Threshold Structures in the Multiphoton Detachment Yield from the H^- Ion

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Multiphoton detachment from H^- has been studied using linearly polarized light for photon energies from 0.15 to 0.39 eV. Characteristic threshold structures were observed. The electron detachment rates are estimated at peak laser intensities from 2 to 12 GW/cm^2 in the laboratory frame. The two-photon detachment data reveal intensity-dependent shifts of the H^- electron detachment threshold energy.

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Multiphoton detachment (MPD) from H^- provides a window on fundamental multiphoton physics.[1] Structures in the continuum[2] far above the detachment threshold need not be considered for moderate laser fields. Because high-power lasers have had limited wavelength tunability, with the exception of the measurements using relativistic Doppler shifts, the general behavior of the MPD rate (cross section) versus photon energy has been explored only in theory.[3]

Comparisons between theory and experiment involving multiphoton ionization (MPI) of neutral atoms are usually complicated by intensity-induced shifts, resonances, and the laser intensity distribution. In addition, subtle gauge problems[4] call for experimental data. Thus, it is desirable to study a simple system like H^-, using a tunable laser, to obtain an unobstructed view of MPI processes.

The first observation of MPD in H^- was reported recently.[5] However,[6] difficulties precluded detailed data analysis. Here, we report new observations of MPD of H^- in moderate laser fields. These data reveal clear threshold phenomena occurring near photon energies where a transition from one order of absorption to another can take place (e.g., N-photon absorption to $N+1$). Double-electron detachment from H^- was not observed. Briefly,[5,6] an 800-MeV beam of H^-, at the Los Alamos Clinton P. Anderson Meson Physics Facility (LAMPF), was made to intersect with a focused CO_2 laser beam. The H^- beam, roughly 3 mm in diameter, consisted of 0.25-ns micropulses, with a separation of 5 ns. A transversely excited atmospheric CO_2 oscillator-amplifier, including a low-pressure gain cell in the oscillator, was operated in the TEM_{00}, single-axial mode with wavelength of 10.6 μm. Linearly polarized, temporally smooth laser pulses had a duration of $\sim$130 ns, full width at half maximum (FWHM), and a 1-μs long tail. The measured intensity distribution in the focal region of a 25.4-cm focal-length lens demonstrated that the beam was nearly diffraction limited. The beam diameter at the focus was 110 ± 5 μm (FWHM), corresponding to a transit time of 0.24 ps in the ion's frame when the beams cross at 90°, and a Rayleigh range of 2.6 mm. The highest peak laser intensity in the experiment was estimated to be 12 GW/cm^2 ($\pm$10%) in the laboratory frame. The photon energy (E_{lab}) was Doppler shifted to $E_{c.m.}$ in the center-of-mass (c.m.) frame of the H^- ion according to

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

where α is the angle of intersection ($\alpha=0$ when head on) and β (0.842) and γ (1.85) are the usual relativistic parameters. The photon-energy spread in the ion's frame is mainly due to the angular divergence of the focused beam, $\Delta\alpha\cong40$ mrad (FWHM), leading to an energy spread $\Delta E_{c.m.}$ ranging from 2.2 ($\alpha\sim20°$) to 7.2 meV ($\alpha\sim90°$). The laser intensity in the c.m. frame ($I_{c.m.}$) transforms from the laboratory value I_{lab} as[5]

$$I_{c.m.}=\gamma^2(1+\beta\cos\alpha)^2 I_{lab}\,; \tag{2}$$

however, the ponderomotive energy, $E_p \propto I_{c.m.}/E^2_{c.m.}$, does not vary with α. The photodetached electrons (or H^0's) are detected with 100% efficiency using a scintillator-photomultiplier combination.

The nominal threshold photon energies are submultiples of the electron affinity (0.754 eV) of H^-. Even though a given photon energy range may be labeled as belonging to, for example, the N-photon region, the total electron yield can, in general, contain contributions from higher-order processes (i.e., $N+1, N+2,\ldots$) depending on the laser intensity. Clear thresholds around the nominal energies for the onset of the two-, three-, four-, and five-photon MPD of H^- are shown in Figs. 1(a)-1(c), where the peak laser intensity (in the laboratory frame) is approximately 12, 6, and 4 GW/cm^2, respectively. These data show the relative signal measured at each photon energy without normalization to the angle-

