Cross sections for multiphoton ionization of alkali metal atoms

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The direct process of two-, three-, four-, and five-photon ionization of alkali metal atoms is investigated. In all cases the relation $W = \alpha_{k_0} F^{k_0}$ is satisfied, where W is the ionization probability, F is the radiation intensity, α_{k_0} is the multiphoton process cross section and $k_0 = \langle I/\hbar\omega + 1 \rangle$ the number of quanta required for ionization of an atom with an ionization potential I. The experimental values of the cross sections for these processes are in satisfactory agreement with perturbation-theory calculations.

1. INTRODUCTION

The character of multiphoton ionization of an atom is determined by the ratio of the energy of the total number of photons to the energy of the bound states of electrons in the atom. We are concerned with states perturbed by a strong light field.

When the difference between the energy of the perturbed bound states and the energy of the n photons is large, the ionization process occurs without excitation of real intermediate levels and proceeds via a single-stage transition of the electron to the continuous spectrum. This is called direct ionization.

The ionization probability W in the case of this direct process is related to the radiation intensity F by the formula

$$W = \alpha_{k_0} F^{k_0}, \tag{1}$$

where $k_0 = \langle I/\hbar\omega + 1 \rangle$ is the number of photons necessary for the ionization of the atom, I is the ionization potential, and α_{k_0} is the cross section for the multiphoton process. This cross section depends on the spectrum of bound states of the atom, and the frequency and polarization of the radiation. We shall calculate the cross section on the basis of k_0-order perturbation theory.[1-4]

Alkali metal atoms are convenient objects for measurements of the direct multiphoton ionization cross section. Analysis of the spectra of these atoms shows that, in most cases, if we use neodymium laser radiation ($\hbar\omega$ = 1.18 eV) and its second harmonic ($\hbar\omega$ = 2.36 eV), the difference between the energy of the n photons ($n < k_0$) and the energy of the bound states is large. Relatively low values of k_0 ensure the possibility of observing the ionization process for relatively low electric fields ($\varepsilon \sim 10^6$ V/cm). Perturbation theory calculations[5] and measurements[6,7] show that in a field of this order there is no change in the above energy difference. The first experiments on four-photon ionization of potassium and five-photon ionization of sodium by neodymium laser radiation showed that Eq. (1) was, in fact, satisfied.[8,9]

These experiments have thus confirmed qualitatively the assumed direct nature of the ionization process. A quantitative description of this process requires data on the size of the cross section.

We have measured the cross sections for two-, three-, four-, and five-photon direct ionization processes on alkali metal atoms. The two- and three-photon processes were observed for the ionization of potassium and sodium atoms, using the second harmonic of the neodymium laser, while the four- and five-photon processes were observed for the same atoms using the fundamental frequency. Preliminary data were reported previously in[10,11]. The two- and four-photon ionization cross sections of potassium were also reported in[12]. The three-photon ionization cross section of cesium at the frequency of the ruby laser radiation was measured in[13,14]. The cross sections for the above processes have been calculated on the basis of perturbation theory[2-4], which enables us to make a comparison between theory and the experimental data.

2. DESIGN OF THE EXPERIMENT

We have measured the cross sections by a direct method which involves the detection of the resulting ions. This method was used in the early work on multiphoton ionization of atoms,[15] The basic difference between this method as applied to multiphoton processes and photoionization is connected with the nonlinear nature of the ionization process. In addition to the usual measurements of the number of photons incident on the target, the density of atoms in the target, and the number of resulting ions, it is also necessary to determine the intensity of the radiation, i.e., the number of photons per unit area of the target per unit time.

In accordance with Eq. (1), the number of resulting ions is

$$N_i = n_0 \alpha_{k_0} \iint F^{k_0}(x, y, z, t)\, dv\, dt, \tag{2}$$

where n_0 is the density of neutral atoms and the integrals are evaluated over the volume within which the incident radiation interacts with the target and over the length of the laser pulse.

In practice, the intensity F is most conveniently measured by determining independently the space and time distributions of the radiation. This separation of variables is possible when the time dependence of the intensity is the same at all points within the target. The necessary condition for this is that the laser must radiate a single transverse mode and this was achieved in all our experiments.

