

Electron polarization in multiphoton ionization of atoms

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The role of level mixing in the polarization of photoelectrons during multiphoton ionization of atoms via resonance on the fine structure p -level doublet is investigated. The effect is taken into account in the perturbation theory formalism for close quasi-energy levels. Ionization of the ground states is investigated in detail for atoms whose level system can be described by the L - S coupling scheme. Concrete calculations are carried out for alkali metal atoms by the Simons model-potential technique. It is shown that allowance for mixing can eliminate the discrepancy between the theoretical and experimental results. The most favorable conditions for experimental realization of the photoelectron-polarization mechanism considered in the paper are indicated on the basis of the results.

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The problem of obtaining polarized-particle beams in photoionization of unpolarized atoms has lately attracted much attention of both the theoreticians and experimenters.¹⁻⁶ Various polarization mechanisms are being considered. Historically, the first mechanism investigated was based on the Fano effect.² A detailed theoretical and experimental investigation has revealed, however, that this mechanism has significant shortcomings, namely a small cross section of the process in the vicinity of the Cooper minimum, the fact that it cannot be realized for all atoms, and the rigid requirement of a definite external-radiation frequency.

Andryushin and Fedorov² proposed for this mechanism a generalization free of these shortcomings. In this case the electrons are polarized on account of the interference between the channels of the one-photon ionization from the ground state and a certain excited state of the atom. An experimental realization of this process is made difficult, however, by the lack of a laser having a quantum energy sufficient for one-photon ionization of the ground state of the atom.

One more mechanism for obtaining polarized-electron beams, based on a two-step process, was proposed recently.⁴ During the first stage a photon of frequency ω_1 excites an isolated atomic p -state, which is ionized during the second stage by a photon of specially chosen frequency ω_2 . The condition imposed on ω_2 is that, owing to violation of the Bethe rule, the dominant branch of the ionization should be the branch in which the electron angular momentum decreases. An attractive aspect of this mechanism would be the possibility of obtaining beams of completely polarized electrons at an arbitrary polarization of the external radiation. The feasibility of this mechanism, however, calls for further study. The reason is that in ionization of the lower p -states of heavy alkali-metal atoms the Bethe rule is violated at photon energies exceeding the ionization potential of the ground states. This leaves open the one-photon ionization channel, in which the photoelectrons are not polarized. The possibility of separating a polarized beam against this background is connected with the need for energy selection of the emitted electrons.

As a result of these difficulties, the polarization mechanisms listed above were not realized in practice. More realistic from the point of view of experiment is polarization of photoelectrons produced in multiphoton ionization of atoms by circularly polarized radiation in the presence of an intermediate resonance with the doublet of the fine³ or hyperfine⁵ structure of the atomic levels. Total polarization of the photoelectrons is then possible, at least in principle, only in the first of these cases.

An experimental study of the polarization of electrons in ionization of a sodium atom via an intermediate resonance with a fine-structure doublet of $3p$ -levels is the subject of Ref. 7. An interpretation of the results of these experiments on the basis of nonstationary perturbation theory³ cannot explain the small degree of polarization of the photoelectron beam. The reason is that the formalism employed does not take into account the strong perturbation of the resonance levels. Field broadening and resonant mixing of the atomic levels were taken into account by Fedorov.⁶ In an approximation in which only resonant levels are taken into account, the problem of calculating the degree of polarization of a photoelectron beam reduces to solution of a secular equation. Within the limits of a weak (intensity $\mathcal{E} \ll 5 \times 10^5$ V/cm) and strong ($\mathcal{E} \gg 5 \times 10^5$ V/cm) fields, analytic expressions can be obtained for the degree of polarization. The results of Ref. 6 cannot be used to interpret actual experiments on electron polarization in multiphoton ionization of alkali-metal atoms. This is due, on the one hand, to the fact that these experiments are carried out at intermediate fields $\mathcal{E} \sim 5 \times 10^5$ V/cm, and on the other because the smoothing of the resonance, due to the field broadening of the level, may make the contribution of the nonresonant terms to the ionization cross section substantial. Actual calculations show that it is precisely this situation which obtains in experimental fields in ionization of light alkali metal atoms. In this case no analytic expressions can be obtained for the degree of polarization of the photoelectron beam.

