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Observation of Narrow Resonances in the H⁻ Photodetachment Cross Section near the n=3 Threshold

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Two pronounced dips in the H⁻ photodetachment cross section have been observed at photon energies of 12.650 ± 0.004 and 12.837 ± 0.004 eV, just below the hydrogenic n = 3 threshold. These energies were reached by crossing a uv laser beam with a relativistic H⁻ beam. The dips are interpreted as the first two members of a ¹P Feshbach resonance series, of the "+" type, converging on n = 3. Fano line shapes have been fitted to these structures to extract their intrinsic widths and line profile indices.

The H⁻ ion, the most elementary two-electron system, has strictly speaking only one bound state.¹ Yet the results of experimental studies² on the excited states of this system indicate the existence of a complex resonance structure based on the excitation of both electrons followed by the autodetachment of one of them. Theoretical studies of these autodetaching resonances suggest that they can be surprisingly narrow and hence difficult to detect. In this Letter we report the first observation of two such resonances in the H⁻ photodetachment cross section near the hydrogenic n=3 threshold.

The experimental method used was an improvement of the high-resolution laser-induced photodetachment technique developed by Bryant *et al.*³ at the Clinton P. Anderson Meson Physics Facility (LAMPF). The crossing of a uv photon beam from a standard laboratory laser with the relativistic 800-MeV H⁻-ion beam at LAMPF resulted in photon energies in the H⁻ ion's center-of-mass frame which were tunable over a large range by varying the angle of intersection. This Dopplershifted energy of the photons is given by

$$E = \gamma E_L (1 + \beta \cos \alpha), \qquad (1)$$

where E_L is the laboratory photon energy, $\beta = v/c$ of the H⁻ beam, $\gamma = (1 - v^2/c^2)^{-1/2}$ and α is the angle of intersection with the ion beam, defined so that $\alpha = 0$ is head-on. The new precision optical system of the apparatus allowed the angle of intersection to be varied in 0.314 mr increments. Using the 4.659-eV photon beam from the fourth harmonic of the 1.06- μ m emission of a Nd-doped yttrium-aluminum-garnet laser,⁴ E was continuously tunable from 1.36 to 15.9 eV for the 800-MeV H⁻ beam. The detached electrons were swept from the beam by a small magnetic field and were detected with a silicon solid-state detector and identified by their unique energy (435 keV) and timing with respect to the laser pulse. The resulting electron signal was found to be ex-

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ceedingly clean with a negligible background produced by gas stripping of the H⁻ beam in the 10^{-8} -Torr residual vacuum.

The energy resolution of this experiment was determined principally by the width and variation in the centroid of the H⁻ beam energy, and by the angular divergences of the ion and laser beams. The beam energy spread, typically 500 keV, and drifts in the energy centroid, each resulted in a spread of ~0.004 eV in the center-of-mass photon energy. The angular divergences of the ion and laser beams each contributed another 0.003 eV. The estimated total energy spread of 0.006 to 0.007 eV full width at half maximum (FWHM) was in good agreement with measurements made of the n=2 Feshbach resonance width, which is a direct measure of the energy resolution of the system,³ since the true width of the resonance is predicted to be narrow.⁵

Since the photodetachment cross-section measurement depends in part upon the difficult to determine overlap (temporal as well as spatial) of the intersecting beams,⁶ only relative cross sections were obtained. Variation in this overlap during a measurement produced random fluctuations in the cross section that exceeded the uncertainty expected from the electron counting



FIG. 1. Photodetachment cross section vs photon energy below the n = 3 threshold showing the two prominent dips. The cross section has been normalized to agree with the theoretical value of Broad and Reinhardt (Ref. 5) at 10.90 eV. The dashed line represents a fit to the large dip at 10.650 eV using a Fano line shape convoluted with a Gaussian resolution function of 0.007 eV FWHM. See text for a discussion of error bars and energy assignments.

statistics. The uncertainties were therefore determined by the consistency of repeated measurements.

