# 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 number of longitudinal modes variable from 1 to 100, has been used to investigate laser temporal-coherence effects on the 11-photon ionization of xenon atoms. When the laser pulse has an over-all bandwidth of 4 $cm^{-1}$, the number of ions formed is enhanced by $10^{6.9 \pm 0.3}$ over that produced by a monomode laser pulse with the same average laser intensity. The observed enhancement in the number of ions due to the change in temporal coherence of the pulse is in good agreement with recent theoretical prediction.

## I. INTRODUCTION

Multiphoton ionization processes have been the subject of a considerable amount of recent theoretical and experimental works.[1-5] A significant disagreement has been observed between experimental and calculated multiphoton ionization probabilities of atoms in the past few years by several authors. Thus the experimental value of the six-photon ionization probability of atomic hydrogen has been found to be greater by three orders of magnitude than the corresponding probability calculated by different authors.[1] This apparent discrepancy can be explained in terms of coherence of the laser radiation.

A previous experiment has demonstrated the influence of laser temporal-coherence effects on multiphoton ionization of xenon atoms when the number of adjacent longitudinal modes is varied from one to seven.[6] The purpose of the present paper is to extend this investigation by increasing the number of longitudinal modes of the laser pulse up to about 100, corresponding to an over-all bandwidth of a few inverse centimeters.[7] This corresponds to the experimental conditions with which most multiphoton ionization experiments have been performed in the past few years with Nd-glass lasers.

It should be pointed out that the theoretical calculations of the multiphoton ionization probability of atoms assume the laser radiation to be a single-mode laser source. However, when the laser radiation contains numerous longitudinal modes (off-axial modes are not considered here), the interaction between these modes can modulate the temporal laser pulse. The laser intensity becomes more and more irregularly fluctuating as the number of longitudinal modes increases. Multiphoton ionization processes are very sensitive to the transient fields of large amplitude of the laser intensity, since the $K$-photon ionization probability is proportional to the $K$ power of the instantaneous laser intensity. It is the purpose of this paper to investigate the influence of the fluctuations of the laser intensity on multiphoton ionization processes by varying the mode-structure of the laser pulse.

## II. DEFINITIONS AND OBJECTIVE OF EXPERIMENT

The instantaneous laser intensity seen by atoms can be expressed in the form

$$I = \bar{I}_M G(t) i(t), \quad (2.1)$$

where $\bar{I}_M$ is the maximum time-averaged intensity. $G(t)$ is the normalized temporal distribution function of the laser intensity; this function is quasi-reproducible from one laser shot to another, and its duration is about $10^{-8}$ sec for a $Q$-switched laser pulse. $i(t)$ is a periodic function which fluctuates from shot to shot. It has a stochastic pattern which depends on both phases and relative amplitudes of the modes at the top of the laser pulse and corresponds to short-term peak intensities, as shown in Fig. 1. This figure gives some temporal laser pulses with corresponding spectral bandwidths. The duration of these peak intensities $\Delta t$ is the coherence time of the laser pulse. It is the characteristic time of the most rapid variations of the laser intensity. It gets shorter and shorter as the number of modes increases, since $\Delta t = \Delta\nu^{-1}$, where $\Delta\nu$ is the spectral bandwidth of the laser pulse. Two adjacent longitudinal modes are separated by $1/2L$, where $L$ is the length of the oscillator cavity, i.e., $6\times10^{-3}$ $cm^{-1}$ for $L = 85$ cm, with the laser used in the present experiment. Thus when this laser operates in ten adjacent modes $\Delta\nu = 5\times10^{-2}$ $cm^{-1}$ and the corresponding coherence time is 600 psec. The instantaneous laser intensity can be measured only if the time resolution of the experimental apparatus is better than the coherence time of the laser pulse. This is possible only when the number of modes is small. When the number of modes becomes large, the time dependence of the laser intensity is not explicitly known. This was the condition in which most multiphoton ionization experiments have been performed in the past few years with Nd-glass lasers.

