Laser measurement of the photodetachment cross section of H^- at the wavelength 1064 nm

Mickaël Vandevraye, Philippe Babilotte, Cyril Drag, and Christophe Blondel*

Laboratoire Aimé-Cotton, Centre National de la Recherche Scientifique, Université Paris-Sud, Ecole Normale Supérieure de Cachan, F-91405 Orsay Cedex, France

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A method is described to measure photoexcitation cross sections, relying on the expected behavior of the signal in the saturated regime, when excitation is provided by a Gaussian light beam. The method is implemented on a negative ion beam, with a single-mode pulsed Nd:YAG laser, to make a laser measurement of the photodetachment cross section of H^-, at the wavelength 1064 nm. This cross section is of importance both as a photodetachment cross section of the most elementary negative ion and as a key parameter for the production of fast neutral H^0 or D^0 atoms, by photodetachment of accelerated anions. The obtained value $4.5(6) \times 10^{-21}$ m^2 is greater than the one known from older measurements and most *ab initio* calculations.

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The concept of an effective cross section, to characterize the fraction of an incident flux to be cut out by target particles, is as old as the kinetic theory of gases. Clausius [1] already used ρ, the radius of the "effective sphere" (*Wirkungssphäre*) of a molecule, to express its probability per unit of time $\pi\rho^2 nv$ to have a collision with another molecule, with v their relative velocity and n the vapor density. The cross section σ of the sphere appears here conspicuous, as the factor $\pi\rho^2$. At the only expense of a change of reference frame, the same formula gives the probability P, for a single target, to undergo a collision from a particle flow of velocity v and density n:

$$\frac{dP}{dt} = \sigma n v. \tag{1}$$

The efficiency of all processes where a linear fraction of an incoming flux is scattered by a target can be described by an effective cross section σ, defined by Eq. (1).

Absorption, as long as the intensity remains low enough for the atomic response to remain linear, can be described by the same formula. When the light quantum introduced by Einstein [2], after the discovery of the Compton effect [3], finally caught on as a particle and was named a photon [4], the interaction of light with matter reverted to the familiar problem of particle collisions. Yet transitions between discrete states offer a variety of possible nonlinear regimes, for which the cross-section concept does not provide appropriate modeling. Cross sections thus remain mainly used for discrete-to-continuum transitions, such as photoionization or photodetachment (i.e., photoemission of an electron from a neutral atom or from a negative ion, respectively). Discrete-to-continuum excitation cross sections are essential atomic quantities, inasmuch as they are determined by both a bound and an unbound wave function, and the way these wave functions overlap. Atomic photoionization and photodetachment experiments thus provide important tests of the consistency of calculated atomic wave functions and related models of light-matter interaction.

Among possible reactions, photodetachment of H^- deserves special interest. Atomic photodetachment can be considered as a case even simpler than atomic photoionization, which is complicated by the presence of an infinite-range Coulomb potential in the final state. With this consideration in mind, photodetachment of H^-,

$$H^- + h\nu \rightarrow H + e^-, \tag{2}$$

appears as the simplest example. From now on and for the sake of simplicity, we shall often tell of photodetachment only, with an initial ion and a final neutral atom. The method we describe would nevertheless apply to photoionization as well.

Making a precise measurement of a photodetachment cross section has been a frustrating game, especially when compared to the tremendous accuracy achieved by spectroscopy. If the photoexcitation probability can be assumed small, integration of Eq. (1) for a finite time t immediately gives the number N of photodetached ions:

$$N = N_0 \sigma \Phi t \tag{3}$$

with N_0 the number of illuminated ions and Φ the incident photon flux. Knowing N_0, Φ, and t, and measuring N is, in principle, enough to determine the cross section σ. Unfortunately, measuring these quantities with an accuracy better than a few percent has remained a difficult task. Even when particle counting happens to be possible, the question remains of the ratio of detected events with respect to the actual number of detached ions (i.e., the quantum efficiency). Experimental estimates of photoionization or photodetachment cross sections thus seldom go beyond a $\pm 15\%$ accuracy, which often prevents these measurements from making a significant discrimination between available theoretical models.

