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Contribution of above-threshold ionization to the total ion yield for xenon at 0.53 and 1.06 μm

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The relative cross sections for 6-, $(6+1)$-, and $(6+2)$-photon above-threshold ionization of xenon at 0.53 μm are experimentally determined. The influence of these transitions on the total ion yield is discussed for different intensity regimes, at 0.53 and 1.06 μm.

In preliminary work it has been shown that, for the case of multiphoton ionization of xenon, the energy spectrum of the outgoing electrons displays several peaks with an energy separation equal to the photon energy $\hbar\omega$.[1,2] The physical process responsible for this spectrum was shown to be a multiphoton "above-threshold ionization" (ATI); i.e., when the minimum N-photon ionization produces electrons with a kinetic energy E_0, the atom can be ionized by absorption of $N+S$ photons and eject one electron with an energy $E_0+S\hbar\omega$. This model was then confirmed and further investigated in other experiments.[3–5] The experimental results can be summarized as follows. First, at 0.53 μm ($N=6$), peaks corresponding to S up to 4 have been detected with laser intensity of 1.6×10^{11} W cm^{-2}. Second, at 1.06 μm ($N=11$) and intensities between 10^{12} and 10^{13} W cm^{-2}, S values up to 10 have been observed. Furthermore, it was shown that $W_{N,S}$ the probability per unit time, of absorbing S extra photons in the continuum, is proportional to the laser intensity raised to the power $N+S$, as expected from lowest-order perturbation theory. That is, $W_{N,S}$ is given by

$$W_{N,S}=\sigma_{N,S}I^{N+S}\ , \qquad (1)$$

where $\sigma_{N,S}$ is a generalized cross section and I the laser intensity. From (1), it can be easily deduced that the total ionization rate W should be given by

$$W=\sum_{S=0}^{\infty}W_{N,S}=\sum_{S=0}^{\infty}\sigma_{N,S}I^{N+S}\ . \qquad (2)$$

On the other hand, older experiments,[6] based on measurements of the ion yield, have shown that W depends on I according to

$$W=\sigma_N I^N \qquad (3)$$

with excellent precision. For instance, at 1.06 μm and intensities up to 1.5×10^{13} W cm^{-2}, the experimental value of N was found to be 11 ± 0.2.[6] So far, no ion measurement has been able to show any deviation from the law (3) (at least far from resonances and saturation), in apparent contradiction with the conclusion drawn from electron energy measurements and summarized by Eq. (2).

The aim of this paper is to investigate in more detail this apparently contradictory experimental situation which was previously noted in Refs. 4 and 5.

First, let us note that (3) is the first term of the expansion (2), and that (2) and (3) lead to different results only if the higher-order terms in (2) are important. It is therefore necessary to know at least the relative values of the coefficients $\sigma_{N,S}$. Second, since Eq. (3) is valid only far from saturation, one must also know how close to saturation the experiment was performed. These are the two key factors in understanding the contradiction mentioned above. We report here the values of $\sigma_{N,S}$ measured at 0.53 and 1.06 μm with a pulsed Nd-YAG laser. The experimental setup has been described in Refs. 3 and 4.

(a) At 0.53 μm, the photoelectron energy spectrum consists of several peaks corresponding to $S=0,1,2,3$. The intensity dependence of the three main ones is shown in Fig. 1. The points are experimental data; the solid lines are the result of a numerical integration of rate equations governing the motion of atomic populations in the ground state and the final state in the continuum. The validity of using the rate equation for ATI was established in Ref. 7. Here we have assumed partial rates of the form (1) and a Gaussian pulse shape. The integration was carried out for different values of the intensity and a given set of coefficients $\sigma_{N,S}$. The best set of coefficients was then found by trial and error and the intensity parameter matched to the maximum value used in the experiment, 1.6×10^{11} W cm^{-2}. The resulting saturation intensity derived from the calculation is 10^{11} W cm^{-2}. The relative values of the $\sigma_{N,S}$ obtained from the fit are

$$\frac{\sigma_{6,0}}{\sigma_{6,1}}=2.57\ 10^{11}\ \mathrm{W\,cm^{-2}},$$

$$\frac{\sigma_{6,1}}{\sigma_{6,2}}=5.26\ 10^{11}\ \mathrm{W\,cm^{-2}}\ .$$

Note that these ratios have the dimensions of intensi-

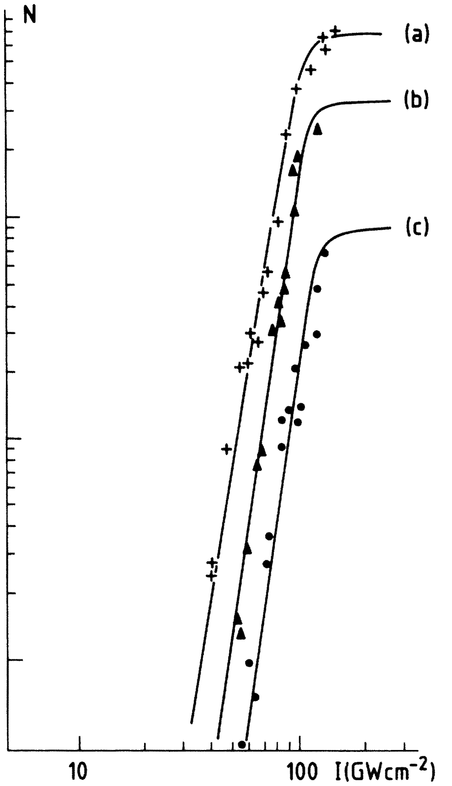

FIG. 1. Total number of counts per laser shot for photon energy equal to 2.34 eV ($N=6$) for Xe ($3P_{3/2}$) lines of the electron spectra as a function of laser intensity in doubly logarithmic scale. (a) Line $S=0$; (b) line $S=1$; (c) line $S=2$. We have reported only the $\frac{3}{2}$ configuration to clarify the figure. Solid lines are calculated (see text).

