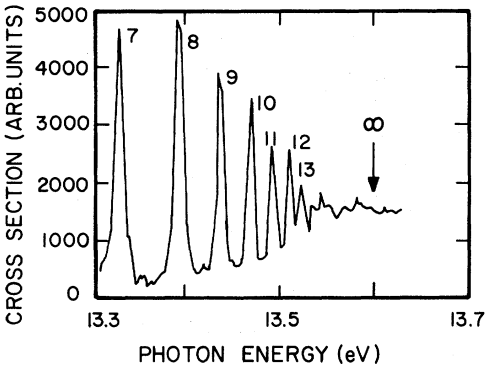

FIG. 2. Relative cross section for the photoexcitation of an H^0 beam followed by ionization in a motional electric field of 820 kV/cm. The numbers refer to the principal quantum numbers of the Lyman series.

unresolved near threshold. We believe that the net effect was an apparent threshold for Reaction (3) which was approximately 0.05 eV lower than the energetically allowed threshold at 14.352 eV. The anomalous threshold required us to leave the threshold energy a free parameter in our fits with theory. The proper quantitative approach to this problem is to take high-precision data at several different field settings, fit the data with the various threshold laws, and then extrapolate the results to zero field. Since our beam time was limited, our data at the higher fields do not have sufficient precision for this task. Such an analysis will probably have to await a second-generation experiment with ten times the data acquisition rate.

The energy scale was calibrated by using the narrow Feshbach resonance. The energy at the maximum of the Feshbach resonance was taken to be 10.925 eV from the theoretical calculation of Broad and Reinhardt[16] using the hydrogen reduced Rydberg. Our energy resolution was determined to be 0.007 eV full width at half maximum by measuring the width of this resonance.

The threshold cross-section data are shown in Fig. 3. The errors are statistical only and the flat background is due to an incomplete subtraction of the residual-gas stripping. The solid line in Fig. 3(a) is the best fit by a power law of the form

$$\sigma(E) = A(E - E_t)^m + B \ . \tag{5}$$

The fit was done by minimizing the χ^2 sum between the data and the theoretical curve (5) using the nonlinear optimization routine MINUIT.[17] The fit results were $A = 38.5 \pm 1.5$, $B = 0.68 \pm 0.05$, $E_t = 14.305 \pm 0.005$ eV, and $m = 1.15 \pm 0.04$. The χ^2 was 76 for 66 degrees of freedom giving a confidence level of 19%.

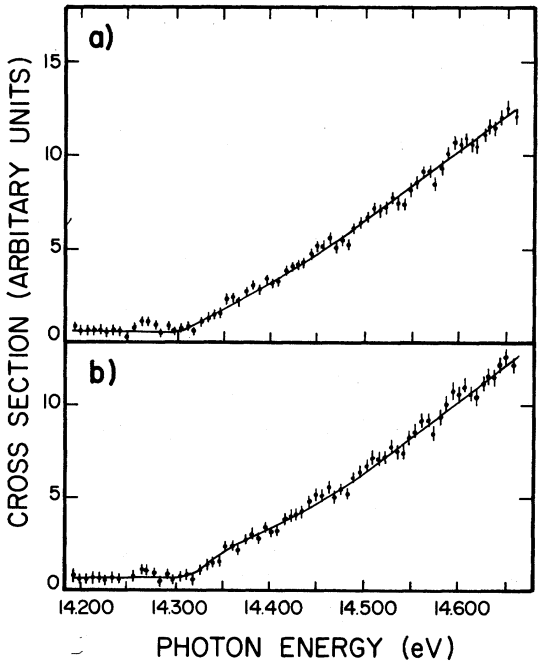

FIG. 3. Cross section for the two-electron photoionization process. The errors are statistical only. (a) The curve is the result of a best fit by a power law. (b) The curve is the result of a best fit by a modulated linear law.

The value obtained for the exponent is consistent with the Wannier law and not with a linear law. If we assume that the power law with $m = 1.000$ or with $m = 1.127$ are the only two possible discrete hypotheses, we can use the method of likelihood ratios to calculate betting odds of 250:1 in favor of the Wannier law using statistical errors only. The recent theoretical work of Greene and Rau[10] predicted Wannier threshold behavior for the $^1P^\circ$ final state.

We have also fitted the data by a modulated linear law of the form suggested by Temkin,[2]

$$\sigma(E) = (E - E_t)\{A + D \sin[C \ln(E - E_t) + F]\} + B \ . \tag{6}$$

The results of the fit were $A = 33.5 \pm 0.4$, $B = 0.69 \pm 0.04$, $C = 41.6 \pm 0.9$, $D = 1.5 \pm 0.4$, $E_t = 14.321 \pm 0.002$ eV, and $F = 5.2 \pm 0.8$. The fit had a χ^2 of 71 for 64 degrees of freedom which gave a confidence level of 25%. The fit is shown in Fig. 3(b).

We have presented the first data on two-electron

photoionization of the H^- ion and the data are consistent with a Wannier threshold law over the 0.3-eV region above threshold. We can discriminate against a simple linear law but we cannot tell the difference between the Wannier and the modulated linear law because of the field-ionization background. The present experiment has provided us with limits on the parameters which appear in these hypotheses. These limits can serve as a guide to theory and to the design of second-generation experiments.

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Instabilities, Self-Oscillation, and Chaos in a Simple Nonlinear Optical Interaction

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It is shown that two light beams interacting in a third-order nonlinear medium undergo transition from a stationary to periodic and chaotic states, as their intensities are increased. A threshold for the onset of instabilities is calculated and verified by computer simulations. It is therefore proved that external feedback is not necessary for self-oscillations in nonlinear optical systems.

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The stability of nonlinear optical systems has been the subject of an intense study lately, especially since Ikeda[1] has shown that a Fabry-Perot ring resonator containing a saturable absorber can show instability in regions considered stable before. Later Ikeda, Daido, and Akimoto[2] proved that a ring resonator which contains a third-order dispersive nonlinear medium undergoes successive bifurcations as the incident power is increased, leading to chaos or "optical turbulence." Similar behavior was later predicted for standing-wave resonators[3] and for distributed-feedback resonators.[4] Gibbs *et al.*[5] recently demonstrated the main features of these predictions in a synthesized Fabry-Perot resonator.

In all these works the external feedback, supplied by the mirrors of the resonators, was necessary for the onset of self-oscillations. In this Letter we show that self-oscillations and chaos can be obtained in an optical system without any external feedback. Specifically, we consider two monochromatic waves interacting in a third-order dispersive nonlinear medium. A steady-state solution always exists, according to which the two

