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Two-photon excitation of the $n = 3$ level in H and D atoms

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Tunable coherent ultraviolet radiation has been used to excite selectively the $n = 3$ level in hydrogen and deuterium atoms via two-photon absorption from the ground state. The resulting Balmer- α fluorescence at 656 nm was observed as well as resonant three-photon ionization. This work demonstrates several advantages over other techniques for selective detection of neutral H and D atoms, including three-dimensional spatial resolution and a remote monitoring capability afforded by the use of laser-induced-fluorescence detection.

We report the first observation of two-photon excitation from the $n = 1$ to the $n = 3$ level in hydrogen atoms. The process was detected by both laser-induced fluorescence from the 656-nm Balmer- α transition and two-photon resonant three-photon ionization. Both techniques were highly sensitive, and provided high spatial and temporal resolution. While two-photon resonant three-photon ionization of atomic hydrogen via the $n = 2$ state has been previously demonstrated as a sensitive detection scheme,^{1,2} the use of the Balmer- α fluorescence diagnostic provides an attractive alternative in many experimental situations. In addition, our basic technique opens up new possibilities for laser spectroscopy in atomic hydrogen, a subject of great fundamental interest.

A schematic of the excitation, fluorescence, and ionization processes is shown in Fig. 1. Two photons are absorbed to excite the $3s$ and $3d$ states. An examination of the relevant transition matrix elements³ shows that excitation of $3d$ dominates over $3s$ by at least an order of magnitude for our case of linearly polarized light. As discussed below, two-photon excitation was observed with either $\lambda_1 = \lambda_2 = 205$ nm or $\lambda_1 = 193$ nm and $\lambda_2 = 218$ nm. The excited atoms may then decay to the $n = 2$ level via fluorescent emission at 656 nm, or they may be ionized by absorption of an additional ultraviolet photon.

The excitation source used for these experiments was a tunable argon fluoride excimer laser,⁴ which was frequency shifted via stimulated Raman scattering in D_2 . The fundamental laser wavelength was

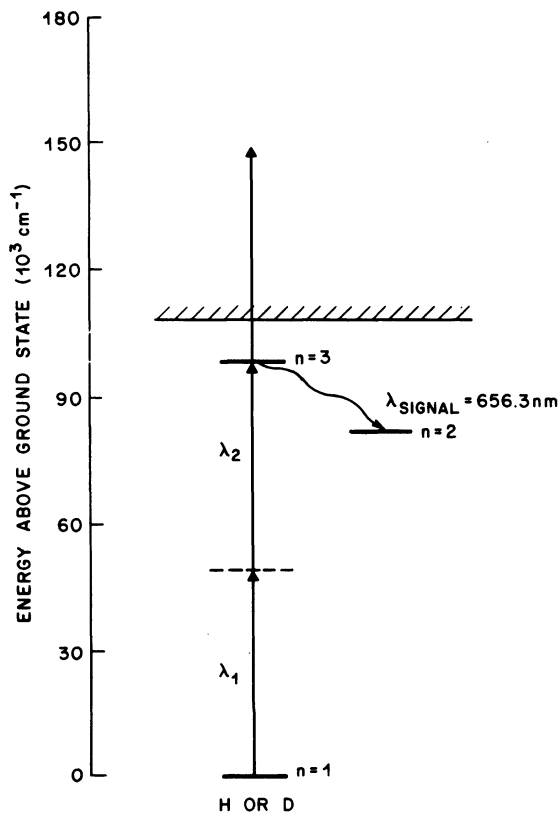


FIG. 1. Schematic energy-level diagram of atomic hydrogen showing the excitation, ionization, and fluorescence processes involved in this experiment.

tunable near 193 nm, the first Stokes output was tunable near 205 nm, and the second Stokes output was tunable near 218 nm. When a deuterium pressure of approximately 100 psi (gauge) in the Raman cell was used, the available pulse energies (at 10 pulses per sec) were approximately 50 mJ at 193 nm, 2 mJ at 205 nm, and 0.1 mJ at 218 nm. The pulse width was 4–5 nsec, and the bandwidth was 1.5 cm^{-1} for all three beams. Figure 2 shows the experimental setup for the specific case of $\lambda_1 = \lambda_2 = 205 \text{ nm}$ (first Stokes) excitation. In the two-wavelength excitation experiments, the fundamental and second Stokes beams were individually isolated, focused, and spatially overlapped in the center of the experimental cell. The focal spot sizes were of the order of 10^{-3} cm^2 .

