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Photodetachment of the H^- Ion.*

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ABSTRACT

Detachment of electrons from the H^- ion is investigated with an experimental technique whereby an H^- beam moving at a relativistic velocity (2.5×10^{10} cm/sec) is intersected with a fixed frequency laser. The Doppler effect allows systematic variation of the center-of-mass (CM) photon energy over a wide range (factor of 10) by simply adjusting the angle between the ion and laser beams. The focused output from a pulsed, linearly polarized, CO_2 TEA laser operating at 10.6 μm, with peak intensities on the order of 10 GW/cm^2, was used to examine the multiphoton absorption process in H^-. The fourth harmonic (266 nm) of a Nd:YAG laser was used to investigate some of the doubly-excited state resonances in H^-. In the multiphoton absorption work, electron detachment was observed at photon energies where as few as 2 and as many as 8 photons are required to get above the 1-electron detachment threshold (EDT) of H^- (0.754 eV). Electron yield vs photon energy plots exhibit structure that is laser intensity dependent. Electron yield vs laser pulse energy data was obtained at a few selected CM wavelengths and laser pulse energies. In the single-photon uv laser work, numerous resonances within the H^- photodetachment continuum corresponding to one-photon two-electron excitation processes were observed. The doubly-excited resonances appear to be of the Feshbach type. A simple, semi-empirical recursion formula predicts the resonance energy levels. The experimental techniques described here can be used to accurately determine accelerator beam and ion source parameters such as beam energy, energy-spread, and ion density spatial distribution.

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1. Introduction

The H^- ion is one of the simplest three-body systems available for experimental and theoretical investigations of a variety of photo-physical processes of great current interest, including electron detachment induced by multi-photon absorption, and one-photon absorption transitions where two atomic electrons are simultaneously excited. It has only one bound state: the ground state. All electronic transitions in H^- are autoionizing.[1]

H^- is a very attractive candidate for multiphoton absorption studies for several reasons. It has no electronic states below the one-electron detachment threshold (EDT). The lack of internal structure greatly simplifies the theoretical description of the absorption process, as the there are no effects due to intermediate state resonances to be concerned about. Detailed quantitative comparisons between experiment and theoretical models should be possible. Since the EDT is only 0.754 eV, low frequency lasers can be used to induce electron detachment. Because of the low frequency required, the effects due to ponderomotive forces on the absorption physics can be significantly enhanced. Commercially available CO_2 lasers can produce laser intensities that lead to ponderomotive potential energies larger than the energy of the laser photons or the EDT. Thus, it should be possible to obtain data in a laser intensity regime where tunneling effects can be important. Finally, the departing electron is subject to a short range potential since it leaves behind a neutral atom. Processes such as above-threshold-ionization (ATI) and harmonic generation are likely to be significantly different when compared to cases involving ionization of neutral atoms where a long-range Coulomb force is at work.

H^- is also a good candidate for investigations concerned with physical processes in which electron correlations are important. Simultaneous excitation of two electrons with a single photon is one example. A proper treatment of electron correlation effects in H^- is extremely important for an accurate description of doubly-excited state resonances as well as the ground state.[1] The resonances in H^- can be thought of as arising from the polarization induced in an electronically excited hydrogen atom by a second, outer, electron.

The experimental technique described below that is used to investigate photodetachment physics in H^- is basically a crossed laser/ion beam approach, except that the velocity of the ion beam is relativistic; 0.84 times the speed of light. One of the principal advantages of this approach is that the Doppler effect allows systematic and carefully controlled variation of the center-of-mass (CM) photon energy over a wide range (about a factor of 10) by simply changing the angle of intersection between the fixed-frequency laser beam and the ion beam. Another experimental feature worthy of note is that the transit time for an ion passing through a tightly focused, short-wavelength laser beam can, in principle, be made less than 10^{-15} s, so that photoabsorption physics in which the ion is exposed to only a few optical cycles can be investigated.

2. Relativistic Kinematics

The photon energy, E, in the H^- frame (moving) is given in terms of the laboratory laser photon energy (E_L) by

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

where α is the lab angle between the laser and ion beams such that $\alpha = 0$ when the laser propagates in a direction opposite to that of the H^- beam. γ and $\beta = v/c$ are the usual relativistic parameters. The energy resolution that any given experiment achieves can be estimated from this equation by carrying out a Taylor series expansion. It can be shown that for $\cos \alpha = -\beta$, broadening due to variations in β (longitudinal beam velocity or beam energy) vanish to first order (Doppler free angle). Similarly, for $\cos \alpha = \pm 1$, broadening due to variations in α (due to H^- beam and laser divergence) vanish to first order.

