Branching ratio of the $H^-(n=2)$ shape resonance

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(Received 15 June 1992)

The relative photodetachment cross section for decay into the $H(N=2)$ channel by the $^1P^\circ$ shape resonance in H^- was measured, as well as that for decay into all channels. The branching ratio $\sigma(N=2)/\sigma(\text{total})$ was computed for a series of energies between 10.95 and 11.3 eV after normalizing the cross sections to theoretical peak amplitudes. The maximum branching ratio (≈ 0.8) appears at an energy about 20 meV higher than the central energy of the resonance. Results are compared with recent theoretical calculations.

PACS number(s): 32.80.Fb, 31.50.+w, 32.80.Cy

I. INTRODUCTION

The H^- shape resonance (SR), lying just above the threshold for production of $H(N=2)$, was first predicted in 1967 [1] by Taylor and Burke for electron-hydrogen scattering, and by Macek for photoionization of H^-. The first observations of this resonance to our knowledge were reported in 1969 by McGowan, Williams, and Carley [2] and later by Williams and Willis [3] in 1974. Both measurements involved electron scattering from neutral hydrogen atoms. A series of high-resolution photodetachment experiments [4] on H^- at the Los Almos Meson Physics Facility (LAMPF) pinned down the position of this feature and led to some understanding of its behavior when subjected to external electric fields.

The autoionizing $n=2$ shape resonance is the largest resonance in the H^- photodetachment spectrum and a key feature in understanding electron correlations. Unique in that its potential has three classical turning points, it is well known as the only resonance of its type yet observed in the H^- photodetachment spectrum (or equivalently in electron-impact excitation of H atoms). The SR results from a centrifugal barrier potential, and appears above the threshold for excitation of the $H(N=2)$ state. It can therefore autodetach to either the H(2) or H(1) continuum. Its slightly-lower-lying neighbor—the ^1P $n=2$ Feshbach resonance at 10.9264 eV [5]—is energetically capable of decay only to the ground state of neutral hydrogen, the $N=1$ channel. Since the SR is the only H^- resonance that has been observed to decay to its parent state, the measurement of its branching ratio (Sec. VI), which demonstrates the substantial effects of electron correlation, confirms the important dynamical differences between shape and Feshbach-type states. Details of the parameters such as widths and asymmetries (Sec. V) may be especially useful in current theoretical studies of static electric-field effects on the SR.

Here we report measurements of relative cross sections for total decay of the $H^-(n=2)$ SR and for its partial decay into the $H(N=2)$ channel. We normalized these cross sections to theoretical peak values to arrive at branching-ratio values. This analysis makes use of data acquired at the LAMPF accelerator [6,7].

II. EXPERIMENTAL METHOD

In these experiments, relativistic H^- ions ($\beta=0.842$) collide at a precisely controlled angle with photons produced by a Nd:YAG laser. In the coordinate system of the moving ion the Doppler-shifted energy of the photons is given by

$$E_{\text{c.m.}}=\gamma E_{\text{lab}}(1+\beta\cos\alpha)\ , \tag{1}$$

where β and γ are the usual relativistic quantities, E_{lab} is the energy of the laser, and α is the angle of intersection between the beams, defined to be zero when the laser beam meets the ion beam head on.

A schematic diagram of the apparatus is shown in Fig. 1. The setup is similar to that used in other photodetachment experiments at LAMPF, except that in the partial cross-section measurement the ions encounter two photon beams of different energies.