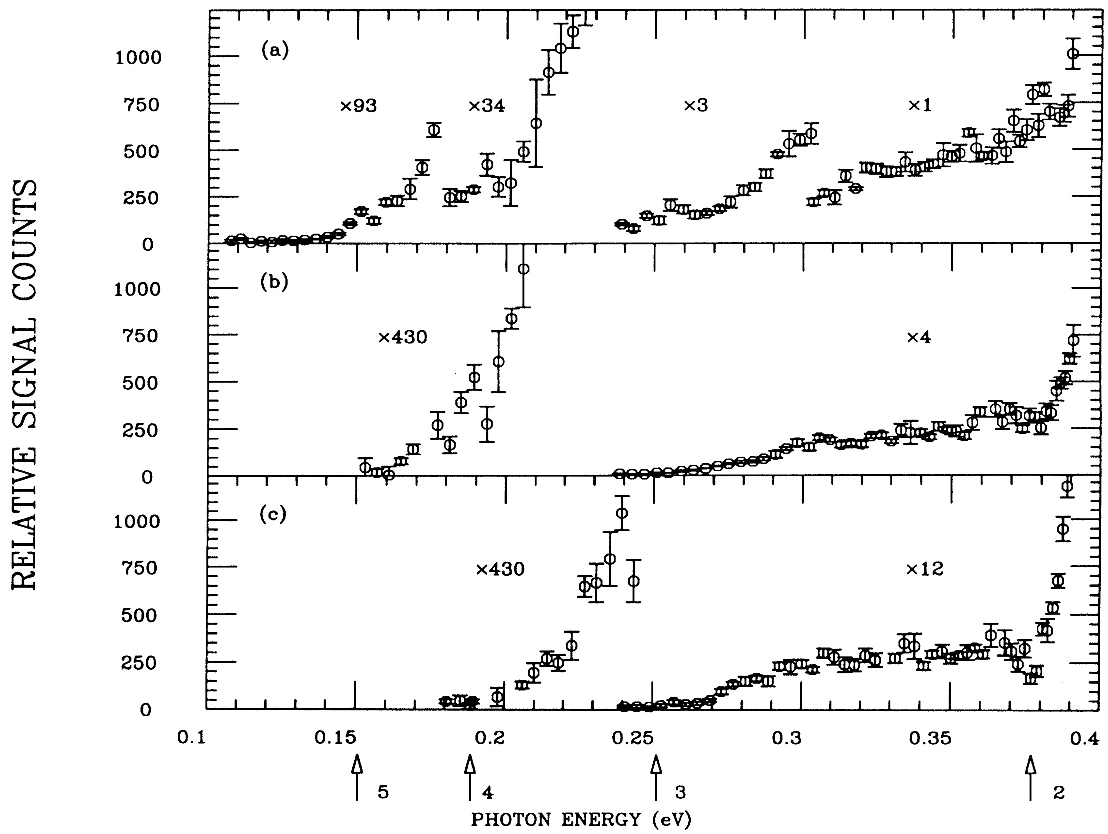

FIG. 1. Dependence of the MPD yield from H^- on photon energy for (a) $I_{\mathrm{peak}}=12$ GW/cm^2, (b) $I_{\mathrm{peak}}=6$ GW/cm^2, and (c) $I_{\mathrm{peak}}=4$ GW/cm^2. The multiplication factors indicate the magnification in the corresponding signal counts, and the arrows indicate the nominal photon energy required for the onset of N-photon absorption.

dependent interaction time. The thresholds, clearer at lower laser intensities, can be seen as rapid changes in the relative signal counts with photon energy.

In a distribution of laser intensity, the H^- ions decay in a complicated manner. Simplistic estimates of the average electron detachment rate ($\bar{\Gamma}$) are obtained as follows:

$$\bar{\Gamma} \sim \frac{N_s}{(\text{interaction time})N_0}, \qquad (3)$$

where N_0, the total number of H^- going through laser focus [roughly 10^5/(laser pulse)], and N_s, the absolute signal counts, have a relative uncertainty of about 30% mainly due to the instantaneous H^- beam fluctuation within the interaction volume. No attempt is made to correct the transit time for the effective compression of the pulse width due to higher orders of interaction,[7] and the interaction time is simply taken as that required for H^- to traverse the laser beam (FWHM), ignoring nonuniform beam-overlap problems. Taking into account the possible systematic uncertainties, $\pm 50\%$ in the estimate of N_0, an effective compression factor of roughly $1/\sqrt{N}$ in the interaction time for an N-photon process, the results shown in Fig. 2 should be in agreement with more accurate detachment rates within a factor of 5 or so. The positions of the dips are generally not good indicators of the thresholds, because near a threshold comparable contributions to the detachment rate from adjacent orders can obscure where one order ends and another