Let us write the radiation intensity in the form

$$F(x, y, z, t) = F_{max} f(x, y, z, t) = F_{max} \varphi(x, y, z) \psi(t). \tag{3}$$

Accordingly, the energy per laser pulse is

$$Q = N_i \hbar\omega = \hbar\omega \iint F(x, y, z, t)\, ds\, dt$$

$$= F_{max} \hbar\omega \int \eta(x, y)\, ds \int \psi(t)\, dt = F_{max} \hbar\omega S\tau. \quad (4)$$

We find from Eqs. (2)—(4) that the cross section is given by

$$\alpha_{k_0} = \frac{N_i S^{k_0} \tau^{k_0} (\hbar\omega)^{k_0}}{n_0 Q^{k_0} V_{k_0} \tau_{k_0}}, \qquad V_{k_0} = \int \varphi^{k_0}(x, y, z)\, dv. \quad (5)$$

It follows from Eq. (5) that, before we can determine the cross section, we must measure the density n_0 of the neutral atoms, the number N_i of the ions, the energy Q per laser pulse, the space distribution of the radiation $\varphi(x, y, z)$, the time distribution $\psi(t)$, and the exponent k_0 in Eq. (1).

The principle of the experiment was similar to that described in[15]. The target was an atomic beam produced by a multichannel collimator[6,16]. The neutral atom density in the beam, n_0, was measured with a piezoelectric probe to within about 5%. We found that, typically, $n_0 \sim 10^9$ cm^{-3}.

Ions produced in the region of intersection of the laser beam and the atomic beam were accelerated by a constant electric field of about 100 V/cm and were intercepted by the detector. The mass composition of the ion beam was monitored in accordance with the time of flight over the path to the detector which took the form of an electron multiplier. Absolute measurements of the number of generated ions were carried out with a Faraday cylinder. The number of recorded ions lay between 100 and 10 000. The limiting resultant error in the absolute number of ions was about 25%.

The energy per laser pulse was measured by a standard IKT-1M calorimeter to within 10%. The energy per pulse was typically a few hundredths of a joule.

We used a neodymium laser (λ = 9439 cm^{-1}) Q-switched by a rotating prism. A diaphragm was inserted into the cavity to produce the TEM_{00} mode. A Glan prism placed outside the cavity was used to isolate plane-polarized radiation. The line width at half-height was ~ 10 cm^{-1}. The second harmonic was produced with the aid of a KDP converter mounted outside the resonator. The second harmonic was isolated using filters made of the colored glass SZS-21.

The space-time distribution of the radiation was determined by independent measurements of the spatial distribution integrated over the length of the laser pulse and time distribution integrated over the laser beam.[15,17]

To determine the spatial distribution one beam from the beam splitter was directed onto the objective similar to the objective focusing the laser beam on the atomic beam. Both objectives were at the same distance from the laser and a photometric method was used to measure the illumination in the focal region of the auxiliary objective. The distribution of the radiation over a number of sections perpendicular to the objective axis was imaged on a photographic film by a microscope with a magnification of ~ 100. The intensity distribution took the form of a diffraction pattern in which the diffraction depth was a few millimeters. The microobjective had a field depth of the order of a few microns, which was less by a factor of about 100 than the distance over which there was a noticeable change in the distribution of the radiation. The photometry of the image on the photographic film gave the quantity

$$S = \int \eta(x, y)\, ds.$$

Integration was carried out up to intensities amounting to 10% of the maximum intensity. The typical size of the effective focal circle within which maximum illumination was produced was about 10^{-5} cm^2. The limiting resultant error δ in the determination of S was ±15%.

The spatial distribution of the intensity was determined from data on the distribution in the different cross sections of the region within which the radiation was focused. The effective ionization volume was found from the formula $V_0 = \int \varphi(x, y, z)dv$ and, typically, $V_0 = 10^{-6}$ cm^3. The limiting resultant frequency was $\delta V_0 \approx \pm 25\%$. Accordingly, the uncertainty in the quantities in which we were interested lay between

$$\delta_{V_2} = \begin{matrix} +50\% \\ -40\% \end{matrix} \text{ and } \delta_{V_5} = \begin{matrix} +154\% \\ 72\% \end{matrix}.$$

The time distribution of the intensity was determined by directing the other beam from the beam splitter onto the fast photodiode FÉK, whose output was fed into the 2-7 oscillograph (total time resolution of the entire system was ~ 1 nsec).

All possible measures were taken to prevent ultrashort fluctuations in the radiation distribution connected with mode locking. The laser was operated in the single transverse mode, and the cavity contained no nonlinear elements other than the active rod. It was shown in[18] that under these conditions spontaneous mode locking was not very likely. Therefore, we are justified in assuming that there was no synchronization of the phases of the individual modes in our experiment.

A typical value is

$$\tau = \int \psi(t)\, dt \approx 3 \cdot 10^{-8} \text{ sec}.$$

The limiting resulting error was $\delta_\tau = \pm 15\%$.

The error varied between $\delta_{\tau_2} = \pm 25\%$ and $\delta_{\tau_5} = \pm 40\%$ for the two- and five-photon processes.