The first beam, whose angle of intersection is varied, detaches electrons from the H^- ions by the reaction

$$H^-+h\nu\rightarrow H^-(n=2)\rightarrow H(N=1 \text{ or } 2)+e\ , \tag{2}$$

where $h\nu=E_{\text{c.m.}}$ is the Doppler-shifted energy of the fourth harmonic of the laser ($E_{\text{lab}}=4.66$ eV). The inter-

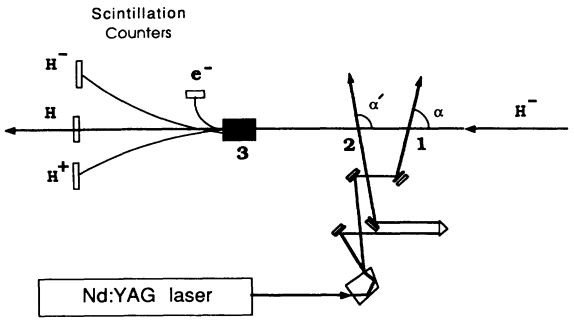

FIG. 1. Diagram illustrating the basic principle of the experiment. In area 1, the fourth harmonic (4.66 eV) of the Nd:YAG laser intersects the 800-MeV H^- beam at an angle α which produces the reaction $H^- + h\nu \rightarrow H^- \rightarrow H(1 \text{ or } 2) + e$. In area 2, the first or second harmonic of the laser intersects the atomic beam at an angle α' which produces the reaction $H(2) + h\nu' \rightarrow H(N')$. In area 3, the stripping magnet [6] or electron spectrometer [7] produces a field in the H^- center-of-mass frame which strips the $H(N')$ atoms of their electrons, but has no effect on atoms in lower states. Decay products, protons [6] or electrons [7], are detected using scintillation counters.

mediate excited state has an extremely short lifetime, on the order of 30 fs.

About 1.5 ns later the H atoms, some in the $N=2$ excited state, encounter the second laser beam in the "promotion" chamber. This beam, derived from the first [6] or second [7] harmonic of the Nd:YAG laser, intersects the ions's trajectory under an angle fixed to produce the Doppler-shifted energy $h\nu'$ needed to excite $H(N=2)$ to a higher excited state N'. Atoms arriving in the ground state remain unaffected.

Downstream from this reaction region, the atoms encounter the field of a stripping magnet. The Lorentz transformation changes the laboratory magnetic field to a very large electric field in the rest frame of the particles according to $F_{\text{c.m.}} = \gamma\beta c B_{\text{lab}}$. The field is tuned to be strong enough to immediately strip all $H(N')$ atoms of their electrons. It has no effect on the H ground-state atoms. The detection reaction for the partial cross-section measurement is thus

$$H(N=2) + h\nu' \rightarrow H(N') \Longrightarrow H^+ + e \ , \tag{3}$$

where the double arrow indicates that the second step proceeds in an electric field. The stripping magnet also separates the resulting protons from the H and H^- ions.

In the 1983 experiment $H(N=2)$ was promoted to $H(N'=7)$, which was then field stripped. The signature for an atom left in the $N=2$ state after the original photodetachment was then the detection of a proton or "H^+" in a scintillation counter. For the total cross-section measurement, only the first laser beam was enabled, and neutral atoms were detected. The atoms exited through a thick foil 13 m downstream of the interaction region, and the resulting protons immediately entered our scintillator-photomultiplier-tube counters.

In a recent experiment, $H(N=2)$ was promoted to $H(N'=11)$ by the second laser beam [7]. Electrons were stripped from H(11) and collected by an electron spectrometer in the partial decay measurement. As in earlier work [6], only the first laser was used for the total cross-section measurement, but the signature was the detection of photodetached electrons in the electron spectrometer.

This detection scheme is flawed in that the angular momentum content of the observed H(2) atoms is not well characterized. That is, we do not know how many of those arriving at the electron spectrometer are in the $2s$ state and how many in $2p$—for two reasons.

First, the promotion chamber is located 27.75 inches downstream from the H^--photon interaction chamber. This means that, in the frame of the moving atom, 1.5 ns elapses before it reaches by the second laser. The lifetime of the $H(2p)$ state is only 1.6 ns, so that approximately 60% of the $H(2p)$ atoms decay to $H(1s)$ before reaching the promotion region. Second, a stray field in the lab could cause Stark mixing of the $2s$ and $2p$ levels. We estimate the *maximum* field possible along the flight path to have been 1 G. Given this field strength and assuming dipole oscillations only, the probability that a $2s_{1/2}$ ($2p_{1/2}$) atom leaving the interaction region will occupy that state after traveling 28 in, was calculated to be 0.51 (0.36). Regarding the transition probabilities in the second interaction region, we note that the fraction of $2s$ atoms promoted to the N' state by the second laser is equal ($\pm 5\%$) to the fraction of $2p$ atoms promoted [8], and small differences here may be disregarded.