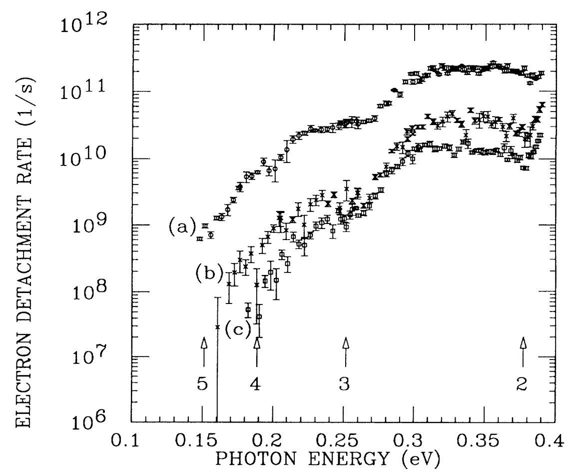

FIG. 2. Intensity-averaged electron detachment rates for the three cases in Fig. 1.

TABLE I. Results of power-law fitting, $\bar{\Gamma} \propto I_{\text{peak}}^{s}$.

Photon energy (eV)	Number of photons	Fitted exponent (s)
0.390	2	1.48 ± 0.02
0.335	3	2.7 ± 0.1
0.242	4	3.4 ± 0.1
0.172	5	4.3 ± 0.1

starts.

The detachment rates, in general, do not follow the power law $\bar{\Gamma} \propto I_{\text{peak}}^{N}$, where N is the minimum number of photons required for MPD. In contrast, Table I displays the observed effective exponent s for selected photon energies.[8] It is unlikely that saturation of the MPD rate due to depletion of the interaction H^- ions occurs. Even in the five-photon range, where detachment probabilities are 0.2% or less, the deviation of s from N still exists. More likely, intensity-dependent variations in the detachment rate due to the ponderomotive shift in the detachment threshold are responsible.[9]

When the laser power is so low, $I_{\text{peak}} \simeq 2$ GW/cm^2, that higher-order signals below the two-photon threshold are suppressed, a clear transition from three- to two-photon detachment can be seen (Fig. 3). A least-squares fit by the Wigner threshold law[10] yields a shift ΔE_0 of 9 ± 1 meV, significantly smaller than the ponderomotive energy (20 meV) expected from the peak laser intensity. This result can be attributed to the variation of laser intensities, and consequently ΔE_0's.

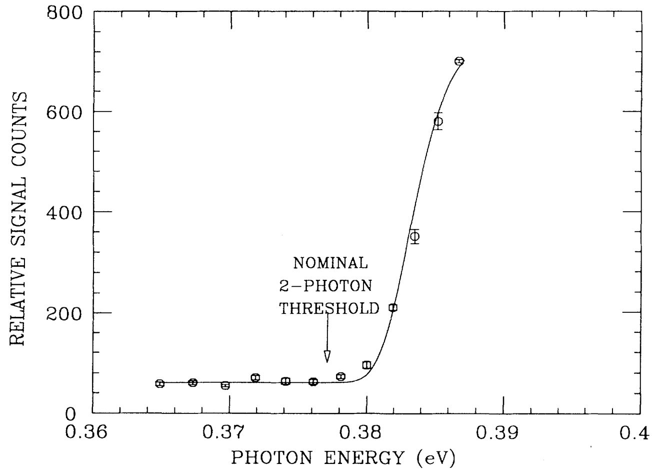

FIG. 3. A fit, for $I_{\text{peak}} = 2$ GW/cm^2, by the Wigner threshold law, $Y = [A(E-E_2)^{1/2} + B] \otimes \delta E$, where A is a constant, E_2 is the field-dependent two-photon threshold, B is a constant approximating contributions from higher orders, $\otimes$ is the convolution operation, and δE is the photon energy spread, assuming a Gaussian distribution with varying widths with α.

