ANGULAR DISTRIBUTION of PHOTOELECTRONS in MULTIPHOTON IONIZATION

T. HELLMUTH
G. LEUCHS
S. J. SMITH
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T. Hellmuth*, G. Leuchs*, S.J. Smith†, H. Walther**

**Sektion Physik, Universität München and Projekgruppe für Laserforschung der Max-Planck-Gesellschaft zur Förderung der Wissenschaften e.V.

1. Introduction

The alkali atoms are especially attractive for the investigation of multiphoton processes. The ionization potentials are small so that photoionization can be performed by only a few photons in the visible spectral range. However, dye lasers can be used for the experiments and, in particular, make it possible to investigate, among other things, the wavelength dependence of the photoionization cross sections. On the other hand the simplicity of the alkali energy level scheme is an advantage for the theoretical understanding and interpretation of the results.

Beside the total cross section for photoionization and its wavelength and polarization dependence, the angular distribution of the photoelectrons is another experimental result giving important information on the multiphoton ionization process.

The angular distribution in multiphoton ionization is determined by the initial and intermediate states involved in the process, the transition amplitudes to the different partial waves of the free electron, the phase difference of the partial waves, the nature of the radiation field and in addition by the final state of the remaining core.

†Permanent address: JILA, National Bureau of Standards, Boulder, Col. 80302, USA
The angular distribution of photoelectrons has e.g. been investigated in connection with laser-photodetachment /1/. The first experiments with two photon ionization have been carried out by Edelstein et al. /7/ and later by Duncanson et al. /2/.

In the case of single photon ionization of atoms or ions with an equal population of the magnetic sublevels of the initial state, the photoelectron angular distribution can be described by the general formula:

\[
\frac{d\sigma}{d\Omega} = \frac{6\sigma_{tot}}{4\pi} \left[ 1 + \beta_2 \cdot P_2(\cos \theta) \right]
\]  

(1)

$\sigma_{tot}$ represents the total cross section, $\theta$ measures the angle between the direction of the ejected electron and the polarization of the incident light. $\beta_2$ is the asymmetry parameter and $P_2$ the second Legendre polynomial. $\beta_2$ ranges from $\beta_2 = 2$ to $\beta_2 = -1$. For a one electron atom $\beta_2$ has been derived by Bethe /3/.

The formula, however, can also be applied to many electron atoms provided the magnetic sublevels of the initial state are equally populated and the wavefunctions are represented by antisymmetrized products of spin orbitals.

The angular distribution (1) is also valid in the presence of configuration interaction and intermediate coupling. In the case of molecules the same form is obtained when the rotational orientations are averaged /4,5/.

In the case of a multiphoton ionization the angular distribution is in addition determined by Legendre polynomials with an order higher than two /8/. For an n-photon ionization $P_{2n}$ is the polynomial with the highest order which may contribute. In general the differential cross section following an n-photon ionization process can be expressed by:
\[
\frac{d\sigma}{d\Omega} = \text{const} \sum_{\nu=0}^{n} a_{\nu} \cos^{2\nu} \theta
\]  

(2)

where the \( a_{\nu} \) depend on the properties of the initial and intermediate states, the radiation field, the partial waves of the emitted electrons etc. The \( a_{\nu} \) contain the physics of the multiphoton ionization process.

The number of photoelectrons obtainable in a multiphoton-experiment are usually rather small and in general the measurement of the angular distribution is a rather difficult task. However, a considerable enhancement of the number of photoelectrons can be achieved if the laser frequencies used for the ionization process coincide with atomic transitions. In this case, the investigation of the angular distribution is not too difficult.

All the experiments on the angular distribution in two photon ionization have been performed for the resonant case. The first one on Ti atoms, which were excited by light from a nitrogen laser /7/ and the second one on Na atoms which were ionized stepwise by the light of a dye and a nitrogen laser /2/. In the latter experiment the dye laser was either tuned to the \( 3^2P_{1/2} \) or the \( 3^2P_{3/2} \) levels of Na.

