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## Experimental Demonstration of Laser Temporal Coherence Effects on Multiphoton Ionization Processes

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A single-transverse-mode Q-switched Nd-glass laser, which can operate over a variable number of longitudinal modes, has been used to investigate the influence of the temporal nature of the laser pulse on the multiphoton ionization probability of xenon atoms. For a seven-mode laser pulse, the number of ions formed is increased by several orders of magnitude over that produced by a single-longitudinal-mode laser pulse.

Multiphoton ionization processes have been the subject of a considerable amount of recent theoretical and experimental work.<sup>1-7</sup> A significant disagreement has been observed between experimental and calculated multiphoton ionization probabilities. Thus, the experimental value of the six-photon ionization probability of atomic hydrogen has been found to be greater by 3 orders of magnitude than the corresponding probability calculated by several authors. This apparent discrepancy may be explained in terms of coherence of the laser radiation. It should be pointed out that theoretical calculations of the probability  $W$  assume the laser radiation to be a single-mode ideal laser source. However, the laser radiation used in previous experiments contains numerous oscillating modes. The instantaneous laser intensity seen by atoms may be much greater than the average laser intensity  $\bar{I}$  determined in these experiments. The multiphoton ionization probability is

$$W = \alpha g^{(K_0)} \bar{I}^{K_0}, \quad (1)$$

where  $\alpha$  is a factor depending on the atom considered and the polarization of the laser light.  $K_0$  is the next integer greater than the ionization energy of the atom divided by the photon energy.

Thus  $K_0 = 11$  for ionization of xenon atoms by a Nd-glass laser pulse.  $g^{(K_0)}$  is the  $K_0$ th-order normalized correlation function of the laser intensity.<sup>8</sup> This expression (1) is strictly valid for a single-transverse-mode laser pulse only, and when the atom is not ionized through any quasi-resonant intermediate excited states.

The purpose of the present paper is to describe an experiment in which the multiphoton ionization probability has been measured by using a single-transverse-mode Q-switched Nd-glass laser which can be operated with a variable number of adjacent longitudinal modes (from one to ten), thus keeping a narrow spectral linewidth. This laser has been described in detail elsewhere.<sup>9</sup> The laser is Q switched by a Pockels cell. This laser radiation is linearly polarized and can be centered at either 10 603 or 10 643 Å. The number of longitudinal modes is varied from ten to one by using resonant reflectors in the temperature-controlled cavity, and a two-step Q switching. The laser beam emerging from the master oscillator is expanded by a telescope with four-fold magnification for entering a five-stage amplifier. The experimental method is the same as that employed in previous work.<sup>6,10</sup>

To control the longitudinal-mode structure of

the laser radiation, the portion of the laser beam reflected from the window entrance of the interaction chamber passes through two beam splitters, and is then incident on the following:

(1) A Fabry-Perot interferometer which gives information on the longitudinal-mode structure. One to ten adjacent modes can be observed in its free spectral range  $\Delta\lambda = 7.5 \times 10^{-2} \text{ \AA}$ .

(2) A photodiode and an oscilloscope whose combined rise time is 350 psec. Laser intensity fluctuations can thus be recorded from the temporal distribution function  $G(t)$  of the laser intensity.

(3) A photodiode and an oscilloscope whose combined rise time is several nanoseconds. This detection setup is used to determine the duration of the laser pulse from the integrated function

$G(t)$ , and then to determine the average laser intensity  $\bar{I}$ .

(4) A diffraction grating spectrograph to record the laser wavelength.