The reason for this is that the matrix element of the transition $\langle \bar{n}s | r | n_1 p \rangle$ decreases rapidly with increasing difference $n_1 - n$. Estimates using the Bates-Damgaard tables

for the rubidium atom, for example, give a ratio $\langle 5s|r|5p \rangle / \langle 5s|r|6p \rangle = 15$, and even $\langle 5s|r|5p \rangle / \langle 5s|r|8p \rangle = 96$. Since typical distances between the levels in the spectra of alkali-metal atoms are of the order of 10^4 cm^{-1} , and the fine splitting for heavy atoms is $\sim 10^1 - 10^2 \text{ cm}^{-1}$, the contribution of the resonant and nonresonant terms on going through resonance with the $8p$ state are comparable even in the absence of smoothing of the resonance. In this case there is no need to take into account the field broadening of the latter, since it is much smaller than the corresponding energy differences. In transitions through resonance with $n_1 > 8$ levels the situation is even more favorable, since the matrix element decreases approximately in the same manner as in the hydrogen atom, namely $\propto n_1^{-3/2}$, and the fine splitting decreases much more slowly. A similar situation is observed also for the cesium atom.

The contribution of the nonresonant terms can be taken into account in natural fashion in the perturbation-theory formalism developed in Ref. 8 for close quasi-energy levels. The quasi-energy function of a valence electron can be represented in the form

$$|\Phi\rangle = \sum_{i=1}^N a_i |\Phi_i^0\rangle + G_E^p \hat{V} |\Phi\rangle. \quad (1)$$

Here G_E^p is the reduced Green's function, which contains nonresonant terms, $\{\Phi_i^0\}$, $i = 1, 2, \dots, N$; \hat{V} is the operator of dipole interaction with the field. The coefficients a_i are solutions of a certain system of algebraic equations, and the quasi-energies E are the roots of the corresponding secular equation.

We consider the ionization, by a circularly polarized field, of the S state of an atom whose level system is described by L - S coupling via one-photon resonance with a doublet of a p -level fine-structure.

In the dipole approximation, the operator of the interaction with the field is

$$\hat{V} = e(\mathbf{r}, \vec{\mathcal{E}}), \quad \vec{\mathcal{E}} = \frac{\mathcal{E}_0}{\sqrt{2}} \{\cos \omega t, \sin \omega t, 0\}. \quad (2)$$

In the case of ionization from the state $|n_0 s \frac{1}{2} m_j\rangle$, depending on the sign of the projection of the total angular momentum on the quantization axis, m_j , a system of two quasi-energy levels

$$|\Phi_{m_j = \frac{1}{2}}^1\rangle = a_0^{(\frac{1}{2})} \left| n_0 s \frac{1}{2} \frac{1}{2} \right\rangle + a_3^{(\frac{1}{2})} \left| n_1 p \frac{3}{2} \frac{3}{2} \right\rangle \quad (3)$$

or of three levels

$$|\Phi_{m_j = -\frac{1}{2}}^1\rangle = a_0^{(-\frac{1}{2})} \left| n_0 s \frac{1}{2} -\frac{1}{2} \right\rangle + a_1^{(-\frac{1}{2})} \left| n_1 p \frac{1}{2} \frac{1}{2} \right\rangle + a_3^{(-\frac{1}{2})} \left| n_1 p \frac{3}{2} \frac{1}{2} \right\rangle \quad (3')$$

is produced. The coefficients $a_i^{(m_j)}$ are determined from the following system of equations:

$$a_i (E_k - E_0) = \sum_{j=0}^N \langle \Phi_i^0 | \hat{V} | \Phi_j^0 \rangle a_j; \quad i, j = 0, 1, 2, \dots, N, \quad (4)$$

whose solutions in our case take the form

$$\begin{aligned} a_0^{(m_j)} &= (E_k - E_{\frac{1}{2}} + \omega) (E_k - E_{\frac{3}{2}} + \omega) \\ &\times \{ (E_k - E_{\frac{1}{2}} + \omega)^2 (E_k - E_{\frac{3}{2}} + \omega)^2 \\ &+ V_1^2 (E_k - E_{\frac{1}{2}} + \omega)^2 + V_3^2 (E_k - E_{\frac{3}{2}} + \omega)^2 \}^{-1/2}, \\ a_{1,3}^{(m_j)} &= V_{1,3} \text{sign} (E_k - E_{1,3} + \omega) \\ &\times \{ V_{1,3}^2 + (E_k - E_{3,1} + \omega)^2 [1 + V_{1,3}^2 / (E_k - E_{1,3} + \omega)^2] \}^{-1/2}. \end{aligned} \quad (5)$$