Figure 1 displays the cross sections resulting from the combination of three sets of data taken in 0.638-mr steps (~0.004 eV) covering an energy range from 12.567 to 12.855 eV. To compensate for H⁻ beam energy shifts, the energy scales of two of the sets of data were shifted to bring the measurements in the region of the sharp decline near the 12.650-eV structure into agreement with the third set, for which an absolute determination of the intersection angle was made. This set of data also extended down to the n = 2 region and could therefore provide a calibration of both the energy and cross-section scales. An energy scale was determined by assigning a photon energy of 10.930 eV (Ref. 5) to the angle of the ${}^{1}P$ Feshbach resonance just below the n = 2 threshold, and the cross-section scale was assigned by normalizing to the calculated values of Broad and Reinhardt⁵ in the continuum region at 10.90 eV. The other data sets were normalized by equalizing the area under the curves. The systematic error in assigning the energy by this method was estimated to be less than $\pm 0.004 \text{ eV}$; the error bars shown represent the standard deviations of the combined data in each energy bin.

Figure 2 displays the combined results of four



FIG. 2. Photodetachment cross section taken in 0.002eV energy steps across the 12.837-eV dip. The same cross-section normalization as in Fig. 1 was used. The dashed line again represents a fit utilizing a Fano line shape convoluted with the same Gaussian resolution function of Fig. 1.

additional sets of data which scan in finer steps $(\sim 0.002 \text{ eV})$ the small dip seen just below the n = 3 threshold in Fig. 1. The same energy shifts used in Fig. 1 were used here, and the cross section was normalized to agree with Fig. 1 in this region. This dip appears to be a small replica of the large one.

Both features have been fitted to a Fano line shape, which arises from the interference of a Breit-Wigner resonance with a constant continuum. An expression for this shape is given by⁷

$$\sigma(\epsilon) = \sigma_a \left[(q + \epsilon)^2 / (1 + \epsilon^2) \right] + \sigma_b , \qquad (2)$$

where $\epsilon = (E - E_0)/(\frac{1}{2}\Gamma)$, *q* is the line profile index, σ_a is the resonant cross section, and σ_b is the nonresonant cross section. Table I presents the energies and widths obtained by fitting the cross section for the two states with a convolution of this line shape and a Gaussian resolution function of 0.007 eV FWHM, the experimentally observed resolution. The errors on the widths include the effects of varying this resolution by ± 0.001 eV.

In this analysis q was allowed to be a free parameter for each state. The two values found, -0.81 ± 0.02 for the lower state and -0.67 ± 0.14 for the upper state, are similar and within the uncertainty could be the same. According to Fano and Cooper,⁷ resonances having similar q's are likely to be members of the same Feshbach series. Another quantity of interest is the effective strength⁸ of the resonance, defined as $\rho^2(q^2+1)$, where $\rho = \sigma_a / (\sigma_a + \sigma_b)$. It is interesting to note that the fits gave essentially the same effective strength for each resonance, i.e., 0.32 ± 0.02 and 0.26 ± 0.06 for the lower and upper states, respectively. The higher-energy state only appears weaker in the spectrum due to the filling in of the narrow structure by the experimental resolution.

A dip similar in appearance to the lower-energy dip in the present data and near the same energy has been observed by Williams⁹ in an e^{-} H⁰ elastic-scattering study in which it was identified as a ¹*P* resonance. In the absence of magnetic or electric fields, the present experiment should populate only ¹*P*° states by single photon excitation. Table I lists some theoretical predictions¹⁰⁻¹³ for ¹*P* structures near the n = 3 threshold for comparison to the results of the present study. Based on the measured energies and widths, the two states reported here most likely correspond to the first and fourth predicted resonances as listed in the table.