We used Eq. (5) to calculate the limiting error in α_{k_0} which was the upper limit for the accuracy of the experiment. The complicated nature of the errors in all the quantities in Eq. (5) complicates estimates of the mean error of the experimental data since, in many cases, the errors are systematic while, in other cases, they are random.

3. EXPERIMENTAL RESULTS AND DISCUSSION

In the experiments designed to determine the function $W \approx f(F)$ we measured the amplitude of the ion signal as a function of the energy of the radiation transmitted by the region in which the two beams intersected. The experimental data were represented by a power law using a least-squares procedure. To determine the cross section α_{k_0}, we measured the beam density n_0, the spatial distribution of the radiation $\varphi(x, y, z)$, the time distribution $\psi(t)$ in the focal region, and the energy per pulse. In all cases, the measured exponents were found to be equal to the corresponding k_0 to within experimental error (a few percent). The cross sections determined in this way are given in the table.

When theory and experiment are compared it must be remembered that the radiation used in our experiments had the multimode character. The statistics of multimode radiation has not been adequately investigated experimentally. In our experiment we measured $\overline{\mathscr{E}^2}$ whilst the probability of multiphoton ionization was determined by $\mathscr{E}^{2k_0}$. The relation between these quantities

is $\overline{\varepsilon^{2k_0}} = \eta_{k_0}(\overline{\varepsilon^2})^{k_0}$, where η_{k_0} is the k_0 order correlation function.

Calculations for the case of nonlocked modes with a random distribution of phases, which was realized in our laser (see above), is known to give $\eta_{k_0} = k_0!$. For two-, three- and four-photon processes the quantity $k_0!$ is appreciably less than the experimental error, but for the five-photon process it is comparable with the error. The data given in the table are not corrected for the correlation function.

The four-photon ionization cross section of the potassium atom was measured previously using the saturation of the ion signal and the formula $W\tau_{k_0} \approx 1$.[10] The cross section α_4 was determined to within a substantial error because the accuracy of processing of the experimental data was low. However, this result is in good agreement with the value of α_4 determined directly in the present experiment to within experimental error.

The cross sections α_2 and α_4 of the potassium atom were also reported in[12], but these results are higher than our measurements. We are looking into the reasons for this discrepancy.

The table gives the values of the cross section calculated from perturbation theory. Different approximations were used in[2-4] to calculate the combined matrix element. In[2] direct summation of the "principal" terms was performed and the oscillator strengths were determined in a Coulomb-like approximation. In[3] an effective oscillator strength was employed for virtual transitions over bound states. In[4] the Green function for the optical electron in the atom based on the quantum defect approximation was used.

It is clear from the table that, at the radiation frequency in which we are interested, the various calculated results based on different approximations are not substantially different.

The main conclusion which we may draw from the comparison between existing experimental data and calculations is that, to within experimental error, and apart from the uncertainties introduced by the various approximations, the perturbation theory provides a good description of two-, three- and five-photon ionization of alkali-metal atoms. The same conclusion follows from[13,14], where measurements were reported of the three-photon ionization of cesium.

On the whole, the above results can be regarded as only the first step in the study of the direct process of multiphoton ionization of atoms. A complete description of the process will require experimental data for circularly polarized radiation (the first results in this field were published in[13,19]) and measurements of the cross sections at a number of frequencies in the intervals between resonances.

Experimental and theoretical cross sections α_{k_0} for the ionization of potassium and sodium by radiation with $\hbar\omega = 1.18$ eV and $\hbar\omega = 2.36$ eV. The dimensions of $[\alpha_{k_0}]$ are $\text{cm}^{2k_0} \cdot \text{sec}^{k_0-1}$

k_0	Atom	$\hbar\omega$, eV	α_{exp}	α_{theor}		
				[2]	[3]	[4]
2	K	2.36	$10^{-48.8\pm0.8}$	$10^{-48.6}$	$10^{-48.6}$	$10^{-48.8}$
3	Na	2.36	$10^{-79.6\pm1.1}$	$10^{-79.2}$	$10^{-77.5}$	10^{-79}
4	K	1.18	$10^{-107\pm1.4}$	—	$10^{-106.5}$	10^{-106}
5	Na	1.18	$10^{-138\pm1.7}$	—	$10^{-140.3}$	10^{-139}

We note in conclusion that in each specific case, i.e., for each atom, frequency, and polarization, there is an upper limit for the field strength up to which the multiphoton ionization process is described by the cross section measured in such experiments. This restriction is connected with the appearance of resonances due to the perturbation of bound states by the radiation field and, consequently, the violation of Eq. (1).

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50