The matrix elements $V_{1,3}^{(m_j)}$ are defined here as follows:

$$V_{1,3}^{(m_j)} = \left\langle n_0 s \frac{1}{2} m_{j_1} \left| \hat{V} \right| n_1 p \frac{1}{2} \left(\frac{3}{2} \right) m_{j_2} \right\rangle$$

and, in accordance with the dipole selection rules, $V_{1,3}^{(3/2)} = 0$. The quasienergies are solutions of the secular equation corresponding to (4), which ensure the needed asymptotic behavior of the wave function (3) and (3') when the external field is turned off.

The probability of a transition from the state $|\Phi_{m_j}^1\rangle$ into a continuum state $|f\rangle$ can be calculated by perturbation theory. In this case, since the initial state of the atom is not polarized, the total probability of its ionization is obtained by averaging over the projections of the total angular momentum on the quantization axis, whose direction is determined in this case by the direction of the momentum of the photon of the circularly polarized field. In the case of two-photon ionization, the probability can be expressed in terms of radial composite matrix elements of perturbation theory, in the form

$$\begin{aligned} R_{1,3}^{(m_j)} &= \left\langle n_0 s \frac{1}{2} m_j \left| r g_{\frac{1}{2}, (\frac{1}{2})}^p r \right| f \right\rangle \\ &+ \frac{1}{2} \sum_{m_j} a_{1,3}^{(m_j)} \left\langle n_1 p \frac{1}{2} \left(\frac{3}{2} \right) m_j \left| r \right| f \right\rangle. \end{aligned} \quad (6)$$

The probability of three-photon ionization via a one-photon resonance is expressed in terms of the matrix elements

$$\begin{aligned} R_{1,3}^{(m_j)} &= \left\langle n_0 s \frac{1}{2} m_j \left| r g_{\frac{1}{2}, (\frac{1}{2})}^p r g_{\frac{3}{2}, (\frac{3}{2})}^d r \right| f \right\rangle \\ &+ \frac{1}{2} \sum_{m_j} a_{1,3}^{(m_j)} \left\langle n_1 p \frac{1}{2} \left(\frac{3}{2} \right) m_j \left| r g_{\frac{1}{2}, (\frac{1}{2})}^d r \right| f \right\rangle. \end{aligned} \quad (6')$$

Here g_j^d is a radial reduced Green's function.

The degree of polarization of a photoelectron beam, defined as

$$P = (N_+ - N_-) / (N_+ + N_-) \quad (7)$$

$[N_+(N_-)]$ is the number of photoelectrons with positive (negative) spin projection, can be expressed in the case of ionization via one-photon resonance in terms of the matrix-element ratio $R = R_{1/2} / R_{3/2}$ in the following manner: for two-photon ionization

$$P_{2\omega} = [9 + (1-R)^2 - (2+R)^2] / [9 + (1-R)^2 + (2+R)^2]; \quad (8a)$$

for three-photon ionization

$$P_{3\omega} = [27 + 2(1-R)^2 - 3(2+R)^2] / [27 + 2(1-R)^2 + 3(2+R)^2]. \quad (8b)$$

These expressions coincide in form with those obtained in Ref. 3, and go over into them when account is taken in (6) of only the resonant terms, and the mixing of the levels is neglected in the latter; this reduces to the substitution

$$a_{1,(3)} \rightarrow V_{1,(3)} / (E_n - E_{n_1} + \omega).$$

As seen from Eqs. (8), complete polarization of the photoelectrons takes place at $R = -2$. Neglecting the mixing of the resonant levels, the ratio R does not depend on the external-field intensity. In the fine-structure interval, depending on the radiation frequency, it varies in this case in the range $-\infty < R < 0$ and reaches the sought value $R = -2$ at $\omega = \frac{1}{3}(2\omega_{1/2} + \omega_{3/2})$.

