Experimental Observation of Laser-Stimulated Radiative Recombination

F. B. Yousif, P. Van der Donk, Z. Kucherovsky, J. Reis, E. Brannen, and J. B. A. Mitchell
Department of Physics, University of Western Ontario, London, Ontario, Canada N6A 3K7

T. J. Morgan
Physics Department, Wesleyan University, Middletown, Connecticut 06457

(Received 13 March 1991)

Spontaneous radiative recombination between protons and electrons to form hydrogen atoms with $8 \leq n \leq 19$ has been measured. CO_2 laser light has been shown to induce stimulated radiative recombination to the $n=11$ and 12 levels with an inferred gain in the cross section of 1720 ± 860 and 4790 ± 2830 for a laser power of 12.6 and 15.3 W, respectively. This is in line with that predicted theoretically.

PACS numbers: 34.80.Kw, 32.90.+a, 35.80.+s

When a thermal-energy electron makes a close collision with a free proton, during a very short interval ($\sim 10^{-15}$ s), the system can have the configuration of a bound electron moving in an excited Bohr orbit around the nucleus. If, during this interval, this quasi hydrogen atom undergoes a transition down to a lower orbit, emitting a photon, then the energy of the electron may be insufficient to escape and the recombination process is stabilized. This is known as radiative recombination and is in fact a very-low-probability process since the characteristic time for radiation emission is of the order of 10^{-9} s. Usually, the electron will simply be scattered by the proton and continue on its way.

Bethe and Salpeter [1] derived the following expression for the radiative-recombination cross section for capture into a given state with principal quantum number n:

$$\sigma = (2.1\times 10^{-22}\,\mathrm{cm}^2)\frac{E_0^2}{nE_e(E_0+n^2E_e)}, \tag{1}$$

where $E_e = \frac{1}{2}mv_e^2$ is the kinetic energy of the electron and $E_0 = 13.6$ eV, the ionization potential of the $n=1$ level of atomic hydrogen. The frequency ν of the stabilizing photon is given by

$$h\nu = \tfrac{1}{2}mv_e^2 + E_0/n^2 . \tag{2}$$

If the electron-proton collision takes place in an intense photon field, then it is possible that the stabilizing transition can be pumped by photon impact. In other words, stimulated emission of the stabilizing photon can occur with the frequency of the pumping and emitted photons being the same. This process, referred to as *laser-stimulated radiative recombination* can be expressed as

$$h\nu + e^- + \mathrm{H}^+ \rightarrow \mathrm{H}(n) + 2h\nu , \tag{3}$$

and leaves the hydrogen atom in a particular n state determined by the pumping photon frequency. Neumann *et al.* [2] have examined this process and its possible application to the production of atomic antihydrogen. They have given an equation for the ratio of the stimulated recombination rate over the spontaneous rate:

$$G = \frac{\sigma^{\mathrm{stim}}}{\sigma^{\mathrm{spon}}} = \frac{Pc^2}{F\Delta\nu 8\pi h\nu^3}, \tag{4}$$

where P is the laser power in watts, F is the cross-sectional area of the laser beam, and ν is the laser frequency. (This equation assumes that there is complete overlap between the laser beam and the electron-ion interaction region.) In order to stimulate a given electron to recombine with a proton, the photon frequency must match the frequency of the photon which would be emitted and this is given by the sum of the binding energy of the level into which the electron capture occurs and the kinetic energy of the electron, as given by Eq. (2).

In practice, the frequency spread of the laser will be much smaller than the energy spread of the electrons in the center-of-mass frame of reference and so the $\Delta\nu$ term in Eq. (2) will be dominated by the electron velocity spread Δv_e:

$$\Delta\nu = (mv_e/h)\Delta v_e . \tag{5}$$

In the experiment reported here, a CO_2 laser is used to stimulate the recombination of electrons and protons colliding at low energies in a merged-beam configuration. The center-of-mass collision energy is given by

$$E_{\mathrm{c.m.}} = E_+ + E_e - 2(E_+E_e)^{1/2}\cos\theta , \tag{6}$$

where θ is the intersection angle between the two beams, E_e is the electron-beam energy, and E_+, the reduced ion energy, is given by $E_+ = E_i m_e/m_i$, E_i and m_i being the ion energy and mass and m_e being the electron mass. If θ is small then $\cos\theta \approx 1-\theta^2/2$ and Eq. (6) can be written as

$$E_{\mathrm{c.m.}} = (E_e^{1/2} - E_+^{1/2})^2 + (E_eE_+)^{1/2}\theta^2 . \tag{7}$$