In the following, experiments on the resonant three-photon ionization of sodium atoms will be described. In addition the quantum interference effects in the angular distribution in resonant two photon ionization being introduced by the hyperfine splitting of the intermediate state are studied in detail. The quantum interference effects have also been studied in the total photoelectron current.

**Three-Photon Ionization**

The experimental setup is shown in Fig. 1. The atoms of a sodium beam were irradiated by two laser beams originating from two different dye lasers pumped by the same nitrogen laser. The two dye laser beams were linearly polarized in the
Fig. 1 Experimental setup used for the measurement of the angular distribution in three-photon ionization

S P D F

Fig. 2 Energy level diagram showing the transitions used for the three-photon ionization
same direction. The direction of polarization could be rotated by means of a $\lambda/2$ plate. In this way the angle $\theta$ between the direction of emission of the electrons and polarization direction could be changed. The interaction region between the laser and atomic beams is electrically shielded. The electrons were detected with an angular resolution of 0.35 rad. Behind the aperture, defining the opening angle of the detection system, the electrons were accelerated and detected by means of an electron multiplier.

The excitation scheme used for the three photon ionization is shown in Fig. 2. The first laser beam ($\gamma'_1$), was tuned either to the $3^2S_{1/2} - 3^2P_{1/2}$ or $3^2S_{1/2} - 3^2P_{3/2}$ transition of the sodium atom (wavelengths 589.5 nm and 588.9 nm respectively). The second laser ($\gamma'_2$) performed a resonant excitation of the $n^0D$ fine-structure states.

The duration of the dye laser pulses was 4 ns and the output power between 10 and 50 kW. The spectral width of the laser was about 0.05 $\AA$. In this way the $2^2P_{1/2}$ and $2^2P_{3/2}$ levels could be excited separately whereas the $n^0D$ states were superposed since the fine-structure splitting is rather small. The photoionization was performed either by laser $\gamma'_1$ or $\gamma'_2$ or by both.

From theory one should expect that the angular distribution in the case of the photolionization via the $2^2P_{1/2}$ state is given by the following expansion

$$\frac{d\sigma}{d\Omega} \sim a_0 + a_1 \cos^2 \theta + a_2 \cos^4 \theta$$  (3)

This formula corresponds in principle to the result for two-photon ionization. This is due to the fact that the substates of the $2^2P_{1/2}$ state are isotopically populated by the excitation with the linearly polarized light. The accurate theory /9/, however, gives a contribution for $a_3$ which is nonzero (s. Table 1). This contribution is due to the perturbing influence of the neighbouring $2^2P_{3/2}$ level. The three-photon ionization via the $3^2P_{3/2}$ state contains a contribution up to the sixth power of $\cos \theta$. 

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Fig. 3 Angular Distribution of photoelectrons in resonant three-photon ionization of sodium via the $^2P_{1/2}$ and $^6D$ states (a), via the $^2P_{3/2}$ and $^6D$ states, and via the $^2P_{3/2}$ and $^10D$ states (c).

The experimental results are shown in Fig. 3 in a polar diagram. The solid line was obtained in a least squares fit of the analytical function (2) to the experimental points. The corresponding coefficients are compiled in Table 1 and compared to the theoretical values which have been calculated by Lambropoulos /9/. The experimental errors of the coefficients are about ± 0.05. There is a very good agreement between theory and experiment.

Table 1 Coefficients for the angular distribution in three-photon ionization (the coefficient $a_2$ is normalized to 1)

<table>
<thead>
<tr>
<th></th>
<th>$^2S_{1/2} - ^2P_{3/2} - n^2D - l1,k$</th>
<th>$^2S_{1/2} - ^2P_{1/2} - n^2D - l1,k$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>exp. n=6</td>
<td>exp. n=10</td>
</tr>
<tr>
<td>a</td>
<td>0.03</td>
<td>0.03</td>
</tr>
<tr>
<td>$a_1$</td>
<td>0.32</td>
<td>0.30</td>
</tr>
<tr>
<td>$a_2$</td>
<td>-1.0</td>
<td>-1.0</td>
</tr>
<tr>
<td>$a_3$</td>
<td>0.87</td>
<td>0.88</td>
</tr>
</tbody>
</table>
From Table 1 it can be seen that there is no change in the coefficients when the photoionization is performed either via the n=6 or n=10 state, which is also expected from theory. However, there is significant change when the stepwise ionization is performed either via the $^2P_{1/2}$ or the $^2P_{3/2}$ state. For comparison with theory it would be very interesting to observe the angular distribution when the laser is tuned between both states. Such experiments are presently underway.