The smoothing of the intermediate resonance, noted in Ref. 9, which is due to mixing of the levels, alters the situation radically. The ratio R in this case is limited in absolute value, and at a certain external field frequency the condition for complete polarization cannot be reached. As seen from (5), the possibility of reaching complete polarization of the beam is determined by the value of the parameter $\alpha = V\mathcal{E} / |E_{1/2} - E_{3/2}|$; at $\alpha \ll 1$ the mixing can be neglected and consequently, complete polarization is attainable. In the opposite limit $\alpha \gg 1$ complete polarization of a photoelectron beam is impossible.

The actual calculations were carried out for alkali-metal atoms. The radial composite matrix elements of the perturbation theory were calculated by using a Green's function constructed in the Simons model potential,¹⁰ and the secular cubic equation was solved numerically. As indicated by the results of these calculations, the \gg (\ll) symbol in the estimate of α can be replaced respectively by $>$ ($<$). This fact is illustrated in Fig. 1, which shows the maximum attainable degree of polarization of photoelectrons as a function of the external-field intensity. The abscissas are, for comparison, the values of the critical field intensities calculated from the condition $\alpha = 1$. It can be seen from the figure that in fields in which the experiments on multiphoton ionization of alkali-metal atoms are usually carried out ($\mathcal{E} \sim 10^6 - 10^7$ V/cm) total polarization of the beam is possible only for rubidium and cesium atoms, in the spectra of which the fine splitting is appreciable (it reaches hundreds of cm^{-1} for the lower p -states of these atoms). Thus, the estimate of the critical fields ($\mathcal{E} \ll 10^5$ V/cm) given in Ref. 6 is on the whole valid for light alkali-metal atoms, and is strongly undervalued for heavy atoms. Since the fine splitting ΔE decreases rapidly with increasing principal quantum number n_1 , the real source of polarized electrons can be ionization via excited levels with small n_1 . Thus, for example, even in the most favorable case of the cesium atom $n_1 - n_0 < 5$ in the fields used in experiment.

An experimental investigation of the polarization mechanism considered in the present paper is reported in Ref. 7. It should be noted that the ionization scheme realized in that experiment differs somewhat from the one considered above. In the experiment, the ground state of the sodium atom was ionized in two stages. During the first stage the doublet $3p_{1/2,3/2}$ was excited by weak-field photons, after which the excited state was ionized by strong-field photons