By differentiating this equation with respect to E_+, E_e, and θ, it is possible to derive an expression for the uncertainty in the center-of-mass collision energy:

$$\Delta E_{\text{c.m.}}=\left\{\left\{\left[1-\left(\frac{E_+}{E_e}\right)^{1/2}\right]\Delta E_e\right\}^2+\left\{\left[1-\left(\frac{E_e}{E_+}\right)^{1/2}\right]\Delta E_+\right\}^2+[2(E_eE_+)^{1/2}\theta\Delta\theta]^2\right\}^{1/2}. \tag{8}$$

It can be seen that when $E_e=E_+$, the first two terms go to zero, and the uncertainty is determined mainly by θ and $\Delta\theta$. Using well-collimated beams and careful alignment, these quantities can be made very small and so in principle the merged-beam approach can produce very-high-energy-resolution measurements even though the uncertainties in the electron and ion energies are quite large. Calibration of the center-of-mass energy was performed using a procedure described in Ref. [3].

A discussion of this experimental method and its relationship to antihydrogen production has been given in previous reviews [4-6].

The merged-electron-ion-beam apparatus has been described in detail elsewhere [7]. A proton beam of 10^{-8} A is produced using a radio-frequency (rf) source mounted in the terminal of a 400-keV Van de Graaff accelerator. The ions are mass analyzed and injected into the collision chamber where they are deflected to eliminate neutral atoms formed in the beam line. An indirectly heated barium-oxide cathode emits a 100-μA electron beam which is merged with the protons using a trochoidal analyzer. The two beams interact over a distance of 8.6 cm, the overlap being measured at two points along the merging region using scanners which intersect the beams horizontally and vertically [8]. After the interaction region the ions are electrostatically analyzed and the protons are deflected into a Faraday cup.

The neutral hydrogen atoms formed in the interaction region, due to both electron-ion recombination and charge exchange with the background gas, pass undeflected into a static electric-field ionizer which consists of two highly polished aluminum surfaces separated by 1.0 mm, each with an aperture of 1.2 mm diameter. This device can generate an on-axis electric field of up to 150 kV/cm along the axis of the beam which is high enough to ionize atoms in excited Rydberg states with $n\geq 7$. Ions produced by field ionization are then deflected onto a surface-barrier detector. Much attention has been given to the elimination of neutral atoms not formed by electron-ion recombination. Charge exchange with the background gas is minimized by maintaining the pressure in the chamber below 10^{-10} Torr, and a high primary ion-beam energy (330 keV) is used so that the total charge-exchange cross section is low ($\sim 5\times 10^{-20}$ cm^2). Furthermore, the principle behind this measurement is to examine the formation of hydrogen atoms in high-n states using the field ionizer and this further reduces the background since the charge-exchange cross section into a particular n state scales as n^{-3}.

Multiple apertures inside the chamber minimize the chance of scattered particles hitting the detector. Final discrimination between background and true electron-ion signals is achieved by modulating the electron beam and counting the detector signal in and out of phase with this modulation. The background count rate is very low (typically 5 counts/min), but given the small cross sections being measured, counting times of up to 24 h per point are required to reduce statistical errors to acceptable levels.

The beam from a 20-W CO_2 laser, operated in cw mode (TEM$_{00}$) at a wavelength of 10.535 μm, is reflected into the collision chamber through a zinc-selenide window. It crosses at right angles with the merged ion-electron beam in the interaction region and then exits through a second window. A computer-controlled feedback system assures that the laser remains tuned to the desired wavelength. By measuring the laser-intensity profile, with a two-dimensional scanning pinhole, just before and just after entry, the divergence of the beam can be determined and losses due to transmission through the windows can be corrected for.

The ionizer was operated with an on-axis field of 105 kV/cm so that hydrogen atoms with $n\geq 8$ would be field ionized. The resulting proton signal was counted as described above. Prior to arriving at the field ionizer, however, the atoms had to pass through the charge-state analyzer where the field was 2.4 kV/cm, which is sufficient to field ionize atoms with $n\geq 19$, so that they did not arrive at the field ionizer. A length of 1.4 mm of the interaction region was illuminated by a 10.535-μm beam from the CO_2 laser and the absolute value of the effective recombination cross section was measured as a function of center-of-mass energy. [Since the measured stimulated recombination cross sections are a function of laser power, illumination, distance, etc., they are referred to as effective cross sections. In the absence of the laser beam (and off resonance), the cross sections are absolute.] The results are shown in Figs. 1(a) and 1(b).