Since we report relative cross sections only, the above effects would not be problematic if the $2s$ and $2p$ profiles had exactly the same shape. According to calculations of Sadeghpour [9] and Callaway [10], however, the cross sections probably differ above about 11 eV, with the $2s$ cross section falling off faster than the $2p$ (Fig. 2). For that reason, we consider the experimental cross sections and branching ratios below this energy to be more reliable than those above it.

For total cross-section calibration, the energy region of the $n=2$ Feshbach peak was scanned. In off-line

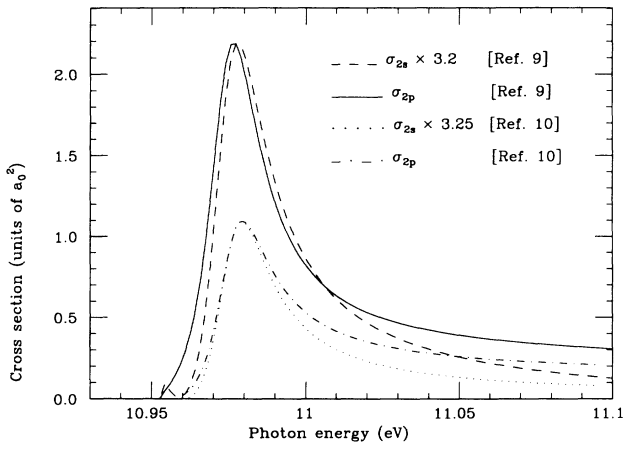

FIG. 2. $2s$ and $2p$ profiles for the $H^-(2)\ ^1P^0$ shape resonance from R-matrix calculations of Sadeghpour for photodetachment [9] and variational calculations of Callaway for electron-impact excitation [10]. The $2s$ profile has been normalized to the $2p$ cross section at its peak in both cases to emphasize the comparative drop-off in the high-energy region.

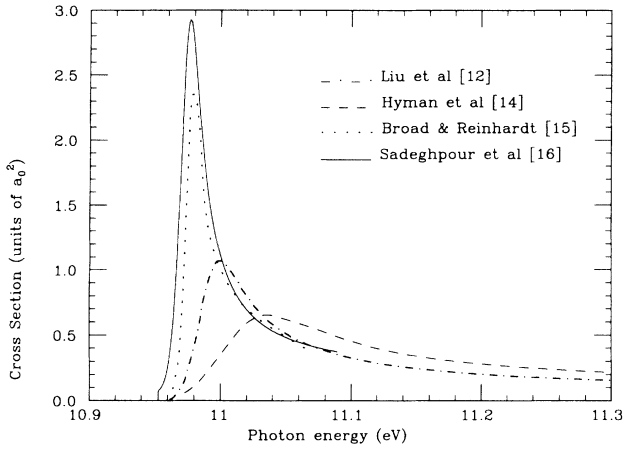

FIG. 3. Theoretical photodetachment partial cross section vs energy above the threshold for production of H(2) plus a free electron. Of the curves shown, the most recent calculation is that of Sadeghpour, Greene, and Cavagnero; the earliest is that of Hyman, Jacobs, and Burke.

analysis, energies were calibrated to this narrow peak whose energy is well known from experiment [5]. Since the profile is effectively a δ function ($\Gamma \approx 30\ \mu$eV) [11], its observed width demonstrated our energy resolution of 0.007 ± 0.001 eV. For partial decay cross sections, calibration was to the H($N=2$) threshold at 10.953 eV.