To illustrate the complexities of threshold shifts, we have calculated H^- MPD rates (Γ) near the two-photon threshold ($E_{\text{c.m.}} = 0.377$ eV) using an approximate formula[11] for MPD in a zero-range potential with linearly polarized light:

$$\Gamma \simeq \text{Re}\left\{\omega \left[\frac{4(\epsilon+z)}{i\pi}\right]^{1/2} \int_0^\infty \frac{d\theta}{\theta^{3/2}} e^{i(\epsilon+z)\theta}[e^{iu(\theta)} J_0(z\sin\theta - u(\theta)) - 1]\right\}, \tag{4}$$

where $\epsilon = I_0/E_{\text{c.m.}}$, $z = E_p/E_{\text{c.m.}}$, $u(\theta) = 2z(1-\cos\theta)/\theta$, and J_0 is the zeroth-order Bessel function. Some numerical results are shown as the solid curves in Fig. 4, where a ponderomotive energy (E_p) of 0.01 eV corresponds to I_{lab} of 1 GW/cm^2. We note that (a) the shift in the threshold is roughly equal to the ponderomotive energy and (b) a lower intensity may sometimes produce a higher detachment rate, because a higher laser intensity may shift the threshold such that the nominal lowest-order process is no longer possible (channel closing) and detachment must proceed via higher orders. Far below saturation, the averaged detachment rate is

$$\bar{\Gamma}(E_{\text{c.m.}}) = \int_0^{I_{\text{peak}}} W(I)\Gamma(E_{\text{c.m.}}, I)\,dI, \tag{5}$$

where I is the laser intensity, and W is a weighting function of intensity to be determined by laser-H^- beam-overlap conditions. We simulated the experiment with a uniform H^- beam (diameter 3 mm) and a Gaussian distribution of laser intensity, in space and time, with the same values of FWHM as in the experiment. Restricting the photon energy range, 0.37–0.39 eV (α varies from 33° to 18°), approximating $W(I)$ as that for $\alpha = 25°$, and calculating Γ for $E_p = 0.001$ up to 0.02 eV at a step size of 0.001 eV, with $I_{\text{peak}} = 2$ GW/cm^2, we approximated $\bar{\Gamma}$ as a sum of twenty terms. This simulation is compared directly (no free parameters) with the results from the experiment in Fig. 4. Although none of the twenty Γ curves comprising the simulation fits the experimental data, the weighted sum suggests that the apparent small shift in the threshold may be attributed to the laser intensity distribution.

In summary, with moderately intense laser fields we have observed threshold structures in the MPD of H^-. Transitions between orders become less discernible as the range of laser intensities contributing to the signal increases. The dependence of the MPD rates on the laser intensity deviates from the power law in situations well below saturation, indicating a nontrivial dependence of the absorption cross section on laser intensity. Fitting the two-photon threshold data with the Wigner threshold law reveals an upward shift of the detachment threshold energy, smaller than the ponderomotive energy associated with the peak laser intensity. The good agreement between a theoretical simulation and experimental data illustrates the critical importance of the intensity distribution in the space-time volume of the interaction.

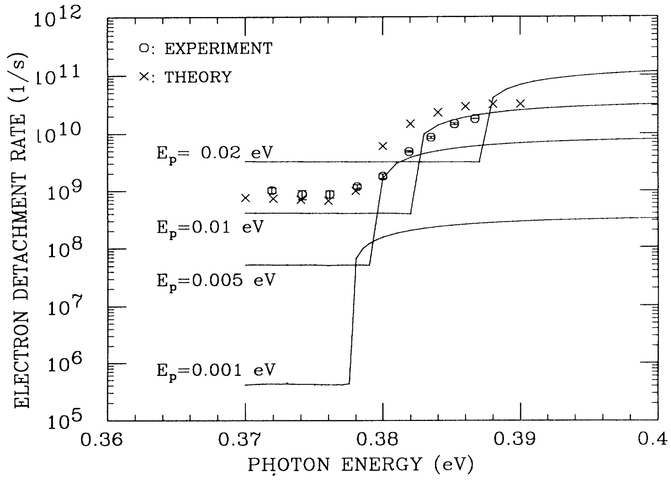

FIG. 4. Four examples of calculations using Eq. (4) for $E_p = 0.001$, 0.005, and 0.01, and 0.02 eV (solid curves), and direct comparison between intensity-weighted detachment rates and experimental data for $I_{\text{peak}} = 2$ GW/cm^2.

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