Generally, only the time-averaged laser intensity is measured:

$$\bar{I} = \bar{I}_M G(t), \tag{2.2}$$

whereas multiphoton ionization of atoms is a highly nonlinear process which is very sensitive to peak-intensities functions $i(t)$, since the multiphoton ionization probability is

$$W = \alpha I^K, \tag{2.3}$$

where $\alpha$ is a factor which depends on the atom considered as well as the polarization of the laser pulse. $I$ is the instantaneous laser intensity. $K$ is the next integer greater than the ionization energy of the atom divided by the photon energy. Thus $K=11$ for ionization of xenon atoms by a Nd-glass laser pulse. This expression (2.3) is strictly valid only when the atom is not ionized through any quasiresonant intermediate excited states.[6]

We can define a peak-intensity moment $f_K$ for a $K$th-order process as

$$f_K = \langle i^K(t) \rangle, \tag{2.4}$$

where the symbol $\langle\ \rangle$ means the ensemble average over several laser shots. This peak-intensity moment $f_K$ can be related to a multiphoton ionization process. The number of ions induced by a multimode laser pulse is given by

$$N_i = \alpha \int I^K(t)\, dt. \tag{2.5}$$

The number of ions induced by a monomode laser pulse with the same average intensity would be

$$N = \alpha \int \bar{I}_M^K(t)\, dt. \tag{2.6}$$

Hence

$$f_K = \langle N_i / N \rangle. \tag{2.7}$$

Thus $f_K$ is the expectation value of the enhancement of the number of ions due to the multimode character of the laser pulse. It reflects, at constant average laser intensity, the influence of the temporal coherence of the laser light on the multiphoton ionization process. This paper investigates the relationships between the eleventh-order moment $f_{11}$ and the resultant enhancement in the number of ions formed through the 11-photon ionization of xenon atoms.

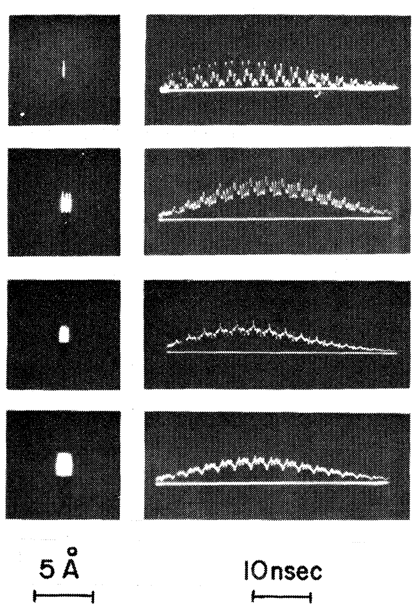


FIG. 1. Laser intensities and corresponding spectral linewidths when the Nd-glass laser operates successively, from top to bottom, in one spectral line (ten modes), three lines (30 modes), seven lines (70 modes), and ten lines (100 modes).

### III. EXPERIMENTAL METHOD

In the present experiment the multiphoton ionization probability of xenon atoms has been measured by using a single-transverse-mode $Q$-switched Nd-glass laser which can be operated with a variable number of longitudinal modes (1–100). This laser has been described in detail elsewhere.[8] The laser is $Q$ switched by a Pockels cell. The number of longitudinal modes is varied by using two-step $Q$ switching and resonant reflectors in the temperature-controlled cavity. The laser radiation is linearly polarized, and is centered at 10643 Å. This wavelength has been selected to investigate a direct 11-photon ionization of xenon atoms, i.e., without any resonant intermediate excited states.[6,9-11] This consideration is important because the enhancement factor in the number of ions is characteristic of the statistical properties of the laser pulse only when a nonresonant multiphoton ionization process occurs. Figure 2 shows a schematic diagram of the experimental arrangement. The laser beam emerges from the master oscillator, is expanded by a telescope with fourfold magnification and then enters a five-stage amplifier. The laser beam is then focused with an aspheric lens (focal length 80 mm) into a vacuum chamber into which the xenon gas has been released at a pressure of $10^{-4}$ Torr. The average laser intensity at the focal point is about $10^{12}$ W cm$^{-2}$. The xenon ions resulting from the laser interaction at the focal point are extracted with a transverse electric field of 400 $V cm^{-1}$ and then detected with a magnetic electron multiplier (Bendix type 308).