The laser intensity in the laboratory frame, I_L, transforms as [2]

$$I = I_L \gamma^2 (1 + \beta \cos \alpha)^2.$$

Because of the manner in which photon energy and laser intensity transform into the moving frame, the ponderomotive energy (quiver energy), which is proportional to laser intensity divided by the square of the laser frequency, does not depend on the photon/ion angle α.

3. Experimental Description

The experiments were carried out at the Los Alamos Meson Physics Facility (LAMPF) using an H^- beam at 800 MeV ($\beta = 0.842$, $\gamma = 1.852$). The temporal structure of the H^- beam consists of "macropulses" that can be as long as 1 ms, occurring at pulse repetition rates as high as 120 Hz. Each macropulse contains a train of "micropulses" which are approximately 0.25 ns wide, spaced 5 ns apart. The experimental configuration used to investigate multiphoton absorption[3] and doubly-excited states resonances[4] in

H^- have been described in detail previously, and is shown schematically in Figs. 1 and 2. Briefly, a laser is made to intersect the H^- beam within a vacuum interaction chamber that contains an optical turntable assembly that allows the laser/ion beam angle to be varied accurately over a wide range of angles. The H^- beam was approximately 3 mm in diameter.

In the multiphoton absorption studies (see Fig. 1), a ZnSe lens with a focal length of 25.4 cm is used to focus a near-diffraction limited CO_2 laser pulse into the H^- beam. Electrons (at about 435 keV in the lab) and hydrogen atoms that are produced via multiphoton absorption are detected downstream of the laser interaction region with scintillator/PMT assemblies which allow near 100% detection efficiency. The focal spot diameter was measured to be approximately 0.11 mm (FWHM), with peak intensities on the order of 10 GW/cm^2 (lab frame). The CO_2 TEA laser system was an oscillator/amplifier design. Within the oscillator, an iris and low-pressure gain cell were used to generate a linearly-polarized, temporally smooth TEM_{00} beam with a maximum pulse energy of about 1 Joule. Approximately half the energy was contained within the initial pulse (130 ns FWHM), which was followed by a low intensity tail. Most of the measurements were taken with the laser operating on the 10.6 μm line (0.117 eV), although some data has been taken with the 9.6 μm line (0.129 eV). Pulse repetition rates were limited to 0.5 Hz. The laser/ion beam intersection angle α was varied from 18° to 162°, which corresponds to photon energies ranging from 0.39 to 0.043 eV. Laser polarization is maintained perpendicular to the H^- beam throughout the scan. The energy resolution in these measurements is dominated by the spread in angles introduced by the focusing lens. The resolution was calculated to be 1.1 meV at 20°, increasing to 3.6 meV at 90°, and decreasing down to 0.4 meV at 162°. The ponderomotive energy (invariant with angle) at the peak of the laser pulse was estimated to be approximately 0.1 eV for the highest laser pulse energies used. At 90°, the H^- transit time through the laser focus is about 240 fs.

In the doubly-excited H^- state resonances (see Fig. 2), the fourth harmonic (266 nm = 4.66 eV) of a Nd:YAG laser is used to excite H^- in regions where doubly-excited autoionizing resonances are likely to be found. The experimental procedure is roughly as follows. The laser is Doppler tuned to excite the transition

$$H^- + \text{photon} \longrightarrow H^{0*}(\leq n) + e^-$$

where the principal quantum number n of the Hydrogen atom is typically 4-8. As the photon energy is scanned upwards, successively higher n states can be produced. Should an H^- resonance occur somewhere along the way, a disturbance in the production of a given $H^0(n)$ would be expected. To be able to monitor specific $H^0(n)$ states, a variable field magnet is used to field ionize $H^0(n)$ atoms into protons that can be detected with a scintillator/PMT assembly downstream. The magnet field is adjusted so that, for example, it can ionize all H^0 with $n \geq 4$, but leaving all $n \leq 3$ unaffected. When the photon energy reaches the threshold for production of $H^0(n=4)$ from H^-, a sudden increase in protons would be observed. When the photon energy reaches the minimum energy required to make $H^0(n=5)$, another sudden increase in the proton signal will occur. In between these sudden increases in proton signal, H^- resonances that affect the production of the states being detected can be observed. By adjusting the field in the ionizing magnet, individual $H^0(n)$ channels can be isolated.