In order to present a possible physical interpretation of these states, we focus on the recent theoretical work of Greene¹³ which has applied the analytical technique of Lin¹⁴ to the region below the n = 3 threshold. This method allows the approximate separation of the two-electron Schrödinger equation, leading to a one-dimensional potential in hyperspherical coordinates. It also utilizes a "+" and "-" classification scheme similar to that used to describe doubly excited states of helium.¹⁵ Using this method, Greene has predicted energies and relative widths for the first two members of both the "+" and "-" Feshbach series below n = 3 as indicated in the table. It should be noted that the two "+" states correspond to the first and fourth resonances predicted in Refs. 10-12, while the two "-" states correspond to the second and third states in those studies. As is evident from the table the predicted energy for the second "+" state agrees very well with the measured energy for the upper resonance, although the prediction for the lower state's energy is somewhat higher than the data

TABLE I. Predicted energies for the first four ¹P resonances near the n = 3 hydrogenic threshold (12.8483 eV for $H^- \rightarrow H_{n=3}^{-0*} + e^-$; electron affinity for H^0 taken as 0.754 22 eV) and corresponding widths (given enclosed in parentheses, if available) in electron volts compared with the energies (widths) of the two states extracted from the present data. See text for discussion of "+" and "-" designations under Ref. 13.

State	Ref. 10	Ref. 11	Ref. 12	Ref. 13	This experiment
1	12,6586 (0.0329)	12.6605	12,6602	12.6687 ^a	$12.650 \pm 0.004^{b} (0.0275 \pm 0.0008^{b})$
2	12,7677 (0.00012)	12.7656	12.7658	12.7640°	•••
3	12.8382 (0.000 30)	12.8330	12.8332	12.8320 ^c	• • •
4	12.8416 (0.0022)	12,8394	12.8408	12.8375 ^a	12.837 ± 0.004^{b} (0.0016 ± 0.0003 ^b)

a"+" designation.

^bErrors assigned to energies or widths in the present experiment.

c"-" designation.

indicate. A simple exponential scaling law predicts the ratio of the widths for the two "+" states to be 0.0597 (Ref. 13) which is in excellent agreement with the measured ratio of 0.057 ± 0.011 . On the basis of the predictions of energies and relative widths, the two resonances observed in this work could be interpreted as the first two members of the "+" Feshbach series. In a simple classical picture, this corresponds to the two electrons being at nearly equal distances from the nucleus at all times. The "-" states, on the other hand, have one electron near the nucleus whenever the other is far away. In this picture the "+" states are expected to autodetach at a much greater rate than the "-" states partly due to the greater perturbative effect of the Coulomb repulsion for two electrons at small separations.¹⁵

As further evidence of the "+" nature of the states seen here, the large resonance structure at 12.650 eV appears to be only slightly affected by a strong electric field (produced in our apparatus by the Lorentz transformation of a modest transverse magnetic field), in striking contrast to the behavior of the Feshbach resonance¹⁶ near n=2 which has "-" symmetry in Lin's analysis.¹⁴ Data taken with an barycentric electric field of 1.2 MV/cm show very little difference from zero field data; q decreases by $(13 \pm 6)\%$ Γ increased by $(13 \pm 7)\%$ and E_0 shifted by $+0.005 \pm 0.004$ eV. In its resistance to quenching by an electric field, this state behaves more like the n=2 shape resonance.¹⁶ which in Lin's picture is a "+" resonance. This supports the conjecture that the two prominent dips observed just below n = 3 and the shape resonance just above n = 2 arise from the decay of states having in common a "+" symmetry.

In addition to the two structures reported here, there is preliminary evidence for further structure below the n=3 threshold; e.g., there appears to be another weak dip near 12.78 eV (which could be the first "-" state in Green's formulation or the second resonance in the other theories.) There is also evidence for some structure near the n=4 threshold, which we plan to study under higher resolution.

In summary, we report here the first observation of narrow resonances in the H⁻ photodetachment cross section near the hydrogenic n = 3 threshold, utilizing a high-resolution, Dopplershifted crossed-beam technique. Even though one of the states (12.650 eV) has been previously reported in e^- -H⁰ scattering, the present measurement represents more than an order-of-magnitude improvement in precision over the electron scattering data. The higher-energy state (12.837 eV) has not been observed to our knowledge prior to this study.

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