To avoid the difficulties of particle detection calibration, a second method can be implemented, looking for the saturation of the excitation signal as a function of the photon flux Φ. Since the excitation probability given by Eq. (1) only applies to the yet unexcited population of ions, integration yields a final photodetachment probability

$$P = 1 - [\exp(-\sigma \Phi t)] \tag{4}$$

that does not grow indefinitely as $\sigma \Phi t$, but exhibits saturation when becoming a non-negligible fraction of unity. Experimental observation of this break in the linear increase of P provides a direct probability scale, with no necessity of an absolute measurement of either the initial atomic target or the product density. A reliable measurement of the photon flux

*christophe.blondel@u-psud.fr

Φ and a proper model of the interaction time are the only requirements left.

Implementing this saturation-based method became easier with the advent of lasers. As soon as 1965, Hall *et al.* [5] calibrated their two-photon detachment experiment of I^- by saturating the photodetachment of H^- and checking the photodetachment cross section obtained in this way (but did not give what value they obtained). Ambartzumian *et al.* [6] explicitly introduced the saturation method as new and used it to measure the photoionization cross section of excited Rb atoms. It became then a standard method for photodetachment cross-section measurements, either in an ion trap [7] or with an ion beam [8] and for multiphoton processes [9,10].

A difficulty, whatever the method, is that very rarely do all illuminated ions receive the same photon flux. Even in a regime of continuous-wave illumination, the ions of the periphery of the laser beam are irradiated by only a fraction of the intensity produced on the laser beam axis. As a consequence, the total detachment signal, as a function of the laser power or laser pulse energy, does not directly reproduce the form given by formula (4). There is a continued increase of the signal, after saturation has been reached on the laser beam axis, due to the expansion of the volume V_S where saturation occurs.

The original method that we present here relies on the observation that when the laser beam has a Gaussian profile, the increase of the signal follows such a mathematical law that the cross section can be deduced from the observation of the saturated regime only.

As a general rule, the photon flux Φ varies in space and time. If the laser beam propagates in direction z with a constant spatial profile in directions x and y, allowing for a motion of the ion at velocity v across the light beam along direction x, with x and y the ion coordinates at $t = 0$, one can generalize formula (4) into

$$P(x,y) = 1 - \left[\exp - \left(\sigma \int_{-\infty}^{\infty} \Phi(x + vt, y, t)dt\right)\right]. \quad (5)$$

The total detachment signal N is just the sum of this probability over all illuminated ions.

A cylindrical Gaussian profile $\frac{2}{\pi w_0^2} \exp[-2(\frac{x^2+y^2}{w_0^2})]$, with a "waist" parameter w_0, is a good approximation of the volume distribution of a mode-cleaned laser beam. An order of magnitude of the length over which the beam can be kept collimated, i.e., the transverse profile kept constant, is given by the Rayleigh length $Z_R = \pi \frac{w_0^2}{\lambda}$. For a wavelength $\lambda = 1\ \mu$m and the smaller waist that we use, $w_0 = 70\ \mu$m, Z_R remains greater than 15 mm. This is one order of magnitude greater than the beam length used in the experiment. Assuming a constant transverse profile thus seems to be a legitimate approximation.

As for the time profile, a practical, though more disputable, hypothesis for a single-mode laser pulse is that it can be represented by a Gaussian too, with a characteristic duration τ. With these hypotheses, the photons of frequency ν carried by a laser pulse of energy E are distributed in a flux

$$\Phi(x,y,t) = \left(\frac{2}{\pi}\right)^{3/2} \frac{E}{h\nu w_0^2 \tau} \exp\left[-2\left(\frac{x^2+y^2}{w_0^2} + \frac{t^2}{\tau^2}\right)\right]. \quad (6)$$

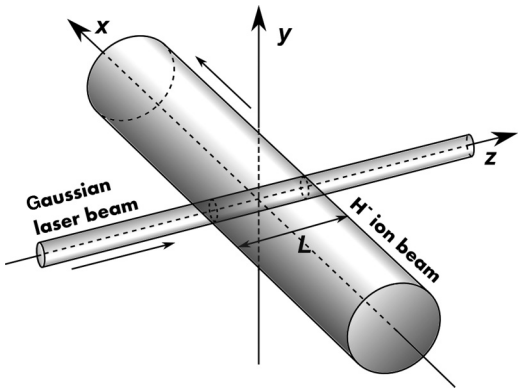

FIG. 1. The situation considered of two beams crossing at right angles, with a laser beam much narrower than the ion beam, the diameter L of which determines the length of the contributing zone of the laser beam.