The experiment consists of a measurement of the number of ions formed,  $N_i$ , as a function of the laser intensity  $\bar{I}$  for different values of the number of longitudinal modes of the laser radiation. Experimental results are summarized in Fig. 1, which represents, on a log-log plot, the variation of the number of ions formed,  $N_i$ , as a function of the laser intensity  $\bar{I}$ , for a laser wavelength  $\lambda = 10\,603 \text{ \AA}$ , when the laser operates successively on one mode, two modes of visibility  $\nu = 0.4$ , and seven modes. It should be pointed out that the observed slope  $K_{\text{expt}} = \partial \ln N_i / \partial \ln \bar{I} = 22 \pm 3$  is found to be higher than  $K_0 = 11$ . A previous experiment<sup>10</sup> has shown that  $K_{\text{expt}}$  can have values smaller or higher than  $K_0$ , depending on the laser wavelength, when there is a quasi-resonant multiphoton excitation of an atomic level. Here, quasiresonant ten-photon excitations of four  $8d$  states of xenon atoms are possible in a wavelength range of  $10 \text{ \AA}$  near the laser wavelength  $\lambda = 10\,603 \text{ \AA}$  used in this experiment. Moreover, the linewidth of the laser radiation is like a damping term in resonant multiphoton processes. The narrow linewidth corresponding to seven adjacent longitudinal modes of this laser is only  $5 \times 10^{-2} \text{ \AA}$ , and allows  $K_{\text{expt}}$  to have values far from  $K_0$ . An important point is that  $K_{\text{expt}}$  remains constant when the number of longitudinal modes is changed from one to seven. Figure 2 shows experimental results obtained with the laser wavelength  $\lambda = 10\,643 \text{ \AA}$ . This figure shows the variation of the number of ions formed,  $N_i$ , as a function of the laser intensity  $\bar{I}$  when the laser operates successively on one mode, two modes of visibility  $\nu = 0.6$ , seven modes, and seven modes which are locked. The observed slope is now  $K_{\text{expt}} = 11 \pm 1$  (no resonant effect appears in that case,  $K_{\text{expt}} = K_0$ ).

In both figures, a very significant enhancement of the number of ions  $N_i$  is observed when the number of modes is increased. This result has been predicted by many theoreticians.<sup>8,11,12</sup> It can be explained classically in terms of temporal peaks of the laser intensity. Figure 3 shows the temporal distribution function of the laser intensity when the laser operates in a single mode, two modes, seven modes, and seven modes which are locked. For ideal single-mode operation, no fluctuation occurs. For a two-mode laser pulse, the modulation of the temporal dis-

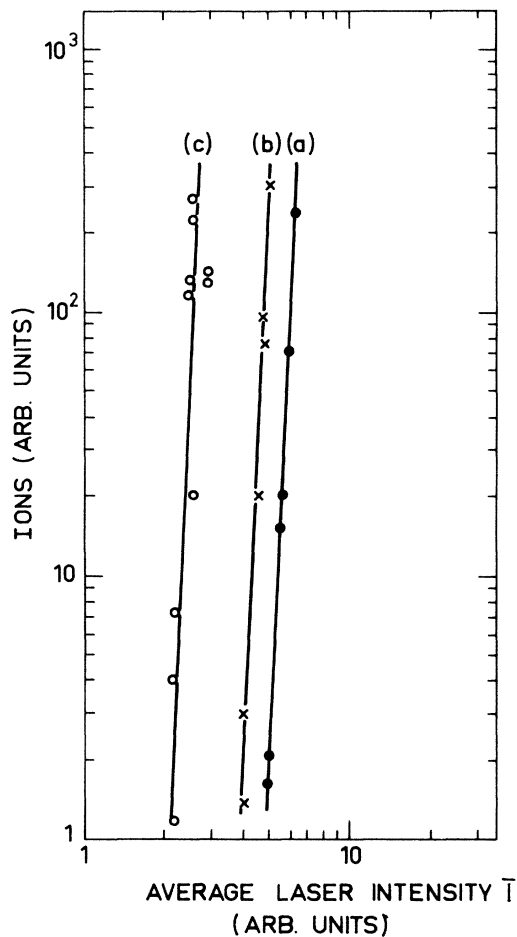


FIG. 1. Log-log plot of the variation of the number of ions  $N_i$  as a function of the average laser intensity  $\bar{I}$  (arbitrary units), when the laser operates in (a) one mode, (b) two modes with visibility  $\nu = 0.4$ , and (c) seven modes. The laser wavelength is  $\lambda = 10\,603 \text{ \AA}$ .