The experimental procedure described above can in principle also be used to detect autodetachments that leave the atom in its ground state H(1), if the fourth harmonic of the YAG is used in the second scattering chamber. This method was not practicable, however, because the two interactions

$$H^- + h\nu \rightarrow H(1) + e \tag{4}$$

and

$$H(1) + h\nu \rightarrow H(N') + e \tag{5}$$

can occur in sequence within the second interaction region. The H^- single-photon photodetachment cross section is large near the energy of the second laser. There are many more H^- ions in the beam at the second laser

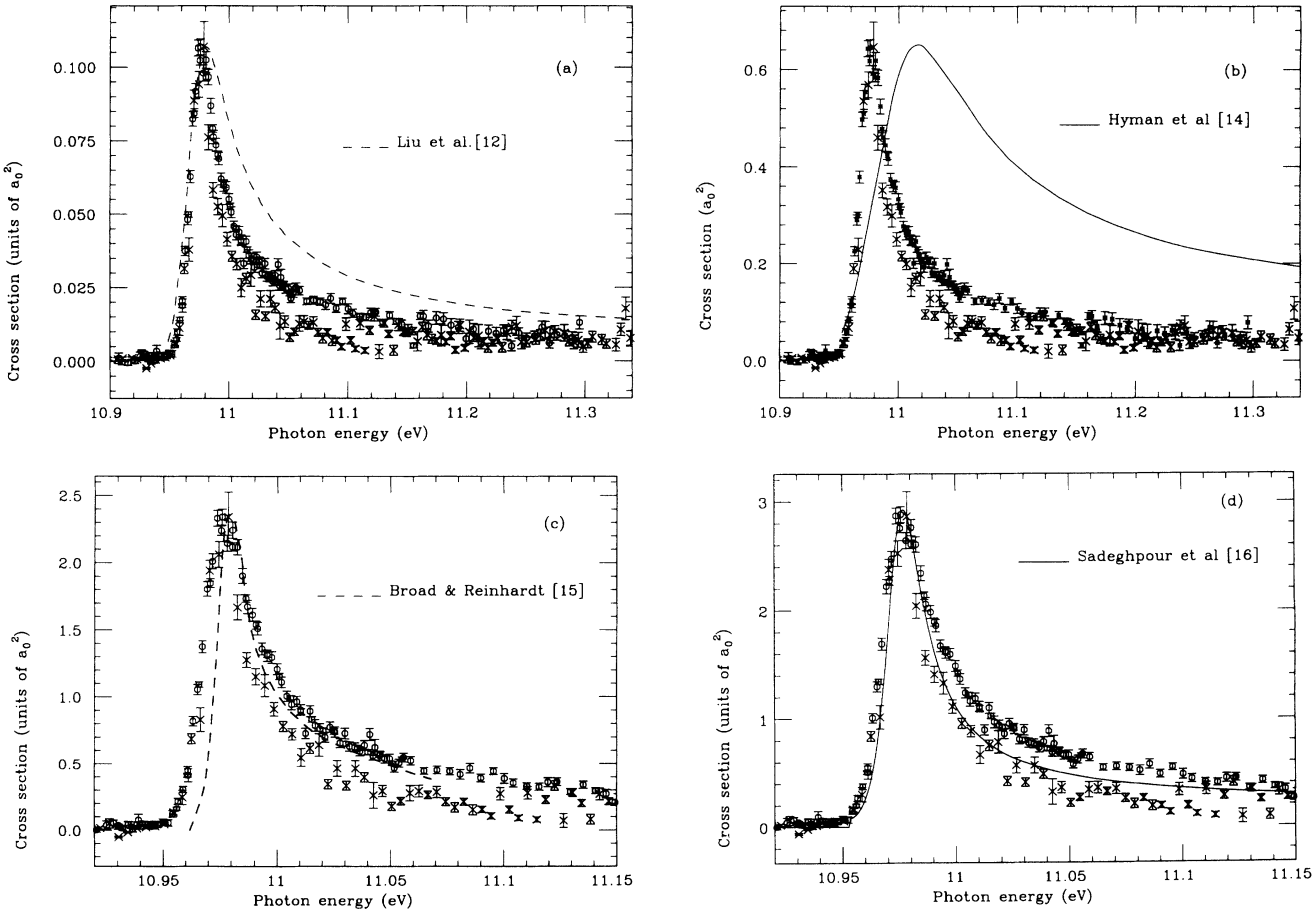

FIG. 4. Partial cross section vs photon energy above the $n=2$ threshold. Open circles represent earlier data [6]. Crosses represent more-recent data [7]. Experimental amplitudes have been normalized to theory. Error bars are statistical only. Shown is a comparison of our data with four theoretical profiles: (a) The broken line is the profile calculated by Liu, Du, and Starace [12]. The theoretical curve has been shifted down in energy by 18.9 meV. (b) The solid line depicts the calculation of Hyman, Jacobs, and Burke [14] with energy shifted down by 20 meV. (c) The broken line is the profile of Broad and Reinhardt [15]. (d) The solid line is the profile of Sadeghpour, Greene, and Cavagnero [16].

than there are H(1) atoms from SR autodetachment. The net result is that far more protons are produced from (4) plus (5) than from decay of the SR, making the measurement of the cross section for $H^-(2) \rightarrow H(1)$ unfeasible.

Our experiments yielded relative cross sections only: These were calculated by normalizing yields to beam current and laser intensity, and then multiplying by $\sin\alpha/(1+\beta\cos\alpha)$. This factor accounts for the changing intensity and beam overlap with change in angle α. Absolute cross sections could not be determined because neither the temporal nor spatial overlap of the beams was accurately established.

III. THE $N=2$ CHANNEL

Although the background was not well known, in the $N=2$ channel the physics supplies the background information, as the cross section must necessarily be zero below the threshold energy for production of H(2). To fulfill this requirement, we subtracted from the data a constant found by fitting to the level below threshold.