4. H^- Multiphoton Absorption

The Doppler tuning capability is an important feature of this experiment. Scanning the CM-photon energy from 0.39 to 0.043 eV allows one to carefully examine and compare multiphoton absorption events where as few as two and as many as 18 photons are required to cause electron detachment. To date, definite signals have been observed at photon energies where a minimum of 2-8 photons are required to reach the EDT. Some of the preliminary results have been described elsewhere.[3] An example of the total electron yield as a function of CM photon energy is shown in Fig. 3. The region between 0.25 and 0.38 eV corresponds to the 3-photon absorption region, where it takes a minimum of 3 photons to get above the EDT. The rapid increase in count rate in the neighborhood of 0.38 eV indicates a transition into two photon absorption. The structure observed is laser intensity dependent. In general, the electron yield is found to increase rapidly as the photon energy increases past the point where a new, lower order of photon absorption can take place (i.e. as N —> N-1). The contrast between orders is highest at low laser intensity. To examine the extent to which the EDT might be shifted by ponderomotive force effects, the transition region between 2 and 3 photon absorption, which is fairly well defined at low laser intensity, was carefully examined. By fitting the data to a Wigner threshold law form, and taking into account the experimental resolution in this photon energy range, we find that there does appear to be a small but observable increase in the EDT. The magnitude of the shift at 2.8 GW/cm^2 is $\approx$ 8 meV, smaller than the ponderomotive potential that would be experienced at the peak of the laser pulse ($\approx$ 31 meV). However, it should be kept in mind that the electron yields measured here represent an unavoidable average over a range of laser intensities due to the spatial and temporal characteristics of the laser beam. Preliminary measurements of the intensity dependence of the electron yield vs laser pulse energy were carried out at selected photon energies for 2,3,4,5 photon absorption. The results were analyzed in terms of a power law of the form: electron signal $\propto \sigma_s I^s$, where σ_s is a generalized multiphoton absorption cross section. In general, least-squares fits yield non-integer values for the exponent s, typically 0.5 less than the expected from perturbation theory. This result suggests that σ_s itself is a function of laser intensity, possibly due (in part) to intensity dependent shifts of the EDT.

5. H^- Doubly-Excited Resonances

Fig. 4 shows some of the H^- doubly-excited resonances observed in the photodetachment continuum between the hydrogenic n=4 and n=5 thresholds (at ~13.5 and 13.8 eV, respectively) that appear as structure in the partial production cross section of H^0(n=4). The solid line is a fit to a set of three Fano lineshape profiles. It is clear that the effect of the H^- resonance is to interfere destructively with the continuum production of H^0(n=4). Similar resonance "dips" were observed in the 5 to 6, 6 to 7, and 7 to 8 hydrogenic threshold regions. In general, the positions of the resonance dips were found to be described reasonably well by a simple, semi-empirical recursion formula for the energy levels of the doubly excited states in H^-[1]:

$$E(n,m) = E_t - R/n^2 + 2R\,[1/2n^2 - 0.696/(n + 0.333)^2]\,\exp[-2\pi(m-n)/\alpha_n]$$

where n is the principal quantum number of the hydrogenic threshold in question, m = n, n+1, n+2, ..., α_n is a dipole parameter associated with channel n, R is the Rydberg constant and E_t is the threshold energy for production of the hydrogenic level n. The doubly excited states observed here appear to be a series of Feshbach-type resonances which are due entirely to the effects of electron correlations.[1]

6. Accelerator Beam Characterization

The laser techniques developed here to address some fundamental atomic physics issues related to the H^- ion have practical applications in the area of accelerator beam characterization. A couple of examples will be discussed in the material that follows.

We have already demonstrated that a the focused output of a CO_2 TEA laser can induce electron detachment in H^- via multiphoton absorption. Since the simultaneous absorption of many photons is a highly non-linear function of laser intensity, electron production can be confined to occur primarily within the focal volume of the laser pulse where the intensity is highest, the electron signal can be used to obtain spatially and temporally resolved information concerning the density of H^- ions. By scanning the laser focal spot around in space one ought to be able to map out the spatial and temporal distribution of H^- in an accelerator beam or perhaps an ion source.[5]