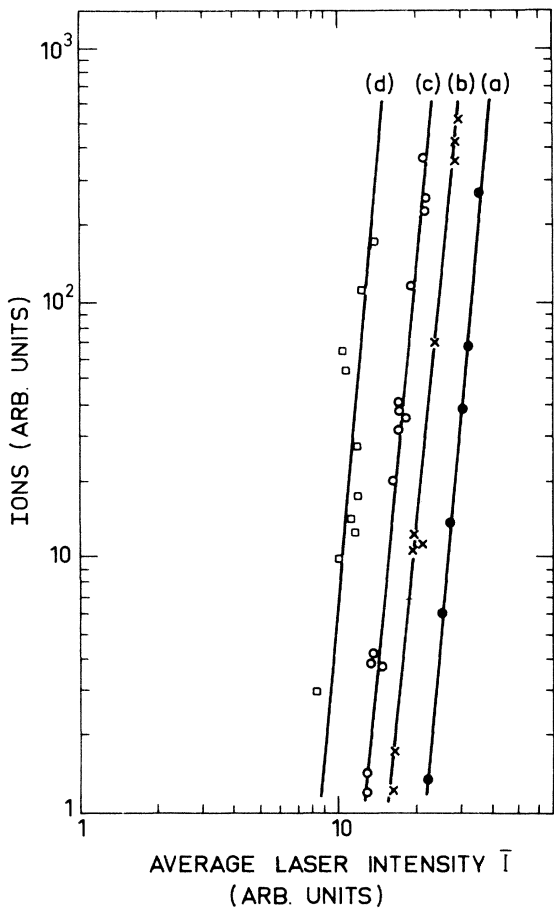


FIG. 2. Log-log plot of the variation of the number of ions  $N_i$  as a function of the average laser intensity  $\bar{I}$  (arbitrary units), when the laser operates in (a) one mode, (b) two modes with visibility  $\nu = 0.6$ , (c) seven modes, and (d) seven modes when these modes are locked. The laser wavelength is  $\lambda = 10\,643 \text{ \AA}$ .

tribution function of intensity is sinusoidal, with a period (5.7 nsec) corresponding to a round-trip time in the oscillator cavity. The modulation depth, at the highest part of the pulse, varies from shot to shot. When the laser oscillates in three, or more than three, modes, the temporal distribution function of laser intensity exhibits a quasiperiodical structure, but its shape greatly varies from shot to shot.<sup>13</sup>

Considering the experimental curves in Figs. 1 and 2, several observations can be made. First, experimental points induced by a single-mode laser pulse are perfectly lined up. The number of ions induced by a single-mode laser pulse is found to be

$$N_1 = \alpha(\lambda) I^{K(\lambda)} \quad (2)$$

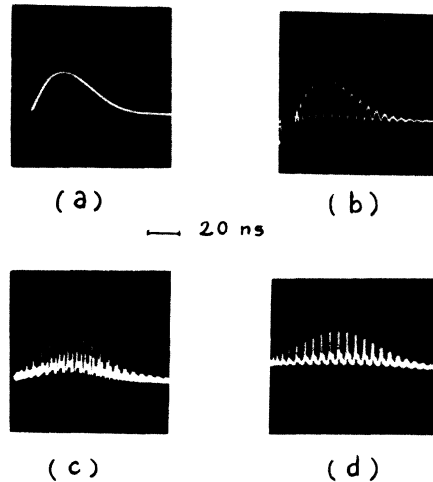


FIG. 3. Temporal distribution function of the laser intensity when the laser operates in (a) one mode, (b) two modes with visibility  $\nu = 0.8$ , (c) seven modes, and (d) seven modes when these modes are locked (sub-pulses are due to electrical ringing in detector circuit).

in the laser intensity range used in the present experiment. The second observation concerns experimental results obtained with a multimode laser pulse. Experimental points are scattered. This scattering is due to the variation in both phases and relative intensities of modes from one laser shot to another one. The number of ions induced by a multimode laser pulse is found to be

$$N_i = N_1 g^{(k)} \quad (3)$$

for the same average intensity  $\bar{I}$ . This factor  $g^{(k)}$  seems to depend only on the statistical properties of the laser light, in the laser intensity range used in this experiment, and for a given laser wavelength  $\lambda$  which determines the value of  $k$ . Table I exhibits values of  $g^{(k)}$  deduced from experimental results at the laser wavelengths  $\lambda = 10\,603$  and  $10\,643 \text{ \AA}$ , for two different laser

TABLE I. Enhancement factor  $g^{(k)}$  of the number of ions formed, normalized to unity for a single-mode laser pulse. This factor  $g^{(k)}$  is given for two different laser light patterns: a two-mode pulse, and a seven-mode pulse, at two different laser wavelengths.