Since no two theories have similar predictions for the width, central energy, or amplitude of the resonance (Fig. 3), we show the comparisons with our data in separate figures, normalizing experimental peak amplitudes to theory. In some cases the theoretical energy was shifted downward in order to make profile comparisons.

Figure 4(a) is a plot of our data compared to a profile calculated by Liu, Du, and Starace [12] within an adiabatic hyperspherical representation in which electron correlations are described in terms of the surface harmonics at a constant hyperradius [13]. For this figure the theoretical curve has been shifted down in energy as it predicts the SR 18.9 meV too high.

Figure 4(b) shows a comparison with a $1s$-$2s$-$2p$ close-coupling calculation of Hyman, Jacobs, and Burke [14] using Hyleraas bound-state wave functions. This profile has been shifted downward in energy so that the onset of production appears at 10.953 eV. One of the earliest, this calculation predicted the resonance to be much wider than experiment shows.

In Fig. 4(c) our data are compared with the multichannel J-matrix calculation of Broad and Reinhardt [15]. Their method solves the pseudostate close-coupling equation for H^- photodetachment using standard configuration-interaction methods and square integrable (L^2) basis functions. Their choice of scale parameter $\xi=0.5$ obtains reasonable values for the width and energy of the SR, and no energy adjustment was necessary.

Figure 4(d) is a comparison with a profile resulting from a recent eigenchannel R-matrix calculation which incorporates an analytic description of electron motion in a dipole field [16]. For this figure we have made no shift in energy. To our knowledge, this prediction of Sadeghpour, Greene, and Cavagnero derives from the only *ab initio* calculation for partial and total cross sections to date. The energy and width are in good agreement with experiment.

IV. THE TOTAL CROSS SECTION

In subtracting the background for the total cross-section measurement, we made the assumption that the $N=2$ partial cross section accounts for 75% of the total at a photon energy of 11.1 eV. The theoretical predictions for this ratio are 77% by Broad and Reinhardt, 76% by Sadeghpour, Greene, and Cavagnero, and 72% by Hyman, Jacobs, and Burke. We further assumed that the background was flat.