The peak in the effective cross section in Fig. 1(a) at 5.33 meV is due to the enhancement of the recombination to the $n=11$ level of atomic hydrogen, by the stimulated emission of radiation induced by the laser beam. This enhancement only occurs when the resonance condition given in Eq. (2) is satisfied. The width of the peak (0.5 meV) reflects the energy resolution of the experiment. Figure 1(b) shows the same process occurring for the $n=12$ level. Here, the peak width is 0.7 meV. The ion energy used for these measurements was 330 keV. In order to achieve such a high energy resolution, Eq. (8) indicates that θ and $\Delta\theta$ must be less than one-tenth of a degree. Off resonance, the measured cross sections are for the spontaneous radiative recombination of protons and electrons to form $H(8\leq n\leq 19)$ and the peaks are super-

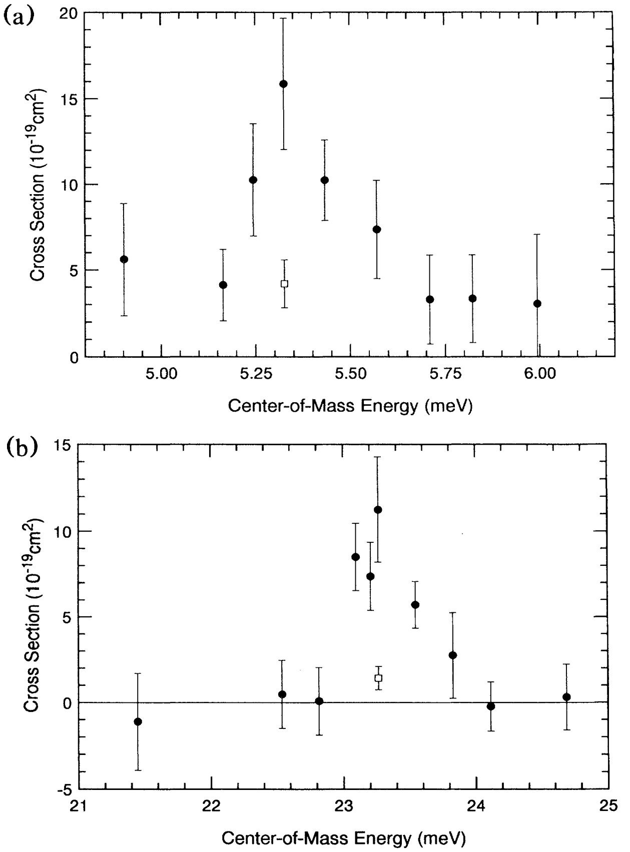

FIG. 1. Measured effective electron-proton recombination cross section with, ●, laser on and, □, laser off. (a) Peak at 5.33 meV corresponds to stimulated recombination to the $n=11$ level. (b) Peak at 23.3 meV indicates stimulated recombination to the $n=12$ level.

imposed upon this background process. The data points shown as squares indicate measurements that were taken with the laser beam off so that there was no stimulated contribution. Using Eq. (1), the calculated values for $\sigma_{\text{spon}}(8 \leq n \leq 19)$ are 4.92×10^{-19} and 9.41×10^{-20} cm^2 at 5.33 and 23.3 meV, respectively. The corresponding measured values are $(4.2\pm1.4)\times10^{-19}$ and $(1.4\pm0.7)\times10^{-19}$ cm^2. Considering the difficulty of measuring such a small cross section, the agreement can be considered to be very good. The peak values of the cross sections, including the stimulated contributions, are $(15.96\pm3.8)\times10^{-19}$ and $(11.2\pm3.0)\times10^{-19}$ cm^2. Subtracting the spontaneous recombination contribution to the cross section in each case yields effective stimulated recombination cross sections of $(11.7\pm4.0)\times10^{-19}$ cm^2 for $n=11$ and $(9.8\pm3.1)\times10^{-19}$ cm^2 for $n=12$. Equation (1) shows that $\sigma_{\text{spon}}(n)/\sigma_{\text{spon}}(8 \leq n \leq 19)$ are 0.097 and 0.089 for $n=11$ and 12. Using these ratios and the measured values of $\sigma_{\text{spon}}(8 \leq n \leq 19)$ at 5.33 and 23.3 meV, one obtains corresponding values for $\sigma_{\text{spon}}(n=11)$ and $\sigma_{\text{spon}}(n=12)$ or $(4.2\pm1.4)\times10^{-20}$ and $(1.25\pm0.6)\times10^{-20}$ cm^2. Taking the ratio $\sigma_{\text{stim}}/\sigma_{\text{spon}}$ gives 28 ± 14 for $n=11$ and 78 ± 46 for $n=12$. The laser beam, however, has a FWHM diameter of 1.4 mm and this intersects the interaction region (length 86 mm) at right angles. The stimulation only applies therefore to the small overlap region. If the entire interaction region had been illuminated, the resulting gains would have been 1720 ± 860 and 4790 ± 2830. The large errors are due to statistical errors in the measurements of the cross sections.