With this form of the laser space-time profile, Eq. (5) can be volume integrated explicitly. Assuming a uniform ion density n, calling L the length of the laser beam to be crossed by the ion beam (as depicted by Fig. 1) and using, for the sake of compactness, auxiliary variables $\rho = \sqrt{w_0^2 + v^2\tau^2}$ and $A = \frac{2}{\pi}\frac{E}{h\nu}\frac{\sigma}{w_0\rho}$ one gets

$$N = \frac{\pi}{2} nLw_0\rho\,[\ln A + \gamma - \mathrm{Ei}(-A)] \quad (7)$$

with Ei the exponential integral [11] and γ Euler's constant. This extends a formula already written by Arutyunian *et al.* [12] to the case of moving ions, at the only expense of an additional hypothesis on the time profile of the laser pulse.

As $\mathrm{Ei}(-A)$ tends towards 0 faster than $\exp(-A)$ when A becomes greater than 1, $N(E)$ rapidly merges with the asymptotic law

$$N_{as}(E) = \frac{\pi}{2} nLw_0\rho \left[\ln E - \ln\left(\frac{\pi}{2}\frac{w_0\rho}{\sigma}h\nu\right) + \gamma\right]. \quad (8)$$

The $\ln E$ leading term, in the saturated regime, is a simple consequence of the Gaussian shape of the beam, given by Eq. (6). When the maximum flux on axis

$$\Phi_0 = \Phi(0,0,0) = \left(\frac{2}{\pi}\right)^{3/2} \frac{E}{h\nu w_0^2 \tau}$$

exceeds the minimum flux Φ_S required for bringing the most intensely illuminated ions to saturation, the photodetachment signal can still grow, when the laser pulse energy E increases, due to the expansion of the volume V_S in which the local maximum intensity is greater than Φ_S. The condition can be written

$$\Phi_0 \exp\left(-2\frac{x^2+y^2}{w_0^2}\right) > \Phi_S, \quad (9)$$

which shows that the relevant cross section of the laser beam has an area $\pi\frac{w_0^2}{2}\ln(\frac{\Phi_0}{\Phi_S})$. The volume, inside the laser beam, that provides the saturated signal thus grows as a linear function of $\ln(\Phi_0)$ or $\ln E$. A linear and logarithmic plot of photodetachment data, an example of which is given by Fig. 2, makes this property conspicuous.

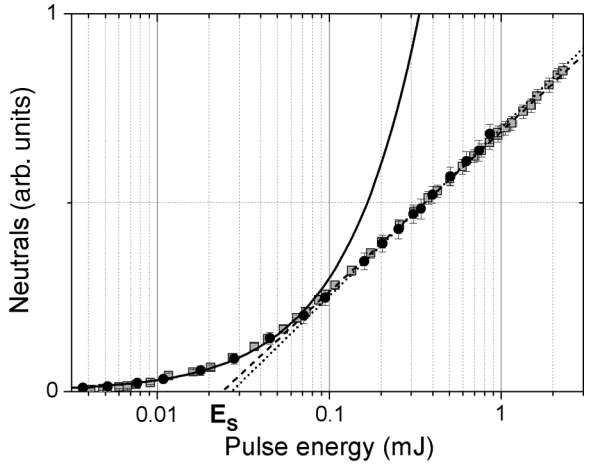

FIG. 2. Detachment signal as a function of the laser pulse energy plotted along linear and logarithmic scales, which makes the representation of the linear regime look like an exponential and the saturated regime be represented by a straight line. The intercept E_S of this line on the $N = 0$ axis is enough to measure the photodetachment cross section. The data represented by light squares (black circles) and interpolated, for the saturated part, by dashes (dots) are for a laser waist $w_0 = 70.6(20)$ μm [80.2(20) μm].