The longitudinal-mode structure of the laser pulse is controlled in the following manner. A portion of the laser beam reflected from the entrance window of the interaction chamber passes through three beam splitters, and is incident on the following.

(i) A photodiode (Radiotechnique XA 1003) with 150-psec rise time; this photodiode is directly connected to the input of a Ferisol 0Z 100B oscilloscope with 150-psec rise time and bandwidth of 2 GHz. The combined rise time of this apparatus is about 200 psec. Laser-intensity fluctuations can thus be recorded as long as the over-all spectral bandwidth $\Delta\nu$ of the laser emission is less than 0.1 $cm^{-1}$.

(ii) A photodiode and an oscilloscope whose combined rise time is several nanoseconds; this detection setup is used to determine the average laser intensity $\bar{I}_M$.

(iii) A diffraction-grating spectrograph which has a dispersion of 1.6 Å/mm and a resolution of 0.08 $cm^{-1}$ at 1.06 $\mu$m; a laser spectrum is recorded through an image converter. This apparatus is used to record laser emission spectra with a bandwidth $\Delta\nu > 0.08$ $cm^{-1}$.

(iv) A Fabry-Perot interferometer which gives information on the laser mode content when the laser oscillates on few modes; one to ten adjacent modes can be observed in its free spectral range of $7.5\times10^{-2}$ $cm^{-1}$. Fabry-Perot rings are recorded through an image converter.

The experiment consists of a measurement of the number of ions formed as a function of the average laser intensity $\bar{I}_M$ for different values of the number of longitudinal modes of the laser pulse. The experiment was performed in two stages. In the first stage the laser operated in a variable number of adjacent longitudinal modes (from one to seven) inside a single spectral line. In the second the laser operated on a variable number of spectral lines from one to ten; each spectral line contained about ten adjacent modes.

## IV. EXPERIMENTAL RESULTS AND DISCUSSION

### A. Single-line laser

Experimental results obtained when the number of longitudinal modes is varied from one to seven inside a single line have been published earlier[6] and will only be discussed here.

When the laser operates in two adjacent modes, the modulation of the temporal laser pulse is sinusoidal with a period $T = 5.7$ nsec corresponding to a round-trip time in the oscillator cavity. The visibility of the fringe pattern is defined as

$$v = (I_{\max} - I_{\min}) / (I_{\max} + I_{\min}). \tag{4.1}$$

The modulation depth, at the highest part of the pulse, varies from shot to shot, depending on the relative intensities of the two modes. A two-mode pulse is a special case for which the phase of the two modes does not play any role. The number of ions induced by a two-mode laser pulse with visibility $v = 0.6$ has been found to be $10^{1.6\pm0.2}$ times larger than those induced by a single-mode pulse of equal average intensity $\bar{I}_M$. The number of ions is proportional to the $K$th power of the instantaneous laser intensity $I$ according to relation (2.5). This instantaneous laser intensity can be accurately recorded for a two-mode pulse, and is given by

$$I(t) = 1 + v\cos(2\pi t/T). \tag{4.2}$$

The experimental enhancement factor $10^{1.6\pm0.2}$ in the number of ions corresponds to the $K$th-order moment $f_K$ defined by (2.4). For a two-mode laser pulse

$$f_K = T^{-1}\int_0^T [1 + v\cos(2\pi t/T)]^K\, dt. \tag{4.3}$$

With $k = 11$ and $v = 0.6$, this expression gives $f_{11} = 10^{1.52}$, which is in good agreement with the measured value $10^{1.6\pm0.2}$.