H and D atoms were produced by flowing mixtures of H_2 and D_2 through a 75-cm-long, 7-mm-i.d. Wood's discharge tube with a dc discharge current of 60 mA and a 10-k Ω ballast resistor. At the entrance to the discharge tube, the total pressure was maintained at about 6 Torr. The gas was pumped from the discharge region through glass tubing into the experimental region. Two separate experimental cells were used. One cell was designed for fluorescence detection as shown in Fig. 2. A 10-nm bandpass filter was used in front of a 1P28 photomultiplier tube. The second cell was designed for ionization detection and consisted of a glass vessel containing two parallel 2.5-cm-square planar metal electrodes separated by 2.5 cm. The ionization signal was then detected as a transient voltage drop across a 200- Ω resistor connected in series with a dc voltage across the electrodes. The fluorescence and ionization signals were further processed using a PAR 162 Boxcar Averter.

Figure 3 shows a typical scan of ionization signal versus excitation wavelength using single beam, first Stokes excitation. The gas mixture used in this case

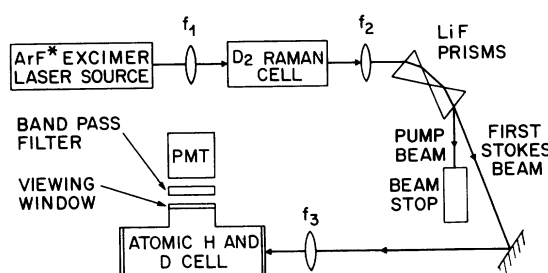


FIG. 2. Experimental arrangement used for observing two-photon laser-induced fluorescence in atomic hydrogen using single beam, first Stokes ($\lambda_1 = \lambda_2 = 205 \text{ nm}$) excitation. For two-wavelength excitation ($\lambda_1 = 193 \text{ nm}$, $\lambda_2 = 218 \text{ nm}$), the fundamental and second Stokes beams were individually isolated, focused, and spatially overlapped in the center of the cell.

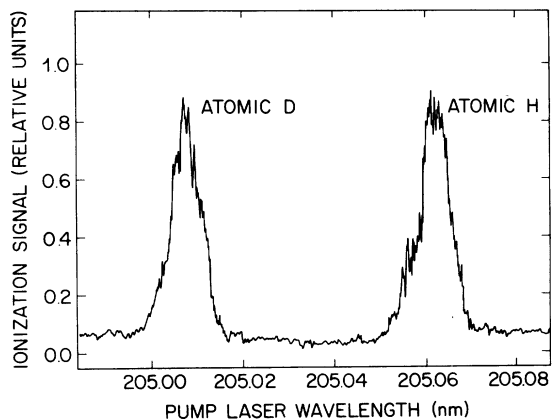


FIG. 3. Ionization signal vs excitation wavelength in the case of single beam, first Stokes excitation.

was 50% H_2 and 50% D_2 . The two isotopic peaks are very clearly resolved. In Fig. 4, an example of a laser-induced-fluorescence spectrum is shown. This particular scan was taken using 100% D_2 in the discharge. The spectrum of Fig. 4 was taken with similar atomic concentration, laser intensity, and signal averaging to that shown in Fig. 3. The lower limit of our sensitivity by fluorescence detection in this apparatus is estimated to be comparable to the value of $3.5 \times 10^9 \text{ atoms/cm}^3$ obtained by Bjorklund *et al.* by ionization detection.¹ In our case, this limit is set by background fluorescence scattered from the Wood's discharge. In any practical application, the

