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Angular distribution of photoelectrons from above-threshold ionization of Xe

H. J. Humpert, H. Schwier, R. Hippler, and H. O. Lutz
Fakultät für Physik, Universität Bielefeld, 4800 Bielefeld 1, Federal Republic of Germany
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The angular distribution of electrons from above-threshold multiphoton ionization (photon energy 1.17 eV) was measured. Photoelectrons are preferentially ejected along the direction of the laser's linear polarization; the distributions become narrower with increasing number of absorbed photons.

Absorption of photons above the ionization threshold in multiphoton ionization has recently received considerable attention. A minimum number (N) of photons (energy $\hbar\omega$) can lift an electron from its bound state into the continuum, thus producing a peak in the photoelectron energy spectrum; absorption of additional photons produces discrete higher-energy peaks which are separated by $\hbar\omega$.[1-6] This phenomenon of above-threshold ionization (ATI) displays interesting features. For example, it has been found[4,5] that with increasing laser intensity, the lowest-order electron-energy peak (corresponding to the absorption of N photons) disappears. If the laser intensity is increased further, the second peak also begins to disappear, etc.[5] Most of these experiments have so far been performed with Xe targets; unfortunately, no theory yet exists for multiphoton processes in such complicated atoms. A rather direct and comparatively simple way to study the ATI process experimentally is a measurement of angular distributions of the emitted photoelectrons. For example, in the simplest case of absorption of circularly polarized photons by an initial s state in the absence of spin-orbit coupling and higher-order processes, the electron angular distributions should shift by one additional angular momentum l for each photon absorbed; to our knowledge, a corresponding ATI experiment has not been performed yet. Without such restrictions, and for linear polarization, the situation is more complex since a range of final angular momentum states may be populated, intricately depending on the initial and intermediate atomic states. A qualitative argument has recently been presented by Rzazewski and Grobe[7] on the basis of a simple model[8] which views ATI as a coherent photon-by-photon absorption process through a flat continuum. For hydrogen atoms they expect only a few of the lowest partial waves in the ATI electron angular distribution, with a shift to higher l for the higher-order ATI peaks (i.e., the more additional photons are absorbed).

To put this assertion on a more quantitative basis, we have for the first time experimentally studied the angular distribution of photoelectrons in a sequence of several ATI peaks. The ATI electrons are produced by multiphoton ionization of xenon using a Nd:YAG laser. Electron-energy spectra have been recorded as a function of the angle between the linear polarization vector and the electron emission direction. Results obtained with the frequency-doubled laser line ($\lambda = 532$ nm) have already been published.[3] These angular distributions involved the lowest-order ATI peaks only. We have now also measured angular distributions of higher-order ATI photoelectrons using the fundamental line of the Nd:YAG laser ($\lambda = 1064$ nm). The electron-energy spectra are similar to those measured by Kruit, Kimman, Muller, and van der Wiel.[4] With a photon energy of 1.17 eV, 11 and 12 photons are necessary for ionization from the $^2P_{3/2}$ and $^2P_{1/2}$ state, respectively. In the energy range from 0 to 6 eV we have recorded the variation of the photoelectron emission with respect to the linear polarization direction for the first five peaks. The laser power in these measurements (50 mJ/pulse) had been adjusted so that the intensity of the first peak was about one-third of the intensity of the second peak (measured at 0°). As an example, Fig. 1 shows the intensity of photoelectrons detected at 0°, as a function of energy. Note that each peak is the result of two ionization channels since the energy difference (0.15 eV) of electrons resulting from ionization of the $^2P_{3/2}$ state (after absorption of N photons) and from the $^2P_{1/2}$ state (after absorption of $N+1$ photons) cannot be resolved in our experiment.

Figure 2(a) displays the angular distributions for the first five peaks. The circles represent the integrated intensities of the measured spectra and the solid line is a fit to the data with a sum of Legendre polynomials of order $2k$,

$$P(\theta) = \sum_k b_{2k} P_{2k}(\cos\theta) \quad ,$$

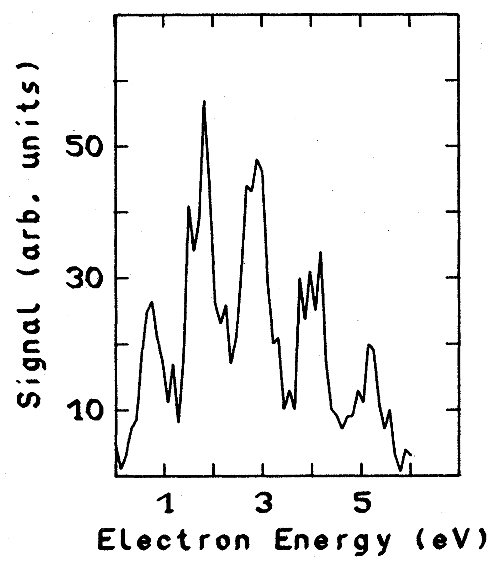

FIG. 1. Energy dependence of photoelectron intensity detected at 0° relative to the laser light polarization direction ($\lambda = 1064$ nm). Laser pulse energy is 50 mJ, corresponding to an effective intensity (Ref. 4) of roughly 10^{13} W/cm^2.