When the laser oscillates in three or more than three modes, the laser intensity exhibits a quasiperiodical structure, but its shape greatly varies from shot to shot, depending mainly on the phases

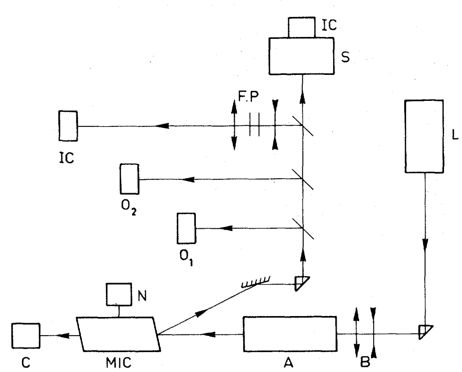


FIG. 2. Schematic diagram of the experimental setup. L: laser oscillator; B: beam-expanding telescope; A: amplifiers; MIC: multiphoton ionization chamber; N: measurement of the multiphoton ions; C: calorimeter; $O_1$: temporal laser-pulse measurement with about 200-psec rise time; $O_2$: temporal laser-pulse measurement with several-nanoseconds rise time; FP: Fabry-Perot interferometer; S: grating spectrograph; IC: image converter.

of the modes. The intensities of the modes can be measured, but no information on their relative phases can be obtained. Statistical considerations are then needed to interpret experimental results obtained with a multimode laser pulse. Most of the authors suppose that the phases are statistically independent and have a random distribution within the interval $(0, 2\pi)$. A simple expression of $f_K$ can be derived by assuming that $M$ modes have independent phases, and amplitudes $\mathcal{A}_m$ which obey a Gaussian law, i.e., the intensity $I_m = |\mathcal{A}_m|^2$ of the $m$th mode follows an exponential distribution law given by

$$P(I_m) = e^{-I_m}. \tag{4.4}$$

The instantaneous laser intensity is

$$I = \left|\sum_{m=1}^{M} \mathcal{A}_m\right|^2, \tag{4.5}$$

while the time-averaged intensity $\bar{I}_M$ is given by

$$\bar{I}_M = \sum_m |\mathcal{A}_m|^2 = \sum_m I_m. \tag{4.6}$$

$f_K$ is defined by

$$f_K = \langle (I/\bar{I}_M)^K \rangle . \tag{4.7}$$

The special properties of the Gaussian law allow us to write

$$f_K = \langle I^K \rangle / \langle \bar{I}_M^K \rangle . \tag{4.8}$$

Equations (4.4) and (4.6) give

$$\langle \bar{I}_M^K \rangle = (K + M - 1)!/(M - 1)! . \tag{4.9}$$

It is easy to show that

$$\langle I^K \rangle = K!\, M^K. \tag{4.10}$$

We obtain from (4.8)–(4.10),

$$f_K = K!\,[M^K (M-1)!/(K+M-1)!]. \tag{4.11}$$

This relation characterizes the mode-number dependence of $f_K$ for phase-independent modes. It should be pointed out that a previous calculation has been performed for $K < 7$; it gave polynomial expressions.[12] For seven adjacent modes and $K = 11$, expression (4.11) gives $f_{11} = 10^{5.2}$, which is much greater than the experimental value $10^{2.6 \pm 0.3}$ corresponding to the enhancement in the number of ions induced by a seven mode pulse compared to that induced by a single-mode pulse of the same average intensity $\bar{I}_M$. This large difference cannot be explained in terms of different intensities of the modes. The present experiments have shown that $f_K$ is relatively insensitive to the distribution of the intensities of the modes. However, the distribution of the phases is the determining factor. The assumption of statistical independence of the phases in a $Q$-switched Nd-glass laser is not obvious, and call for additional investigations.[13] Several adjacent modes may have definite phase relationships that tend to decrease $f_K$. Such correlation properties of the phases can be taken into account by introducing an effective number of modes $\bar{M}$ which would have equal intensities and independent phases. This assumption is supported by an investigation of the statistical properties of the laser modes deduced from direct measurements on temporal laser pulse patterns.[14] The preceding expression (4.11) can then be rewritten