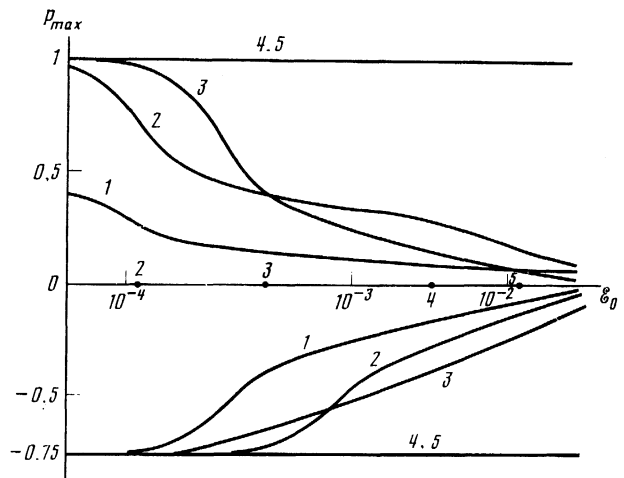


FIG. 1. Maximum attainable degree of polarization of photoelectrons in three-photon ionization of the ground states of alkali-metal atoms via one-photon resonance with the doublet of the fine structure of the lower p -level vs the external-field intensity. The abscissa axis shows also the values of the critical field calculated from the condition $\alpha = 1$. It should be noted that \mathcal{E}_{cr} for lithium is quite small ($\mathcal{E}_{cr} < 10^{-3}$ a.u.) and is not shown in the figure. The numbers 1, 2, 3, 4, and 5 refer to the curves and to the values of \mathcal{E}_{cr} for lithium, sodium, potassium, rubidium, and cesium atoms, respectively. As seen from Eq. (8b), for the mechanism considered in the paper the maximum attainable degree of polarization, corresponding to electron-spin orientation in the direction of the momentum, is equal to unity, and the degree of negative polarization (the electron spin is oriented predominantly opposite to the photon-momentum vector) cannot exceed 0.72. The figure shows that the degree of polarization of the photoelectrons deviates from the maximum attainable in principle in fields stronger than \mathcal{E}_{cr} . The critical fields for heavy alkali-metal atoms, rubidium, and cesium are comparable with the maximum attainable in experiment. This circumstance makes the rubidium and cesium atoms most convenient objects for a practical realization of the investigated photoelectron polarization mechanism.

via resonance with the bound d -state. These differences, however, do not influence the frequency dependence of the degree of polarization P . As can be seen from (5), the shape of the curve depends only on the intensity of the exciting field. In addition, resonant effects on the d -levels can be neglected, owing to the large laser radiation width, which exceeds the fine splitting of these levels. Thus, despite the indicated differences, the form of the frequency dependence of the degree of polarization agrees also under the conditions of this experiment with Eqs. (5) and (8b). A detailed comparison of the theoretical and experimental results is made difficult, however, by the fact that Ref. 7 does not contain information on the intensity of the exciting field. From the data available in that reference concerning the ratio of the intensity of the ionizing and exciting fields one could conclude that the latter has an intensity in the interval $10^{-4} < \mathcal{E} < 3 \times 10^{-4}$ a.u. Figure 2 shows a comparison of the theoretical and experimental results based on this estimate. As can be seen from the figure, the theoretical data agree well with the experimental results, which at the same time contradict strongly the results of the theory that does not take the level mixing into account.

Thus, summarizing the foregoing, we can state that when considering the mechanism of polarization of a photo-

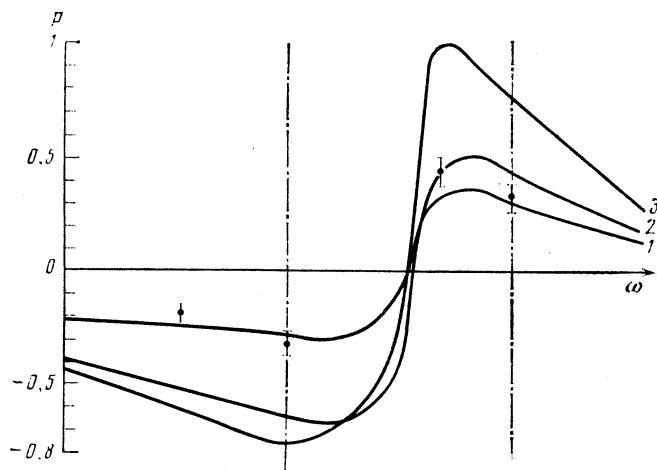


FIG. 2. Frequency dependence of the degree of polarization of photoelectrons in three-photon ionization of a sodium atom via one-photon resonance with the fine-structure doublet of the levels $3p_{1/2}$ and $3p_{3/2}$. The dash-dot lines show the positions of the resonances with unperturbed components of the doublet. Curves 1 and 2 correspond to the exciting-field intensities $\mathcal{E} = 10^{-4}$ and $\mathcal{E} = 3 \times 10^{-4}$ a.u. Curve 3 shows the results of a theoretical paper in which no account was taken of the mixing of the resonance levels.³ The frequency dependence of the degree of polarization of the photoelectrons does not depend in this case on the field intensity. The figure shows also the experimental results of Ref. 7. It is seen from the figure that the experimental results agree well with the present paper and at the same time contradict strongly the theory that does not take into account the resonant perturbation of the levels.

electron beam in multiphoton ionization via single-photon resonance of atoms whose spectra are described by the L - S

coupling scheme, account must be taken of the mixing of the resonance levels. Allowance for this effect lowers greatly the estimated efficiency of the considered polarization mechanism for light alkali-metal atoms. The most favorable case of observation of photoelectron polarization is the ionization of the ground states of rubidium and cesium atoms via one-photon resonance with low-lying p -levels.

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