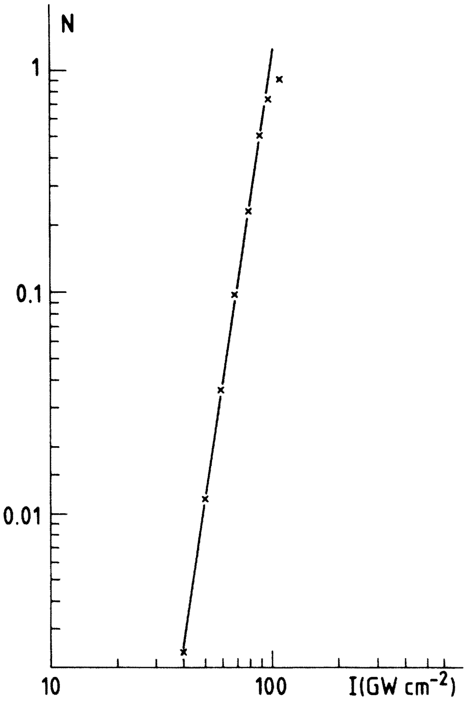

FIG. 2. Total number of counts obtained by adding counts from each line of electron spectra as functions of laser intensity in doubly logarithmic scale. The straight line is calculated (see text).

ty and are nothing but the "threshold intensity" defined by Gontier and Trahin in Ref. 8. With I expressed in GW cm^{-2} the expansion (2) can be written

$$W=\sigma_{6,0}I^6(1+3.8\times10^{-3}I+7\times10^{-6}I^2+\cdots)\ . \qquad (4)$$

This law holds for intensities up to about 100 GW cm^{-2}. Figure 2 shows the total electron signal (obtained by summing all the electron counts), as a function of the intensity. The "straight" line is derived from (4). The measured slope is 6.1 ±0.3 and it is clear that a slope measurement of the ion signal would require quite high precision to detect ATI at 0.53 μm. For intensities larger than 100 GW cm^{-2}, the higher-order terms in (4) would significantly modify the slope but saturation would mask this effect and, again, prevent detection of ATI.

(b) At 1.06 μm, the photoelectron spectrum displays a series of peaks, resulting from up to seven-photon[4] and ten-photon[5] ATI. As shown in Refs. 4 and 5, the interpretation of such spectra is complicated because of the core-fine structure splitting, but this turns out to be of minor importance for the following discussion.

Taking the relative values of the higher-order terms from Ref. 4 the total ionization probability can be written (I in TW cm^{-2})

$$W=\sigma_{11,0}I^{11}(1+9.6\times10^{-2}I+7\times10^{-3}I^2+5.5\times10^{-4}I^3+4.8\times10^{-5}I^4+3.6\times10^{-6}I^5)\ . \qquad (5)$$

However, there is evidence in Refs. 4 and 5 that the experiment was carried out above the saturation intensity I_{sat}. Assuming that (5) holds up to I_{sat}, the slope between 0.1 I_{sat} and I_{sat} is found to increase from 11.1 to 12.4. This is to be compared with the measurement[6] 11.0 ±0.2 for an intensity between 1.0 and 1.5×10^{13} W cm^{-2}. The difficulty here is to know how much above I_{sat} the experiments of Refs. 4 and 5 were performed (since the intensity was only estimated in these works) and how much under I_{sat} the work of Ref. 6 was performed (since the saturation was not observed, even for the highest intensity reported therein, i.e., 1.5×10^{13} W cm^{-2} for 30-ps pulses, which corresponds to 8.5×10^{12} W cm^{-2} for 20-ns pulses). To make the results of Ref. 6 compatible with (5), one has to conclude that the experiment (Ref. 6) was performed with an intensity smaller than 0.3 I_{sat}. This brings I_{sat} up to 2.8×10^{13} W cm^{-2} for 20-ns pulses; that is sensibly above the estimation of Refs. 4 and 5. However, it should be remembered that, especially in the case of high-intensity picosecond pulses, measurements of absolute laser intensities are extremely difficult, so that a precision better than 40% cannot seriously be considered. Taking into account the fact that the intensities used in ATI experiments so far have been only estimated, the results given by the two kinds of experiments can be considered as compatible.

Moreover, recent experiments have been reported[9] where the dependence of ATI spectra of Xe towards the laser intensity for Xe and 1.06 μm. In these experiments, it was carefully checked that the laser intensity was below I_{sat}. Under these conditions, it was found that the amplitude of each peak of the spectrum was varying as I^k with $10 < k < 11$. This is in agreement with the results of Ref. 6 but does not agree with the predictions of lowest-order perturbation theory.

In summary, we have shown, by measuring the relative values of ATI cross sections, that, at 0.53 μm, the accuracy of ion measurements is insufficient to detect ATI. At 1.06 μm, the situation seems more intricate even if there is no true incompatibility between the different experimental results available. It seems that Xe at 1.06 μm is one of the cases where the threshold intensity of Gontier and Trahin[8] is lower than the saturation intensity, and that more theoretical work is necessary to reach a full understanding of this situation.

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