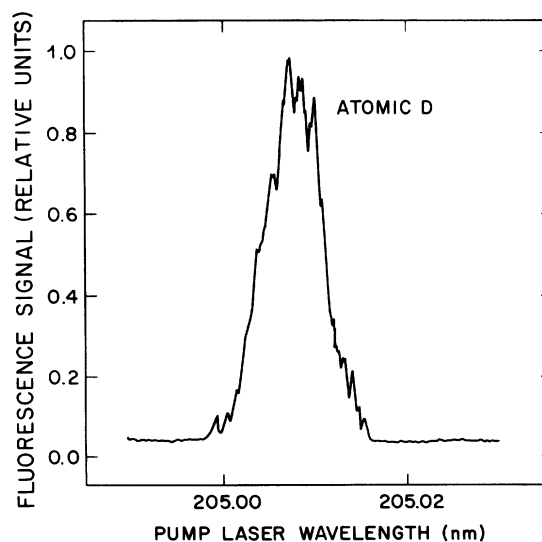


FIG. 4. Balmer- α fluorescence signal vs excitation wavelength in the case of single beam, first Stokes excitation.

detection sensitivity limit will be set by such factors as ambient background fluorescence and fluorescence quenching by both neutral and charged particles.

Similar results were also obtained with two-wavelength excitation where $\lambda_1 = 193$ nm and $\lambda_2 = 218$ nm. In this case we have exploited the simple fact that the Raman shift remains equal to the first Stokes shift for all higher-order outputs. Thus, the sum energy of two photons from the first Stokes beam is identical to the sum energy of one photon from the fundamental beam and one photon from the second Stokes beam. Since the product of the input intensities at $\lambda_1 = 193$ nm and $\lambda_2 = 218$ nm are comparable to the square of the intensity available for $\lambda_1 = \lambda_2 = 205$ nm, the two-photon excitation rates may be expected to be comparable for both excitation methods. However, the higher total intensity used in the two-wavelength case leads to a proportionately larger excited-state ionization rate. Therefore, the sensitivity of the two-wavelength excitation method relative to the single-wavelength excitation method is somewhat enhanced for ionization detection but diminished for fluorescence detection.⁵

The advantage of using two beams of unequal wavelength lies in the ability to probe a well-defined point in three-dimensional space, as was demonstrated by Bjorklund *et al.*¹ This capability was also verified in our experiment. When the focuses of the two beams were spatially overlapped, ionization signals comparable to the single beam excitation case were indeed observed. No signal could be detected due to excitation from either of the isolated laser beams or when the focuses were not properly overlapped. By translating the region of intersection of the two

beams, the three-dimensional distribution of atoms can thus be mapped.

These techniques have several advantages over resonance scattering techniques using either Lyman- α (Ref. 6) or Lyman- β (Ref. 7) sources for applications involving the probing of the interior regions of dense atomic hydrogen distributions such as those found in magnetic confinement fusion machines. As previously discussed, this technique provides high spatial resolution. All of the wavelengths involved may be propagated through the air and readily available window materials. Further, the problems of strong resonance absorption and radiation trapping of the input beam which are inherent in the resonance scattering approaches are eliminated with this two-photon excitation method. While the method described by Bjorklund *et al.*^{1,2} shares these advantages, the approach described here offers the additional advantage of allowing for fluorescence detection rather than ionization detection in situations where it may be inconvenient or overly perturbative to introduce electrodes into the experimental region of interest. Our technique also opens up new possibilities for the study of the dynamical properties of state-selected electronically excited hydrogen atoms. In addition, note that significant improvements in spectral resolution are quite feasible within the general framework of our technique. Single-mode, tunable-excimer-laser systems have been demonstrated.⁸ An improvement of this sort to the present laser system would not only greatly improve the detection sensitivity,⁵ but would also open up additional possibilities for high-resolution spectroscopic studies of atomic hydrogen and its isotopes.

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⁵A full discussion of the excited-state dynamics and fluorescence yield for the general case of two-photon excitation

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