The total cross section in the region of the SR has previously been measured by this group, compared with theory, and reported elsewhere [4]. Therefore, in this section we focus only on total cross sections from theories which have made predictions for both the total partial decay channels. These are plotted against our data in Fig. 5. The cross section was normalized to theory at the peak amplitude in each case. We note that Hyman, Jacobs, and Burke [14] and Wishart [17] also calculated the total cross section, but we do not display their profiles, as they predicted Γ to be much larger than the other theoretically calculated widths and the experimentally observed width.

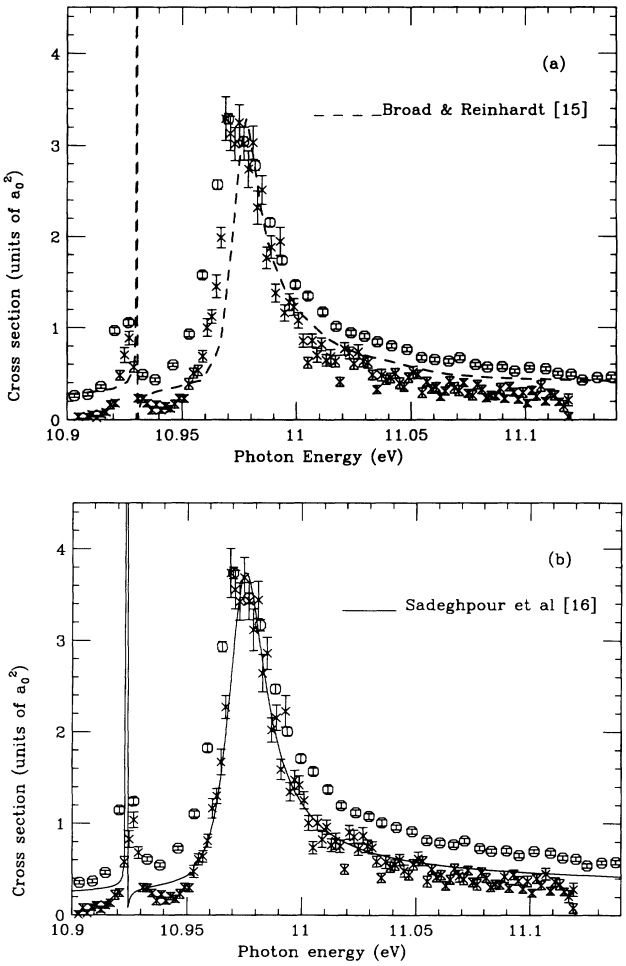

FIG. 5. Photodetachment total cross section vs photon energy near the $n=2$ threshold. Open circles represent earlier data [6]. Crosses represent more-recent data [7]. The narrow Feshbach resonance below threshold is evident near 10.93 eV. Error bars are statistical only. In each figure the experimental peak amplitude has been normalized to theory. (a) The broken line is the profile of Ref. [15]. (b) The solid line is the profile of Ref. [16].

TABLE I. Widths Γ, central energies E_0, asymmetries q, and reduced chi-squared values χ^2/ν obtained from fits to the total and partial sections. *F* indicates a fit to the Fano function. *B* indicates a fit to Broad's modification [20] of the Fano function. *S* indicates a fit to Starace's modification [21] of the Fano function. The error in the parameter value, as assigned by our fitting program MINUIT [19], is the deviation from the best-fit value of the parameter that would increase its χ^2 value by one. All fits were convoluted with a Gaussian function to correct for experimental resolution. The MINUIT error in Γ is combined with that induced by the error in the experimental resolution.

Decay channel	Fit	Γ (meV)	E_0 (eV)	q	χ^2/ν
Total	*F*	21±1[a], 30±1[b]	10.974±0.0003[a], 10.970±0.0003[b]	5.3±0.2[a] 4.5±0.1[b]	2.6[a], 2.5[b]
	B	20±1[a], 33±1[b]	10.974±0.0003[a], 10.970±0.0002[b]	4.6±0.1[a] 4.2±0.1[b]	3.6[a], 4.1[b]
H(2) partial	*F*	20±1[a], 22±1[b]	10.974±0.0003[a], 10.972±0.0001[b]	4.2±0.1[a], 2.7±0.1[b]	5.3[a], 2.6[b]
	B	22±1[a], 22±1[b]	10.973±0.0003, 10.972±0.0001[b]	3.5±0.1[a] 2.7±0.1[b]	4.6[a], 2.7[b]
	S	20±1[a], 22±1[b]	10.974±0.0002[a], 10.972±0.0001[b]	4.3±0.1[a] 2.5±0.1[b]	5.6[a], 3.0[b]

[a]Fits to recent data [7].
[b]Fits to early data [6].