The resonance structures that exist in the electronic excitation spectrum of H^- can also be exploited to characterize the properties of an accelerator beam. Consider, for example, what is commonly known as the n=2 Feshbach resonance. It occurs at a CM photon energy of 10.9264 eV and is predicted, from theoretical calculations, to be as narrow as 30×10^{-6} eV. Excitation of the n=2 Feshbach resonance results in the production of a free electron and a hydrogen atom in the ground state (closed channel). This narrow resonance can be used to measure fairly accurately the H^- beam energy and energy-spread distribution function as follows. Recall from the Doppler shift formula (see discussion above) that the energy resolution observed in any particular experiment is a function of the uncertainties in the experimental parameters such as H^- and laser beam divergence, beam energy spread, laser frequency spread and atomic transition frequency spread. The relative contributions of the terms is a function angle. As pointed out previously, for very shallow angles, where a is close to 0 or 180°, first order contributions to the observed linewidth from uncertainties in the angle between ions and photons vanishes. Thus, given that the atomic transition is very narrow, the width of the resonance is likely to be dominated by beam energy spread effects. Thus, a careful measurement of the n=2 Feshbach resonance position and lineshape will yield precise and detailed information regarding the accelerator beam energy and its variations.[6] During the course of the atomic physics experiments described above, we were able to demonstrate proof-of-principle of this technique.[7] A "momentum buncher" device installed in the LAMPF linac was used to cause variations in the beam energy and energy spread. The variations were monitored by making angular scans of the n=2 Feshbach resonance using the fourth harmonic of a Nd:YAG laser (266 nm). Liberated electrons were swept out of the main beam with a spectrometer magnet assembly and detected with a scintillator/PMT combination. Examples of some of the observed resonance lineshapes are shown in Fig. 5. It is clear that even with the current experimental setup, which is far from being optimum, the changes in the beam energy can be easily monitored.

In a similar manner, if the interest were to monitor say, the transverse divergence of the H^- beam, then by working close to the Doppler free angle ($\cos \alpha = -\beta$), where first order contributions to resonance width from beam energy variations vanish, an accurate measurement of the angle between the laser and ion beams, and its variations, can be made. Precise knowledge of the direction in which the H^- beam is moving (relative to the laser) and its transverse divergence can be obtained. This would require that the laser beam divergence be made significantly smaller that the ion beam divergence. In practice, attempting the H^- beam divergence measurement with the n=2 Feshbach resonance would require a vacuum uv laser source for a relativistic beam, which would be fairly inconvenient. However, if it can be arranged to neutralize a fraction of the H^- beam to produce electronically excited $H^0(n,l,m)$ atoms (with a thin foil or laser photolysis source), then the beam direction and divergence measurement could be easily carried out by causing a transition within the H^0 manifold of states, which are numerous, very narrow in width, and lie in spectroscopically accessible regions. In fact, the angle of the rotating turntable that is used in the experiments described above is routinely calibrated by checking the angular position at which H^0 (n = 2, 3, 4, 5) —> H^0 (n = 11, 12, 13, ...) transitions occur.

7. Conclusion

Using a laser to examine photoabsorption in H^- ions moving at relativistic velocities offers unique and novel ways of examining physics problems of great current interest. The experimental techniques described here have numerous practical applications in accelerator beam characterization and can be applied to investigations involving relativistic heavy ions.

Acknowledgements

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References

[1] For a concise discussion of the spectroscopy of the H^- ion the reader is referred to: "Observation of High-Lying Resonances in the H^- Ion" by P. G. Harris, doctoral dissertation, Los Alamos manuscript LA-11843-T, 1990. This source contains an extensive list of references.

[2] See, for example, L. D. Landau and E. M. Lifshitz, The Classical Theory of Fields, 1st Ed., Addison-Wesley, Reading, MA, 1951, p. 122.

[3] C. Y. Tang, et al., Phys. Rev **A 39** (1989) 6068, W. W. Smith, et al., J. Opt. Soc. Am. B, to be published (1991).

[4] P. G. Harris, et al., Phys. Rev. Letters **65** (1990) 309, and ref. [1] as well.

[5] C. R. Quick, "A Spatially- and Time-Resolved Laser Diagnostic for H- Beam and Ion Source Characterization", Los Alamos document, LA-UR-89-726, 1989.

[6] D. B. Holtkamp and C. R. Quick, Jr., Nucl. Instr. and Meth . **A 287** (1990) 348.

[7] P. G. Harris, et al., Nucl. Instr. and Meth. **A 292** (1990) 254.