$$f_K = K!\,[\bar{M}^K (\bar{M}-1)!/(K+\bar{M}-1)!] . \tag{4.12}$$

By introducing the effective number $\bar{M} = 2$ in this expression (4.12), $f_K$ is calculated to be $10^{2.3}$, in good agreement with the experimental value $10^{2.6 \pm 0.3}$ obtained with a seven mode laser pulse.

### B. Multiline laser

In order to obtain a larger number of modes, we have changed the previous laser cavity used to obtain one to seven modes inside a single spectral line.[8] The resonant reflector has been replaced by a nonselective mirror, the 3-cm-thick interferometer has been removed, and the 10-mm-thick quartz plate has been replaced by a 1-mm-thick Brewster tilted-calcite birefringent filter. When this is done, the laser spectrum consists of a set of sharp lines equally spaced, with a period $\delta\nu$ mainly determined by the thickness of the laser mirrors. $\delta\nu$ equals 0.42 $cm^{-1}$ for the laser used. Each spectral line has a width $\leq 7 \times 10^{-2}$ $cm^{-1}$. The number of modes inside one of these spectral lines has been measured by using a narrow-band interference filter to isolate one of the emission lines. The mode structure of this line was observed with a Fabry-Perot interferometer. The number of modes, inside a line, is found to be ten, ranging from nine to 11 from one laser shot to another. The variation of the number of spectral lines is made by rotating the calcite plate around its geometrical axis. Up to ten lines, that is, 100 modes, have been obtained, giving an over-all laser spectral bandwidth of 4 $cm^{-1}$, that is, a coherence time of 5 psec.

The experiment consists of a measurement of the number of ions produced, $N_i$, as a function of the average laser intensity $\bar{I}_M$ for different values of the number of laser spectral lines. Experimental results are summarized in Fig. 3, which represents on a log-log plot the variation of the number of ions formed, $N_i$, as a function of the average laser intensity $\bar{I}_M$ when the laser operates successively on one line (10 modes), three lines (30 modes), seven lines (70 modes), and ten lines (100 modes). The reference curve (dashed line) corresponds to a previous result obtained with a monomode laser pulse. Experimental points induced by a single-mode laser pulse are perfectly lined up, while experimental points obtained with a multimode laser pulse are scattered. This scatter is due to the variation of both phases and relative intensities of modes from one laser shot to another.

It is important to measure the slope $K = \partial \ln N_i / \partial \ln \bar{I}_M = 11 \pm 1$. When there is no resonant effect, $K$ is the next integer greater than the ionization energy of the xenon atom divided by the photon energy. The slope $K=11$ remains constant when the number of laser lines is changed from one to ten. Thus the eleventh-order moment $f_{11}$ can be directly determined from the enhancement of the number of ions which is observed when the number of laser lines $L$ is increased. Table I sums up the $f_{11}$ experimental values for different number of modes ranging from one to 100. Experimental results obtained with a large number of modes (10–100) appear more explicitly in Fig. 4. This figure gives the experimental values of the $f_{11}$ moment as a function of the number of spectral lines $L$ and (short dashed lines) two curves which have been calculated with the following considerations. Phases of adjacent modes inside a spectral line seem to be correlated in some manner such that $f_K$ decreases, while modes of different spectral lines can be considered as having independent phases. The peak-intensity moment of a spectrum consisting of $L$ lines depends on the correlation properties of the $M$ modes inside a single line. The $K$th-order correlation properties of these $M$ modes are taken into account here by replacing the number $M$ of real modes which have correlated phases by $\bar{M}$, which represents an effective number of independent modes of equal intensity. The over-all peak-intensity moment, with these considerations, is approximated by

$$f_K = K!\,[(L\bar{M})^K (L\bar{M}-1)!/(K+L\bar{M}-1)!]\,, \qquad (4.13)$$

deduced from (4.12) by replacing $\bar{M}$ by $L\bar{M}$. In Fig. 4 experimental values obtained for $f_{11}$ fit well on the calculated curve deduced from (4.13) with $\bar{M}=3$, but do not fit on the curve calculated with $\bar{M}=10$, as it should if the ten modes oscillating in a spectral line had independent phases.