Quantum Interference Effects in Two Photon Ionization

The possibility of observing quantum interference effects in photoionization has been proposed in theoretical publications /10,11/. The first experimental results have been published recently /12/.

In the experiment the two laser pulses used for the photoionization are delayed with respect to each other. The first pulse produces a coherent superposition of closely faced intermediate levels (e.g. hyperfine levels). This is possible if the duration of the pulse is short compared to $\pi/\Delta E$ where $\Delta E$ is the energy splitting of the intermediate state /12/.

The quantum interference effect can be observed both in the total rate of photoelectron emission and in the photoelectron angular distribution if the delay between the two laser pulses is varied. Experiments of both types which have been performed on the sodium atom will be described in the following. As discussed above the angular distribution of photoelectrons for two-photon-ionization is given by

$$\frac{d\sigma}{d\Omega} \sim a_0 + a_1 \cos^2 \Theta + a_2 \cos^4 \Theta \quad (4)$$

For the case of sodium the angular distribution in two-photon ionization via the $^3P_{3/2}$ state has been investigated previously by Strand et al. /2/. In this experiment the ionizing pulse was delayed up to 8 ns in order to measure the sensitivity of the angular distribution on the mixing process resulting from the hyperfine coupling in the $^3P_{3/2}$ level.
In the experiment described here measurements have been performed with delays between 0 and 32 ns, to demonstrate the periodicity i.e. the quantum interference signal due to the hyperfine levels of the $^{3}_2P_{3/2}$ state and to obtain from it, by Fourier analysis, a spectrum of the hyperfine structure. Furthermore the polarization dependence of the signal has been investigated and the quantum interference effect in the total electron current has been demonstrated.

The level scheme for the two-photon experiment is shown in Fig. 4. In an atomic beam experiment a nitrogen laser pumped dye laser, linearly polarized, was used to saturate the transition to the $^{3}_2P_{3/2}$ state. In a second step part of the 337 nm nitrogen laser radiation, also linearly polarized, was used to ionize the atoms producing photoelectrons with a kinetic energy of 0.6 eV. The nitrogen laser pulse duration was 6 ns and the power density at the atomic beam about $10^7 W/cm^2$.

![Level scheme of the two-photon experiment](image)

Fig. 4 Level scheme of the two-photon experiment
Fig. 5 Pulse duration of the laser pulses

The duration of the dye laser was about 4 ns (Fig. 5). The detection of the photoelectrons was performed in a similar way as described in the three-photon experiment (see above).

The electrons correlated with the nitrogen laser pulse are measured with a gated integrator capable of resolving single electrons. For an atomic beam density of $10^8$/cm$^3$ and a collecting solid angle of 0.25 sterad, one to ten photoelectrons were detected per laser pulse for delays not larger than the lifetime of the 3p state. The output of the integrator was averaged for one to three minutes for each angle setting, with the laser pulse repetition rate being 40 pps. The angular distribution was obtained by measuring the signal every 20 degrees over a range of 180 degrees and using the point symmetry of the angular distribution.

By lengthening its optical path, the nitrogen laser pulse was delayed with respect to the dye laser pulse. This was achieved by using up to five plane mirrors to form a simple optical delay line before the Glan-Thompson polarizer and the half wave plate.
observed a periodic variation in the total photoelectron current as a function of optical delay. However, these results were erratic since the variation of the time delay always caused a change in the transmission of the optical delay line and required a new adjustment of the laser beams for optimum overlap with the atomic beam. Therefore, the more tedious technique of measuring the angular distribution of the photoelectrons at each optical delay was used instead. This method is sensitive neither to the changes in transmission of the optical delay line nor to changes in overlap at the atomic beam which result from necessary optical adjustments.