The laser powers used were 12.6 and 15.3 W for the measurements in Figs. 1(a) and 1(b), respectively. Using these powers, Eq. (3) predicts gains of 2388 and 1139, respectively. Given the uncertainties associated with matching the profiles of the three beams, the agreement between the predicted and measured gains can be assumed to be reasonable.

Neumann *et al.* [2] have discussed the consequence of using too much laser power. This would result in the reionization of the recombined atom by the impact of a second photon. This phenomenon becomes important if the photon flux $\phi \geq (\tau\sigma_{\text{ph}})^{-1}$, where τ is the exposure time (or the lifetime of the n state, whichever is shorter), σ_{ph} is the photoionization cross section, and $\phi = P/Fh\nu$. In our experiment $\tau \approx 0.1$ ns and $\phi \approx 3\times10^{22}$ photons cm^{-2}. Since $\sigma_{\text{ph}} \approx 7.7\times10^{-17}$ cm^2 for 10.535-μm photons interacting with the $n=11$ level of atomic hydrogen [9], the maximum photon flux before reionization can be problematic is $\sim10^{24}$ photons cm^{-2}. This phenomenon has little effect on our results.

Clearly, there are many improvements that can be made to this experiment, the most important being to modify the apparatus so that the entire length of the interaction region can be illuminated by the laser beam. The data in Figs. 1(a) and 1(b), however, clearly show that laser-stimulated radiative recombination has been demonstrated in the fundamental electron-proton system. Analysis of these data indicates that, even with moderate laser powers, one can obtain several thousandfold enhancements of the recombination cross sections to moderately high-n states. One can envisage that this new atomic collision process will have numerous applications, including perhaps the modification of, and diagnosis of, the physical and chemical state of ionized media.

The support of the United States Air Force Office of Scientific Research (Grant No. AFOSR-89-0327) and the Canadian Natural Sciences and Engineering Research Council is gratefully acknowledged.

Note added.—The authors have been informed that laser-stimulated radiative recombination to the $n=2$ level of atomic hydrogen has been observed by Schramm *et al.* [10].

[1] H. Bethe and E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Plenum, New York, 1977).

[2] R. Neumann, H. Poth, A. Winnacker, and A. Wolf, Z. Phys. A **313**, 253 (1983).

[3] F. B. Yousif, P. Van der Donk, M. Orakzai, and J. B. A. Mitchell (to be published).

[4] J. B. A. Mitchell, in *Antiproton Science and Technology,* edited by B. W. Augenstein, B. E. Bonner, F. E. Mills, and M. M. Nieto (World Scientific, Singapore, 1988), p. 359.

[5] J. B. A. Mitchell, in *Intersections between Particle and Nuclear Physics,* edited by G. M. Bunce, AIP Conf. Proc. No. 176 (AIP, New York, 1988), p. 447.

[6] T. J. Morgan and J. B. A. Mitchell, in *Dissociative Recombination: Theory, Experiment and Applications,* edited by J. B. A. Mitchell and S. L. Guberman (World Scientific, Singapore, 1989), p. 175.

[7] D. Auerbach, R. Cacak, R. Caudano, T. D. Gaily, C. J. Keyser, J. Wm. McGowan, J. B. A. Mitchell, and S. F. J. Wilk, J. Phys. B **10**, 3797 (1977).

[8] C. J. Keyser, H. R. Froelich, J. B. A. Mitchell, and J. Wm. McGowan, J. Phys. E **12**, 316 (1979).

[9] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (Univ. California Press, Berkeley, 1981).

[10] U. Schramm *et al.*, preceding Letter, Phys. Rev. Lett. **67**, 22 (1991).