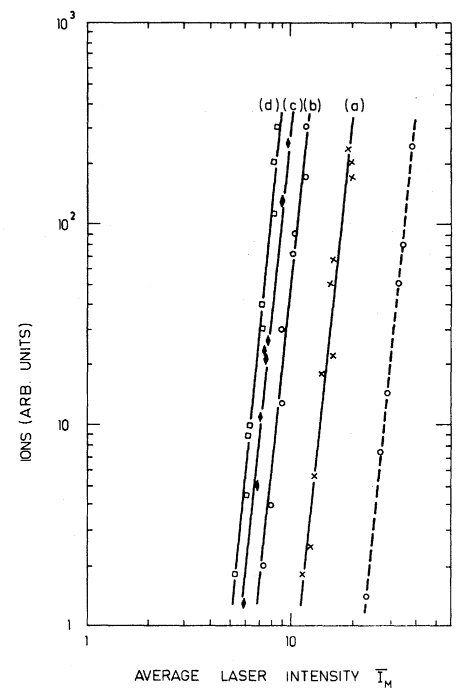


FIG. 3. Log-log plot of the variation of the number of ions as a function of the average laser intensity $\bar{I}_M$ (arbitrary units), when the laser operates in (a) one spectral line (ten modes), (b) three lines (30 modes), (c) seven lines (70 modes), and (d) ten lines (100 modes). Dashed curve corresponds to a single-mode laser operation as given in Ref. 6.

Figure 4 shows that the experimental value $f_{11}$ equals $10^{6.9\pm0.3}$ when the emission spectrum of the laser pulse consists of ten spectral lines, that is, 100 modes, which gives an over-all bandwidth of 4 $cm^{-1}$ and a coherence time of 5 psec. $f_{11}$ tends slowly towards an asymptotic value for large $L$. In the limit of an infinite number of independent modes, this asymptotic value has been calculated to be $K!$,[15–17] that is, $4\times10^7$ for $K=11$. This asymptotic value can be simply explained. When the laser oscillates in a very large number of modes there will be a high probability that, for any pair of modes with a beat frequency $f$, there will be another pair of modes with the same beat frequency but in phase opposition. Therefore, the random character of the phase distribution will

TABLE I. Enhancement factor $f_{11}$ of the number of ions formed, normalized to unity for a single-mode laser pulse.

| Number of modes | $f_{11}$ |
|---|---|
| 1 | 1 |
| 2 (visibility $v=0.6$) | $10^{1.6\pm0.2}$ |
| 7 | $10^{2.6\pm0.3}$ |
| 10 (1 line) | $10^{3.1\pm0.3}$ |
| 30 (3 lines) | $10^{5.6\pm0.3}$ |
| 70 (7 lines) | $10^{6.5\pm0.3}$ |
| 100 (10 lines) | $10^{6.9\pm0.3}$ |