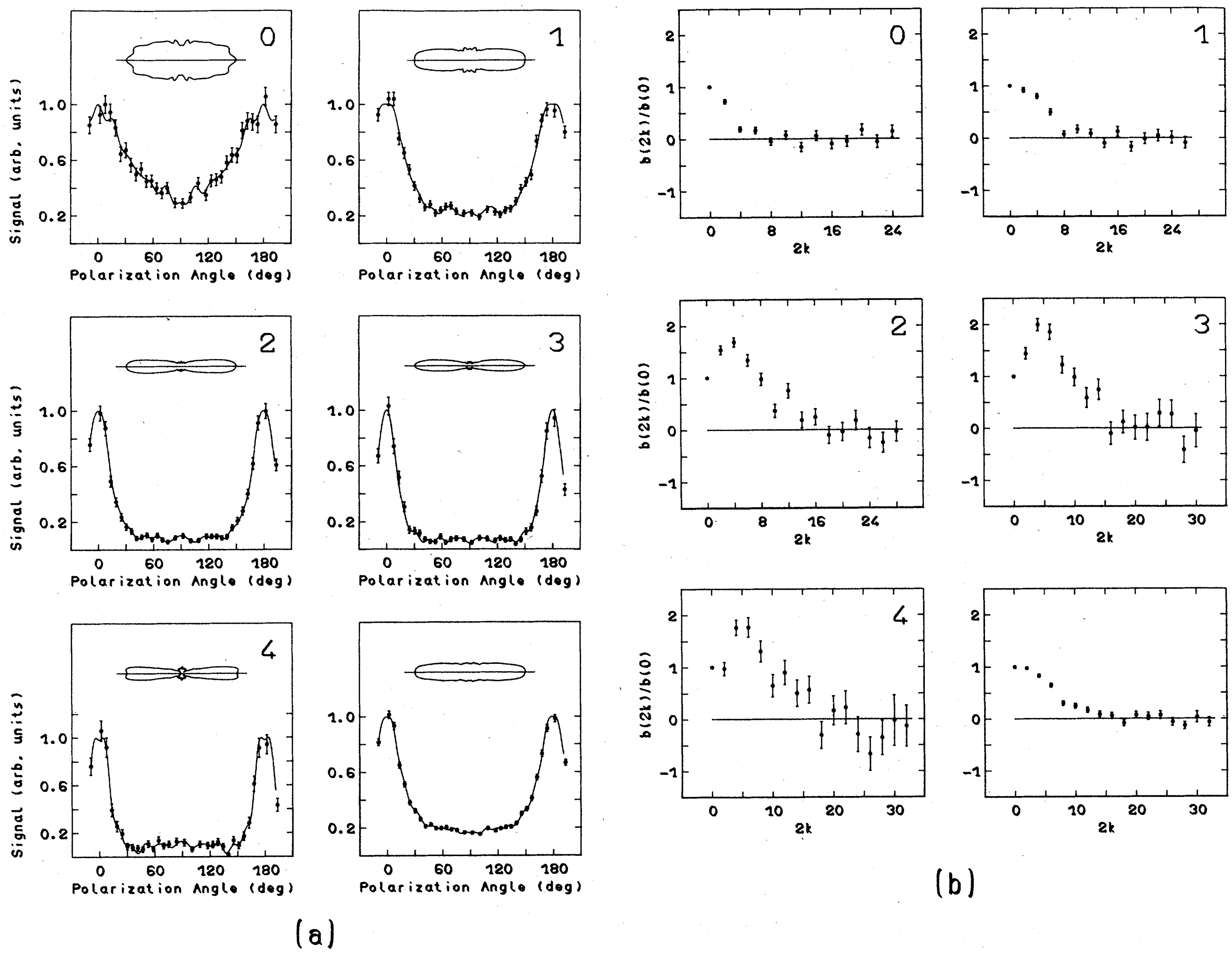

FIG. 2. (a) Angular distribution of the five transitions shown in Fig. 1. The lowest-order process (0) consists of an 11-photon absorption ($^2P_{3/2}$) and a 12-photon absorption ($^2P_{1/2}$). The higher-order processes (1, . . . , 4) refer to absorption of additional 1, . . . , 4 photons. The last figure in the sequence shows the total intensity (sum of all distributions). The curves are fits to a sum of Legendre polynomials. (b) Amplitudes b_{2k} of the Legendre polynomials fitted to the experimental data.

with $0 \leq k \leq L$. L is the maximum angular momentum of the system, i.e., the number of absorbed photons.[9] Photoelectron emission is peaked at 0° and 180°. As can be seen, the measured anisotropy (reflected by the sharpness of the angular distribution) increases with the number of additional photons absorbed.

This behavior is also displayed by the coefficients b_{2k} which are plotted in Fig. 2(b) as a function of order k. Just above threshold, only the lowest k values contribute. With increasing number of absorbed photons higher-order terms become more important.

Our results agree with what has qualitatively been expected from the above model[7,8] although these calculations should not be used to explain the details of the distribution. More quantitatively, Dulcic and Eberly[10] have recently performed calculations of the ATI process in Xe for $\lambda = 532$ nm. For not too high laser intensities their results corroborate the expectation of an increasing sharpness of the angular distributions with the number of photons absorbed in the continuum. However, the results also indicate a pronounced sensitivity of the distributions to the laser intensity above about 10^{13} W/cm^2. Evidently, more detailed calculations for the Xe atom case and experiments such as the one reported here but involving more simply structured target atoms are needed to clear up the ATI process.

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[9]$P(\theta)$ may be written as a single sum of Legendre polynomials even though each electron-energy peak is composed of two ionization channels. The problem lies, of course, in decomposing the fit into partial waves.

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