Fig. 6 Quantum interference effect in the shape of the angular distribution of photoelectrons resulting from photoionization of the $3^2P_{3/2}$ state of sodium.

The upper part of Fig. 6 shows polar diagrams of the angular distributions of photoelectrons measured for various delays and normalized to a constant angle-integrated number of photoelectrons. The linear polarization of the two lasers was parallel. The solid line was obtained by a least squares fit of formula (4) to the experimental data, yielding the asymmetry parameters $\beta_2$ and $\beta_4$. In the lower part of Fig. 6, $\beta_4$ is plotted as a function of the delay of the nitrogen laser pulse with respect to the dye laser pulse. The delay time was varied between 0 and 38 ns. The statistical errors of the experimental points in the upper part of Fig. 6 are at most a few percent.
The observed periodic variation is the quantum beat signal of the four hyperfine levels of the $3^2p_{3/2}$ state. Fig. 7 shows the hyperfine structure of the ground state and of the $3^2p_{3/2}$ state of sodium. The arrows indicate transitions allowed due to the angular momentum selection rule $\Delta F=0,\pm 1$. Consequently the frequencies that are expected to appear in the quantum beat signal are $\Delta \nu_{32}$, $\Delta \nu_{21}$, $\Delta \nu_{10}$, $\Delta \nu_{32}^*+\Delta \nu_{21}^*$ and $\Delta \nu_{21}^*+\Delta \nu_{10}^*$. The frequencies of the periodically varying signal in Fig. 6 were determined by a Fourier analysis. Two frequencies $\Delta \nu_{32}=59.8 \text{ MHz}$ and $\Delta \nu_{32}^*+\Delta \nu_{21}^*=95.3 \text{ MHz}$ show up separately in the Fourier spectrum (Fig. 8). The frequencies $\Delta \nu_{21}$ and $\Delta \nu_{10}$ are not resolved, a peak at 24 MHz is observed instead. The separations of the hyperfine levels obtained from this preliminary evaluation are in reasonable agreement with previous results on the hyperfine splitting e.g. by Figger and Walther /13/.

![Energy level diagram](image)

Fig. 7 Part of the level scheme of sodium showing the hyperfine splitting of the $3^2p_{3/2}$ and of the ground state. The values for the hyperfine splitting are taken from Ref. /13/. The arrows indicate transitions between the two states.
Fig. 8 Fourier spectrum of $\beta(t)$

With the linear polarization of the two lasers oriented perpendicular to each other the angular distribution of the photoelectrons in two-photon ionization is changed since the influence of the magnetic substates differs in both cases. This can be seen in Fig. 9. Therefore, the phase of the quantum interference effect induced by the hyperfine splitting of the $3^{2}P_{3/2}$ state must also be different if the polarization of the two lasers is perpendicular to each other. A corresponding result is shown in Fig. 10.

The total photoelectron current also depends on the polarization of the two laser beams, therefore the quantum interference effect can also be observed when the following function $(i_{II} - i_{I})/(i_{II} + i_{I})$ is evaluated for the photoelectron current $i$ measured with a parallel $i_{II}$ and a perpendicular $i_{I}$ orientation of the polarization directions of the two laser beams. The result of a measurement is plotted in Fig. 11. The signal has the same period as the others shown in Figs. 6 and 10. Plotting the function $(i_{II} - i_{I})/(i_{II} + i_{I})$ the intensity change is eliminated, being introduced by the optical delay line when the time delay is varied.
Fig. 11 Quantum interference effect in the total photoelectron current. $i_\parallel$ and $i_\perp$ corresponds to the photoelectron current obtained with the linear polarization of the two laser beams either parallel or perpendicular to each other.

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Fig. 9 Two-photon ionization with the polarization of the two laser pulses oriented parallel and perpendicular to each other. The time delay between the pulses is zero.

Fig. 10 Quantum interference effect with linear polarization of the lasers perpendicular to each other.
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