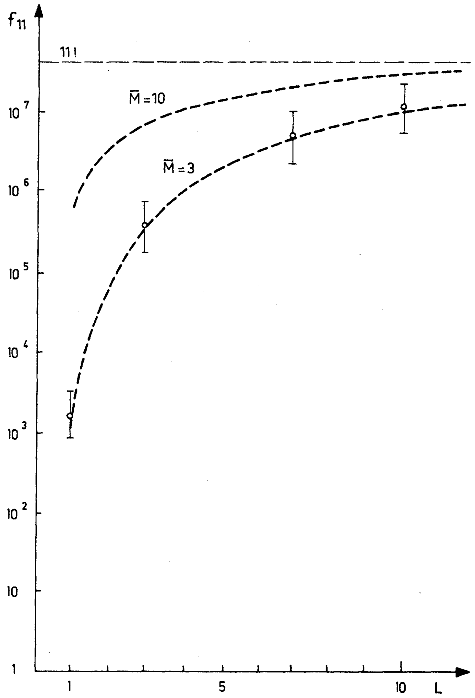


FIG. 4. Eleventh-peak intensity moment $f_{11}$ as a function of the number of spectral lines $L$, each line containing ten modes. The calculated dashed curves are deduced from Eq. (4.13) for $\bar{M}=3$ and $\bar{M}=10$.

tend to smooth out the beats.

Finally, we wish to emphasize the content of this experiment. We have studied here the relative efficiency of 11-photon ionization of xenon atoms by varying the number of laser longitudinal modes from one to 100, that is, a decrease of the coherence time from 40 nsec to 5 psec. But neither the case of single-mode pulse nor that of 100-mode pulse can be considered to be an ideal case. A single-mode pulse cannot be treated exactly as a pure coherent state in the Glauber sense; the laser used does not have a well-stabilized amplitude. On the other hand, a 100-mode laser pulse cannot be considered as being fully Gaussian as a chaotic light.

## V. CONCLUSION

The dependence of the 11-photon ionization probability of xenon atoms on the statistical properties of a $Q$-switched Nd-glass laser pulse has been investigated by varying the number of longitudinal modes from one to 100. The number of multiphoton ions is drastically enhanced when the statistical properties of the multimode laser pulse deviate more and more markedly from those of a single-mode pulse by progressively increasing the number of modes and therefore decreasing the coherence time of the laser pulse. For instance, when the laser spectrum consists of ten spectral lines, i.e., 100 modes, corresponding to an overall bandwidth of 4 $cm^{-1}$ and a coherence time of 5 psec, the number of ions is $10^{6.9\pm0.3}$ times larger than that induced with a single-mode laser pulse of the same average intensity. Most of the Nd-glass lasers which have been used in multiphoton ionization experiments in the past few years had spectral bandwidths of several inverse centimeters. Thus to do a meaningful check of experimental multiphoton ionization probabilities with corresponding theoretical data, a very significant correction factor has to be applied for experimental results obtained with a multimode laser pulse. It is highly desirable in the future that multiphoton ionization probability measurements be done, whenever possible, with a monomode laser. If this is not feasible, great care should be taken in using the correction factor due to coherence. This correction factor depends both on the order $K$ of the nonlinear interaction and on the laser spectral bandwidth, ranging from 1 to $K!$ depending on the number of modes.

Conversely, multiphoton ionization processes allow us to consider an atom under excitation by a laser pulse as an ideal detector of photons concerning the coherence of a laser pulse. The knowledge of the $K$th-order moment of the laser peak intensity is of special interest to characterize a laser field with respect to its nonlinear interaction with atoms.

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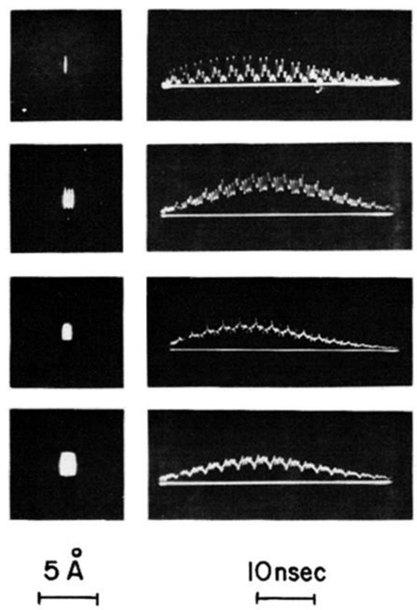


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