No. of modes	$\lambda = 10\,603 \text{ \AA}$ $K = 22 \pm 3$	$\lambda = 10\,643 \text{ \AA}$ $K = 11 \pm 1$
2	$10^{2.1 \pm 0.3}$ ( $\nu = 0.4$ )	$10^{1.6 \pm 0.2}$ ( $\nu = 0.6$ )
7	$10^{8.2 \pm 1.7}$	$10^{2.6 \pm 0.3}$

light patterns: a two-mode pulse, and a seven-mode pulse.  $g^{(k)}$  values are given with a large error which is due to an uncertainty in the value of  $k$ , and an additional uncertainty in the position of the average straight line passing through scattered experimental points as shown in Figs. 1(c) and 2(c). When the number of modes becomes increasingly large, the value of the  $g^{(k)}$  has been calculated to be  $K!$ ,<sup>8,12</sup> i.e.,  $10^{7.6}$  for  $K = 11$ , and  $10^{20.8}$  for  $K = 22$ .

The third point is relative to the influence of mode locking of seven modes of the laser pulse. This experiment was carried out by placing in the oscillator cavity a solvent cell containing a dye to phase lock the seven modes [Fig. 2(d)], and without the dye [Fig. 2(c)]. The number of ions is found to be enhanced by a factor of  $10^2$  when the seven modes have the same phase in comparison with the same average laser intensity when the seven modes have random phase from 0 to  $2\pi$ . Thus the locking of modes by the dye very significantly changes the statistical properties of the laser pulse, and increases the temporal peaks of the laser intensity.

In conclusion, it seems that the multiphoton ionization probability for xenon atoms with a Nd-glass laser pulse is  $g^{(k)}$  times as large as that obtained when the excitation source is an ideal

single-mode laser pulse. Thus, in comparing calculated and experimental multiphoton ionization probabilities, a  $g^{(k)}$  correction factor has to be applied when experimental results are obtained with a multimode laser pulse.

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## Measurement of $S$ - $P$ Coherence in the Beam-Foil-Excited $n = 2$ State of Atomic Hydrogen\*

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The intensity of beam-foil-excited Ly- $\alpha$  radiation has been measured in electric fields alternately parallel and antiparallel to the beam. Measurements were made at three energies—110, 165, and 210 keV—and three field strengths. From these data the  $S$ - $P$  coherence term  $\sigma_{SP}$  has been determined. The magnitude of  $\sigma_{SP}$  is found to be 0.07 at 110 keV, 0.19 at 160 keV, and 0.22 at 210 keV. These results establish that beam-foil-excited hydrogen atoms have a nonzero electric dipole moment.

Since the observation by Bashkin and co-workers<sup>1,2</sup> and by Sellin *et al.*<sup>3</sup> of electric-field-induced modulations in the time decay of beam-foil-excited hydrogen atoms, the study of modulated decays has proved to be of considerable importance in the field of beam-foil spectroscopy. In particular, the discovery of zero-field quantum beats by Andrä,<sup>4</sup> indicating that foil-excited atoms are frequently aligned, prepared the way for the introduction of a large number of high-

resolution techniques into the field of beam-foil spectroscopy. Essentially, the excited atoms must possess a nonzero quadrupole moment  $\langle 3L_z^2 - L^2 \rangle$ , where  $\vec{L}$  is the orbital angular momentum, in order to observe the zero-field beats. Measurable modulations have now been observed for a variety of beam-foil-excited transitions in light atoms, and the coherence implied by  $\langle 3L_z^2 - L^2 \rangle \neq 0$  is now well established as a significant feature of the excitation process.

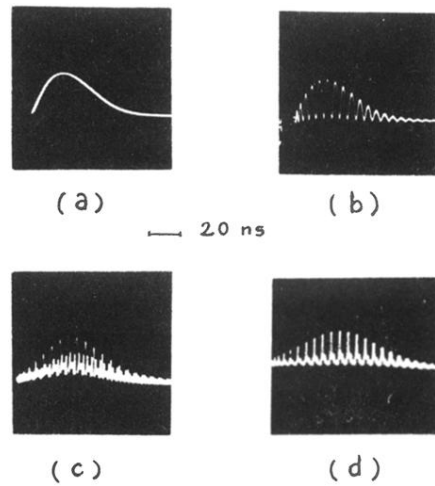


FIG. 3. Temporal distribution function of the laser intensity when the laser operates in (a) one mode, (b) two modes with visibility  $v = 0.8$ , (c) seven modes, and (d) seven modes when these modes are locked (sub-pulses are due to electrical ringing in detector circuit).