Formula (8), however, tells something more, namely, that the intercept E_S of the asymptote with the $N = 0$ axis contains all the information needed to figure out the experimental cross section:

$$\sigma = \frac{\pi e^{-\gamma}}{2}\frac{h\nu}{E_S} w_0 \sqrt{w_0^2 + v^2\tau^2}. \tag{10}$$

As far as orders of magnitude are concerned, formula (10) just tells that saturation begins at an energy E_S such that the number of photons $\frac{E_S}{h\nu}$ flowed in the effective section of the laser beam is of the order of 1 per cross section.

Linear and logarithmic plots of the photoionization yield similar to Fig. 2 have been introduced by Hankin *et al.* [13,14] and Smits *et al.* [15] in the multiphoton regime, but were used to define a "saturation intensity" I_S, which remains a function of both the time profile of the excitation laser and the (generalized) cross section.

In the present experiment, a scheme similar to the one described by Fig. 1 is implemented on an H^- ion beam produced by a Cs-sputtering negative ion source (SNICS II) [16]. The ion beam is crossed by the laser about 2 m downstream from the source by a pulsed Nd:YAG laser. The diameter of the ion beam is controlled by a circular aperture of 2 mm in diameter, a few decimeters before the interaction region. The laser is focused at right angles onto the ion beam down to waist values w_0 of either 70 or 80 μm.

The H^- beam is extracted from the ion source with potential differences of several kV, then decelerated down to 1.2 keV of kinetic energy before being steered to the interaction region. The neutral atoms produced in the photodetachment process, which go on with roughly the same kinetic energy as the ions, are easily detected by an electron multiplier (Hamamatsu R5150-10). Linearity of the neutral detection signal was tested by checking the proportionality of the recorded neutral signal to the variable negative ion current, at a fixed laser power.

Variation of the laser pulse energy without changing the spatial profile was achieved by means of a half-wave plate set before a Glan-Taylor polarizer. The energy was measured with Ophir PE10-C and PE50BF-DIF-C pyroelectric energy sensors, the calibration of which had been factory checked less than one year before. The measurements showed no detectable discrepancy in the common part of the sensor energy ranges, which makes us definitely confident about their $\pm 3\%$ accuracy.

Modeling the variation of the laser intensity with a Gaussian time profile makes sense only if the pulsed laser does not exhibit the rapid modulations usually produced, in nanosecond lasers, by mode beating. Single-mode operation is achieved by injection, into the pulsed laser cavity, of the cw beam of a Nd:YVO_4 laser [17].

Fitting a Gaussian profile to the measured time profile gives $\tau \simeq 17$ ns. With this laser pulse duration and a 479 210 ms^{-1} ion velocity, $v\tau \simeq 8$ mm is much larger than the waist parameter of the laser. The interaction time of the ions with the laser is thus essentially determined by their transit time across the laser beam, much shorter than the laser pulse duration itself. As a consequence, formula (10) can eventually be simplified into

$$\sigma = \frac{\pi e^{-\gamma}}{2}\frac{h\nu}{E_S} w_{0y} v\tau, \tag{11}$$

where w_{0y} is a measure of the laser waist along the direction orthogonal to the ion velocity (in case the laser beam symmetry happens to deviate from circular symmetry). In this transit-time limited regime, variations of the laser waist w_{0x} in the x direction are compensated by a proportional variation of the illumination time: The smaller the waist, the higher the local laser intensity, but the shorter the interaction time, and that makes the illumination integral an invariant with respect to w_{0x}, as shown by formula (11).