Figure Captions

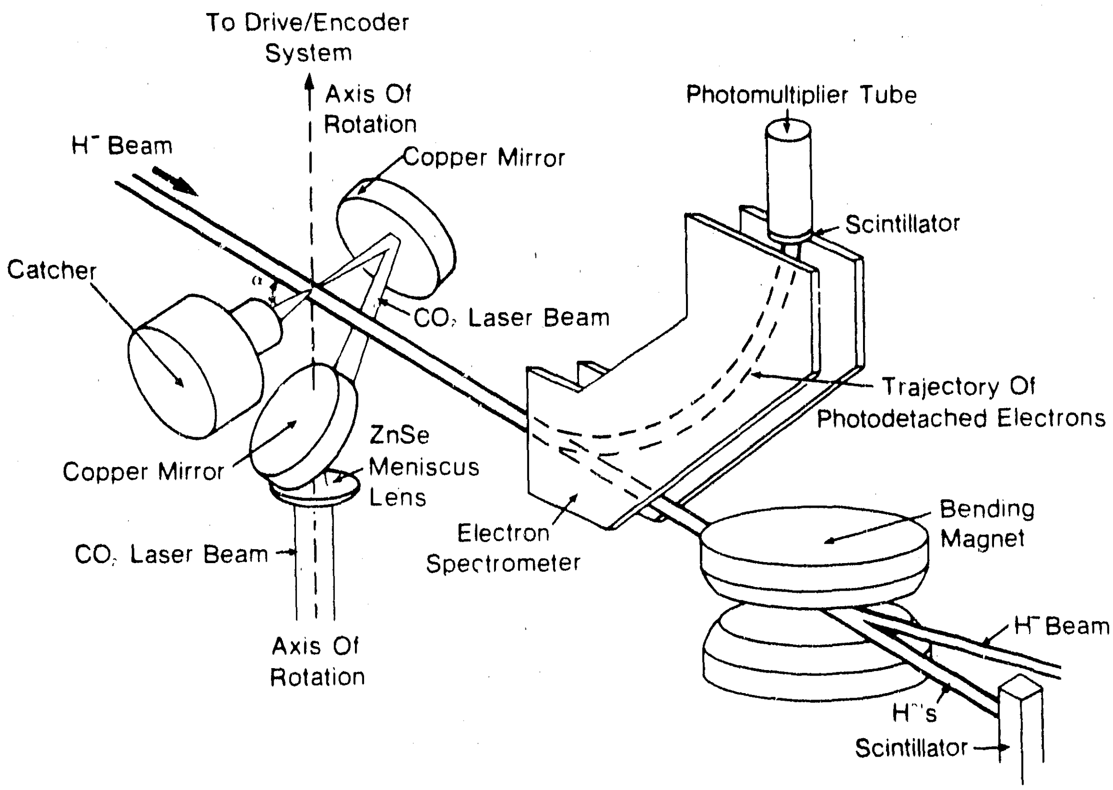
Fig. 1 Experimental arrangement used for H^- multiphoton absorption studies.

Fig. 2 Experimental arrangement used for the investigation of high-lying doubly-excited resonances in H^-.

Fig. 3 Example of total electron yield as a function of CM photon energy in the 2 and 3 photon absorption region, at a peak laser irradiance of 5 GW/cm^2. The error bars denote the statistical fluctuation in the data only. The solid line is a guide to the eye. The data is uncorrected for variations in laser irradiance with angle.

Fig. 4 Example of the resonances observed in the continuum between the hydrogenic n = 4 and n = 5 thresholds. The dips indicate a decrease in the production of H^0 (n = 4). The solid line is a fit to a set of 3 Fano lineshape profiles.