V. FITTING THE DATA

In order to establish the width, central energy, and asymmetry of the SR, we need a function which coincides well with the experimental data. Traditionally, the familiar function formulated by Fano [18],

$$\sigma=\sigma_b+\sigma_a\frac{(q+\epsilon)^2}{1+\epsilon^2}\ , \tag{6}$$

with $\epsilon=2(E-E_0)/\Gamma$, has been used to fit the observed H^- resonant cross sections. The formulation requires, however, that parameters such as width Γ and asymmetry q be constants with respect to energy across the width of the resonance. This may not be the case for the SR.

Other functions have been suggested whose parameters from our MINUIT [19] fits we list in Table I along with the Fano parameters. Broad [20] suggested letting that part of the continuum (σ_a) which interacts with the resonance vary linearly with energy across the range of the fit. Thus we replaced σ_a in (6) with $\sigma_a+\epsilon(\partial\sigma_a/\partial\epsilon)$ with $(\partial\sigma_a/\partial\varepsilon)$ as an extra fitting parameter. The results of fitting to this form are listed in the table as fit *B*.

We also tried the extension of the Fano-type treatment proposed by Starace [21] to fit partial decay cross sections in the vicinity of a resonance. In this approach a new complex variable $\alpha(\mu E)$ is introduced to account for the boundary condition satisfied by the wave function for an outgoing electron in a particular observable channel μ. Our fit using Eq. (26) of Ref. [21] incorporates two new parameters, the imaginary and real parts of α, in addition to the regular Fano parameters. As with the usual Fano function, however, this form is actually meant to be applied to narrower resonances whose parameters have no energy dependence.

In the fits to the total cross section, the original Fano function follows the profile of the data more closely than the function with Broad's energy-dependent background adjustment, as evidenced by the values of the reduced chi-squared χ^2/ν in Table I. For the partial decay cross section, the goodness-of-fit is about the same for the three functional forms we tried. The fit to the partial cross sec-