The results shown in Table I take the loss of 10% of the laser energy in a secondary laser pulse into account (the photodetachment yield of this extra pulse is eliminated by time-selective signal integration). From these data, using formula (10), one can deduce a value of the photodetachment cross section $\sigma = 4.6(8) \times 10^{-21}$ m^2. This can be compared to the $\sigma = 4.5(6) \times 10^{-21}$ m^2 value obtained by fitting the whole detachment curves with the numerical integral of (5) over the interaction volume. The uncertainty budget of the method is the sum of a ± 2 μm uncertainty on the waist value, a ± 1 ns uncertainty on the laser pulse duration, a $\pm 3\%$ uncertainty on the laser pulse energy, and a $\pm 6\%$ statistical uncertainty. Too much of the uncertainty comes from possible systematic errors for any statistical reduction of the uncertainty to be

TABLE I. Measurements carried out with different laser waists and pulse energies. The laser energy is corrected for the loss of 10% of the energy in a secondary pulse, as revealed by a fast photodiode.

w_{0y} (μm)	E_S (μJ)	σ (10^{-21} m^2)
	22.6(60)	4.3(16)
70.6(20)	21.6(50)	4.5(11)
	19.3(40)	5.0(12)
80.2(20)	24.4(70)	4.4(18)
	22.6(40)	4.8(27)

expected when merging both results. The overall result for the photodetachment cross section of H^- at 1064 nm thus remains $\sigma = 4.5(6) \times 10^{-21}$ m^2.

The use of a pulsed laser raises the question whether higher-order processes can perturb the analysis. Two-photon detachment can take place even when one photon is enough to detach an electron [18], but with a rate proportional to the square of the photon flux. With a generalized cross section of the order of 2×10^{-56} s m^4 [19–21], the two-photon detachment rate at the maximum photon flux that we have, about 7×10^{31} photons s^{-1} m^{-2}, remains less than one-thousandth of the one-photon rate to be measured. Another typical effect in pulsed laser fields is the ponderomotive shift they produce on detachment thresholds, due to the quiver energy of the freed electron in the electromagnetic field [22]. With the peak intensity used in the present experiment however, the maximum value of the ponderomotive shift is only 0.13 meV, which has but negligible effects on electrons released with a nominal kinetic energy of 0.411 eV.

The photodetachment cross section of H^-, though of primary importance as a test of photoexcitation models in elementary systems, was seldom measured in photodetachment experiments. Branscomb and Smith [53] measured it with a tungsten filament lamp and color filters between 400 and 900 nm, and found $3.9(5) \times 10^{-21}$ m^2, which nicely agreed with the calculation of Chandrasekhar [27]. Popp and Kruse [54] made quantitative radiation measurements on a low current hydrogen arc and found a slightly smaller value of $3.6(3) \times 10^{-21}$ m^2, which seemed to corroborate more recent calculations.

Bacal and Hamilton [55] measured the fraction of H^- photodetached in a plasma by a ruby laser, as a function of the pulse energy, and found the saturation curve compatible with a $\sigma = 4 \times 10^{-21}$ m^2 cross section, even though that was at a wavelength a little shorter than the cross-section maximum. The experimental uncertainty (visually $\pm 20\%$ at least) did not, however, allow one to detect any discrepancy, as far as the value of the cross section was concerned, with existing literature. Similar plasma diagnostics were made by Nishiura *et al.* [56] with a Nd:YAG laser, but the very sharp increase of the photodetachment yield as a function of the energy, at the wavelength 1064 nm, again ruled out any precise measurement of the cross section beyond confirming its order of magnitude. Only at the third-harmonic wavelength of 355 nm does the more progressive increase of the photodetachment yield suggest a small discrepancy, for the experimental points appear to lie systematically above the calculated curve (Fig. 7 of [56]).

The photodetachment cross section of H^- was the subject of many calculations (see Table II), most of which converged to a 3.6×10^{-21} m^2 value at the wavelength 1064 nm. However, the main reason that led Bell and Kingston [36] to dismiss their velocity-gauge calculated 3.9×10^{-21} m^2 value was that it did not match previous calculations so well. More recently the adiabatic approximation in hyperspherical coordinates produced a significantly greater value of 4.2×10^{-21} m^2 [20,44]. The use of the nonlocal Yamaguchi potential [21] even led to a cross section greater than 5×10^{-21} m^2, if one was to adopt the value of its β parameter obtained by fitting the model to the measured electron affinity [57].