Fig. 5 Angular scans of the n = 2 Feshbach resonance using a 266 nm laser: a) typical Feshbach resonance profile without momentum bunching, b) with momentum bunching, and c) with bunching linac cavity out-of-phase.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H⁻ Beam
Copper Mirror
Scintillator
Catcher
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CO₂ Laser Beam
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H⁰'s
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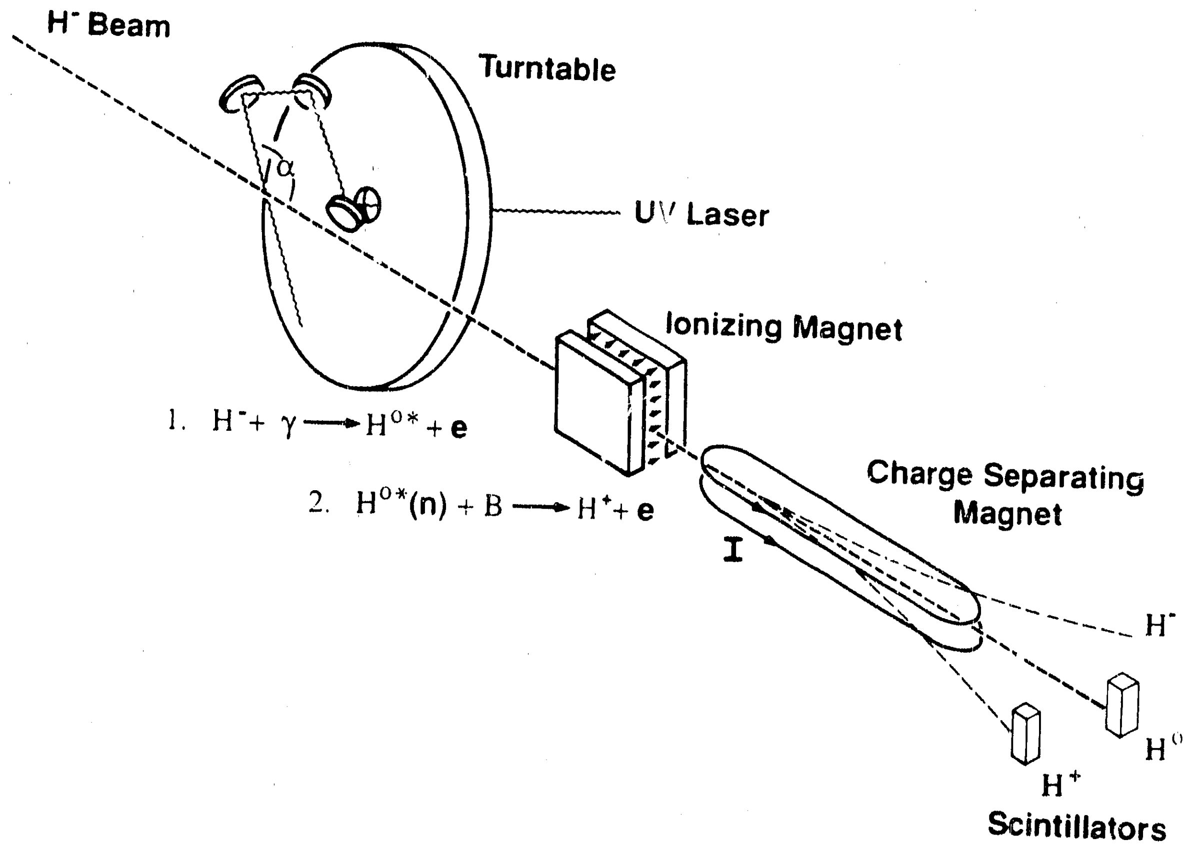
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Turntable
α
UV Laser
Ionizing Magnet
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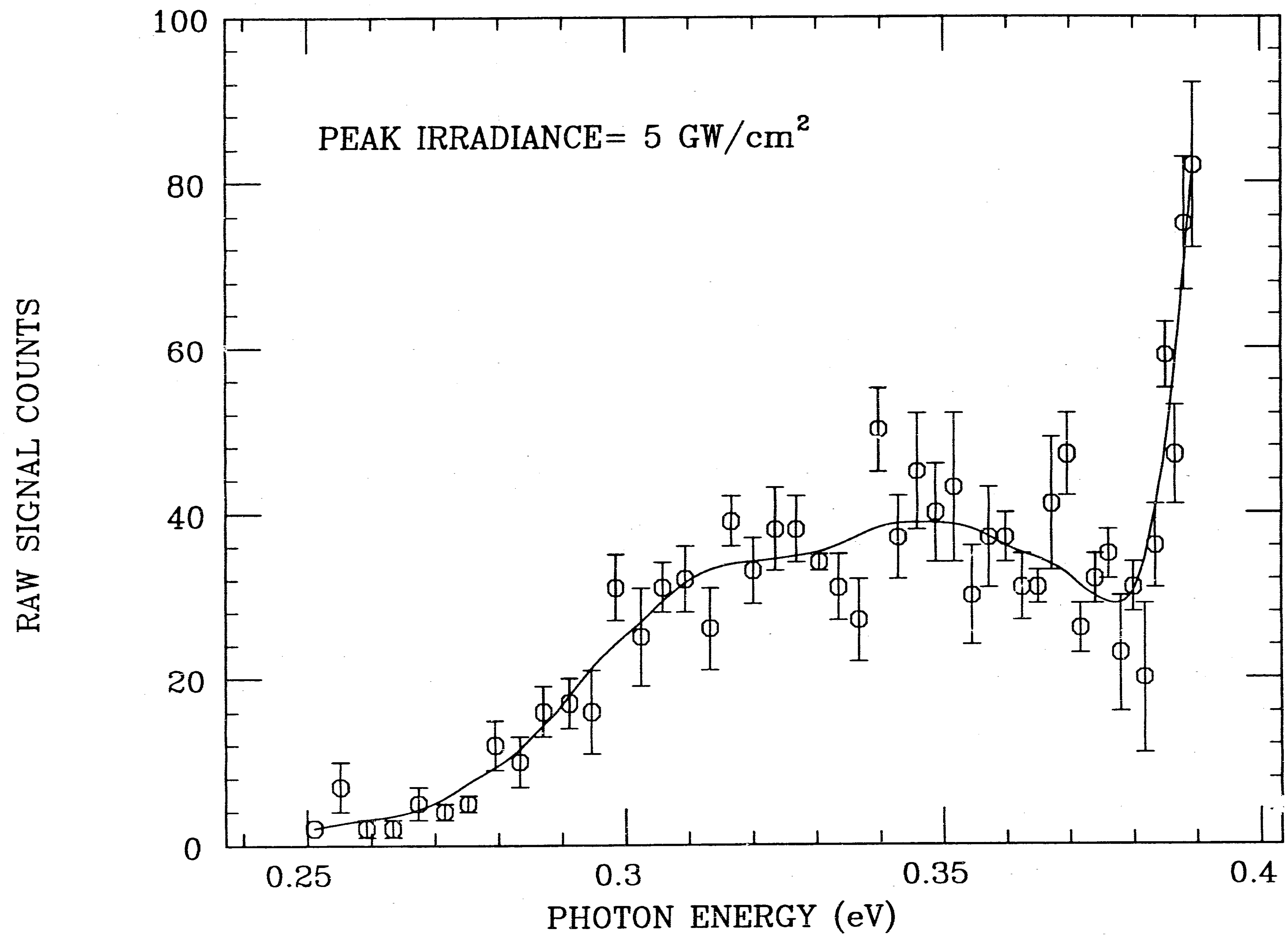
I
H^-
H^0
H^+
Scintillators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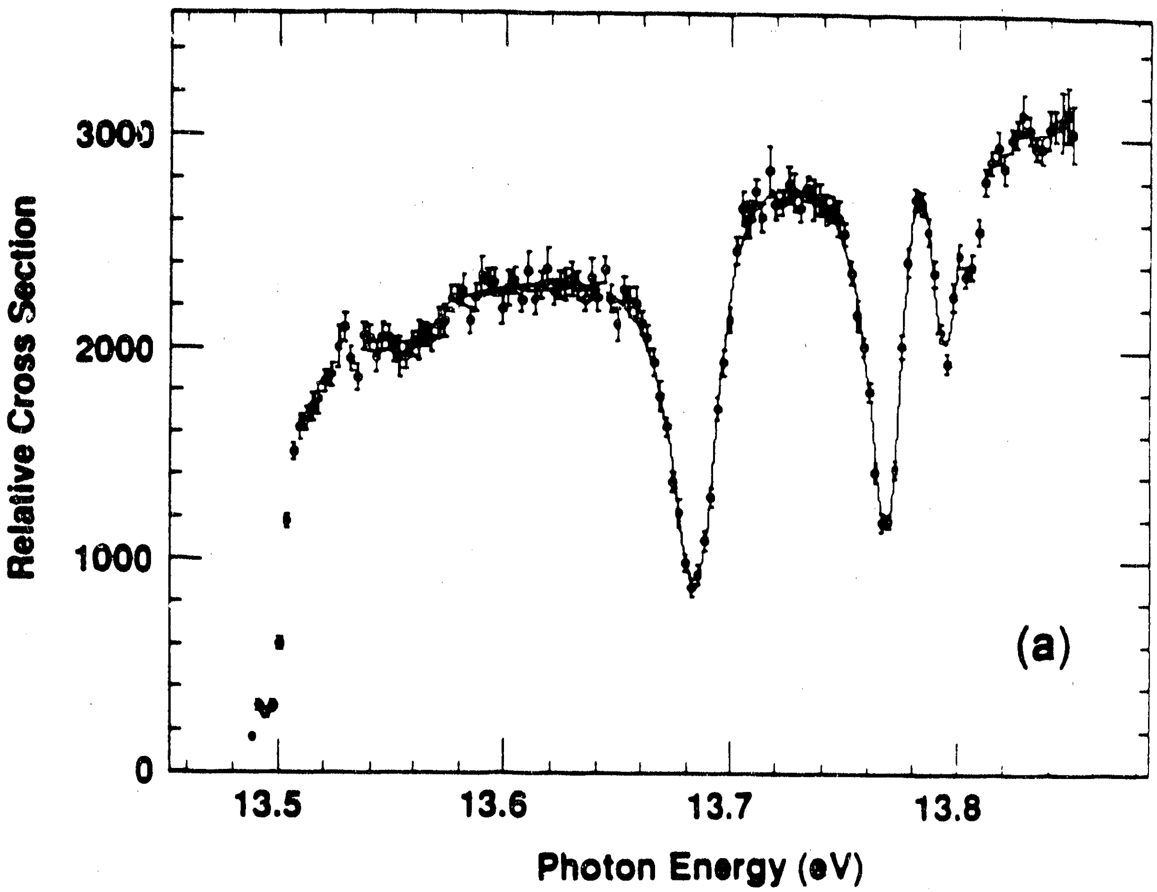
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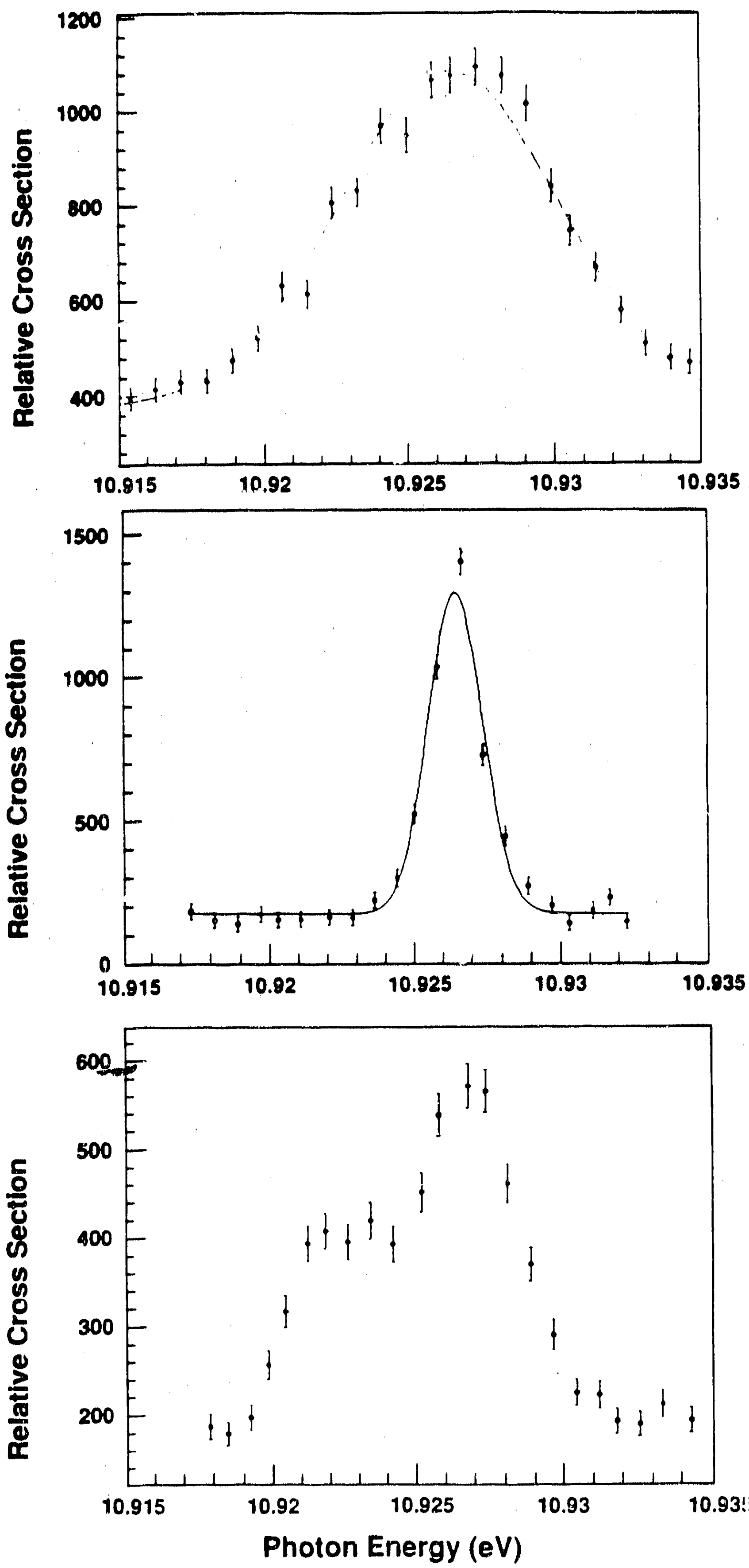
13.7
13.8
Photon Energy (eV)
(a)



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800
600
400
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10.93
10.935
1500
1000
500
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Relative Cross Section
10.915
10.92
10.925
10.93
10.935
600
500
400
300
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Relative Cross Section
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Photon Energy (eV)

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