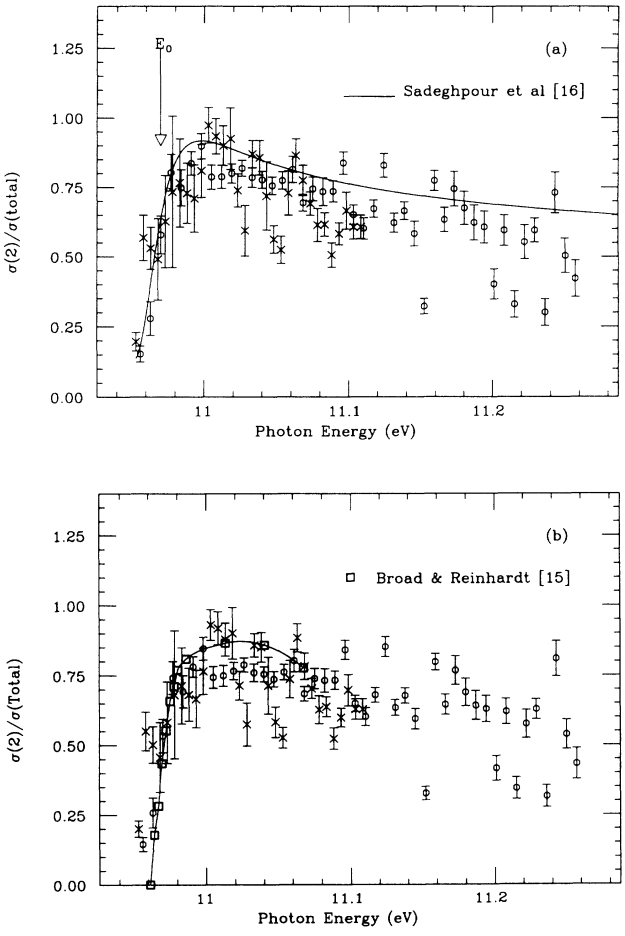

FIG. 6. Branching ratio vs photon energy for the $H^-(n=2)$ shape resonance. The ratio was computed after binning the data into 7-meV bins. Error bars are statistical only. Open circles represent earlier data [6]. Crosses represent more-recent data [7]. (a) The solid line is the branching ratio prediction of Sadeghpour, Greene, and Cavagnero [16]. The arrow points to the approximate central energy position of the shape resonance. (b) The squares, indicating values computed from cross-section predictions of Broad and Reinhardt [15], are joined by a solid line to guide the eye.

tion was better in the earlier work because more data were taken in that year. We also note that the parameters found from fits to the early data show some dependence on the high-energy cutoff value of the fit, while the more recent data do not.

Part of the difficulty in fitting to these functions may be that the low-energy shoulder of the SR lies on the threshold for production of H(2). We therefore tried fitting to a cross-section formula that is a product of the Wigner threshold law and the usual Breit-Wigner resonance formula, as suggested by Peterson [22]. We find that neither the partial nor the total SR cross section is a good fit to this function ($\chi^2/\nu \geq 10$). A variation of width with energy, or the $n=2$ Feshbach resonance lying very near threshold, may explain the poor correspondence of our data with this formulation.

There is an obvious discrepancy between the total widths measured in different years. While stray fields in the laboratory could cause a slight narrowing of the SR (Comtet *et al.* Ref. [4]), we are confident that these did not exceed 1 G. Fields of this magnitude should not be strong enough to cause an observable effect. Comparisons of relative amplitudes and laser intensities have led us to rule out saturation of the reaction as a possible cause for the broader measurement, but other unknown systematic errors must be contributing. We stress, however, that a test of the SR parameters as a function of laser intensity has never been done. We speculate that the intensity may affect the lifetime, which is so short that the resonance decays before leaving the laser field. Multiphoton processes may also be involved. We hope to investigate these ideas in a future experiment.

VI. THE BRANCHING RATIO

The branching ratio we report here, $\sigma(2)/\sigma(\text{total})$, is that fraction of the total SR decaying to $H(N=2)$. We have computed this ratio for 45 photon energies, ranging from 10.95 to 11.30 eV, by binning the data into bins 7 meV wide and dividing by bin. Our data normalized to Ref. [15] give branching ratios only slightly different from those normalized to Ref. [16]. Both cases are shown in Fig. 6. The maximum branching ratio (≈ 0.8) is approximately 20 meV above the central energy of the resonance. Experiment appears to be in good agreement with theory below about 11.1 eV. As discussed in Sec. II, we have less confidence in experimental values above this energy.

VII. SUMMARY

Our cross-section measurements show that, at photon energies between 10.975 and 11.2 eV, the $H^-(n=2)$ shape resonance shows a preference for decay to the H(2) channel. Within this energy range, the branching ratios calculated from our experimental data show good agreement with calculations of Sadeghpour, Greene, and Cavagnero [16] and Broad and Reinhardt [15]. The cross sections normalized to theoretical peak amplitudes also agree well. Various functional forms fit to the data show none clearly superior to the Fano functions for goodness of fit. Discrepancies between cross-section measurements indicate that a careful study of the effect of laser intensity variation is needed.

ACKNOWLEDGMENTS

The authors appreciate the support of the MP-7 staff and technicians at LAMPF. Conversations with John Broad, Chris Greene, David Harmin, Joe Macek, Randall Reeder, Hossein Sadeghpour, and Anthony Starace are gratefully acknowledged. This work was supported by the U.S. Department of Energy.

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