The present result $\sigma = 4.5(6) \times 10^{-21}$ m^2 questions the validity of the models that have predicted the photodetachment cross section of H^- at 1064 nm to lie in the $3.6 \pm 0.1 \times 10^{-21}$ m^2 interval. On the other hand, the new measurement appears quite compatible with the first one, $\sigma = 3.9(5) \times 10^{-21}$ m^2 [53], and marginally compatible with the lower $3.6(3) \times 10^{-21}$ m^2 value deduced from the spectroscopy of a hydrogen-arc discharge [54]. Finding a reason why the latter experiment could underestimate the photodetachment cross section is out of the scope of the present study. The former experiment, with crossed ion and light beams [53], was more similar to our experiment. Using light produced by a lamp or by a laser should not make any difference in the value of the cross section, but knowing the actual illumination of the ion beam may be easier with a laser, the energy per pulse of which can be conveniently measured just before and after the interaction region.

In conclusion, our experiment has shown that recording the variation of the saturated signal only is enough to measure a

TABLE II. Calculated (upper part) and measured (lower part) values of the photodetachment cross section of H^- at the wavelength 1064 nm (or, equivalently, 0.411 eV or 0.0151 atomic unit of photoelectron energy).

Reference	Year	σ (10^{-21} m^2)
C. K. Jen [23]	1933	2.8
H. S. W. Massey *et al.* [24]	1940	0.8
R. E. Williamson [25]	1942	1.1
L. R. Henrich [26]	1944	2.8
S. Chandrasekhar [27]	1945	3.9
S. Geltman [28]	1956	3.59
S. Chandrasekhar *et al.* [29]	1958	3.7
T. Ohmura *et al.* [30]	1960	3.5
T. L. John [31]	1960	3.44
T. Tietz [32]	1961	3.77
S. Geltman [33]	1962	3.52
B. H. Armstrong [34]	1963	3.6
N. A. Doughty *et al.* [35]	1966	3.52
K. L. Bell *et al.* [36]	1967	3.54, 3.90
M. P. Ajmera *et al.* [37]	1975	3.55, 3.43
T. N. Rescigno *et al.* [38]	1976	2.5
M. A. C. Nascimento *et al.* [39]	1977	3.5
J. T. Broad *et al.* [40]	1976	3.4
A. L. Stewart [41]	1978	3.58, 3.60
A. W. Wishart [42]	1979	3.4
M. Daskhan *et al.* [43]	1983	3.5
M. Crance *et al.* [19]	1985	2.9
M. G. J. Fink *et al.* [20]	1985	4.2
C.-H. Park *et al.* [44]	1986	4.2
H. P. Saha [45]	1988	3.58
T. N. Chang *et al.* [46]	1991	3.8
C. Laughlin *et al.* [47]	1993	3.58
A. G. Abrashkevich *et al.* [48]	1994	3.60, 3.56
M. Venuti *et al.* [49]	1997	3.52
A. S. Kheifets *et al.* [50]	1998	3.6
W. H. Kuan *et al.* [51]	1999	3.5
V. A. Pazdzersky *et al.* [21]	2000	3.6, 5.6
A. M. Frolov [52]	2004	3.59
L. M. Branscomb *et al.* [53]	1955	3.9(5)
H. P. Popp *et al.* [54]	1976	3.6(3)
Present data	2014	4.5(6)

photodetachment cross section. The method remains sensitive to possible deviations of the laser beam from the ideal Gaussian profile, in the same way as fitting the detachment yield curve with numerical simulations has shown to be sensitive to the input parameters. The numerical fitting procedure itself, which is not restricted to the case of a Gaussian profile, has shown to remain sensitive to the temporal shape of the laser pulse, for a fixed average duration, even though we have only considered a one-photon process. This observation makes it all the more necessary to monitor the laser time profile accurately and, preferably, work with a single-mode laser. The obtained results are consistent in giving a value of the photodetachment cross section of H^-, at the wavelength 1064 nm, greater than predicted by most *ab initio* calculations. This observation could motivate reinvestigations of the description of electron correlations in H^-, both for the ground and for the excited states of this most elementary three-body system. Having a better known value of the photodetachment cross section also has some importance for dimensioning future photodetachment-based D^0 injectors, in the framework of magnetically controlled nuclear fusion [58].

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