

Polarization of electrons produced as a result of the resonance ionization of atoms by high-intensity electromagnetic radiation

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The polarization of the electrons produced in the course of the resonance ionization of atoms by an intense circularly-polarized field is investigated. The degree of polarization under conditions of resonance between the *S*- and *P*-states is found in the fine and hyperfine level structure schemes a) within the framework of perturbation theory, b) in the field-broadening regime, and c) under conditions when the ionization level broadening predominates. The polarization properties of photoelectrons in very strong fields are investigated with allowance for resonance or nonresonance mixing of the fine structure levels. The electromagnetic-field intensities that are optimal from the standpoint of the use of the resonance-ionization process as a source of polarized electrons are found.

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1. INTRODUCTION

Recently, the possibility of the practical use of the process of resonance ionization of atoms as a source of polarized electrons, i.e., electrons with their spins predominantly oriented along some direction, has been widely discussed in the literature.^[1–8] The basic idea underlying propositions of this sort is that under certain conditions excited resonance states can possess a certain degree of polarization even in the case of the action of radiation on nonpolarized atoms. For this purpose, the radiation must first and foremost have a sufficiently high degree of monochromaticity, which will allow the selective excitation of the fine-structure sublevels of the atomic spectrum. The polarization effects can arise only at the expense of the circularly-polarized part of the radiation (under ideal conditions the field should be circularly polarized 100%).

To ensure a high efficiency of the process, the external field should be sufficiently intense, so as to bring about almost complete ionization of the atoms during the time of action of the pulse. This requirement is fulfilled at, for example, a field intensity $\mathcal{E}_0 \sim 5 \times 10^5$ V/cm under conditions of complete saturation of the resonance transition for a pulse duration $\tau \sim 10^{-8}$ sec if the transition from the resonance state into the continuum is a single-photon transition. On the other hand, the use of radiation of so high an intensity may be dangerous to the appearance of polarized photoelectrons itself, since the reconstruction of the quasienergy spectrum of the atom in the external field can then play a significant role. In view of this, the investigation of the effect of the transition to “strong” fields on the resonance-ionization process and on the degree of polarization of the photoelectrons is important, and it is to this investigation that the present paper is devoted.

The problem of the resonance ionization of atoms in a strong electromagnetic field has lately been investigated in great detail (see, for example, Refs. 9 and 10 and the literature cited in Ref. 10). However, this analysis is inadequate for the study of the polarization

properties of photoelectrons in strong fields in view of the fact that, within the framework of the approaches usually used in the theory of resonance ionization, a) the electron spin is not considered at all and b) the single-resonance-level approximation is used, i.e., the real structure of the resonance levels is not taken into account. In the present paper we shall take these two factors into consideration. Let us limit ourselves to the consideration of the simplest, but quite realistic two-photon resonance ionization model corresponding to the transition scheme $S \rightarrow P \rightarrow D, S$. Let us assume (which is most justified from the practical standpoint) that the excitation of the resonance levels and the subsequent transition into the continuum are effected by two different fields with frequencies ω and $\bar{\omega}$ and intensity amplitudes \mathcal{E}_0 and $\bar{\mathcal{E}}_0$. Let us assume for simplicity that the two pulses have the same width, τ , and act on the atom simultaneously. Let us limit ourselves to the case when both waves propagate in the same direction (along the *z* axis) and both are clockwise-polarized. We shall assume that the natural level width is fairly small, so that not only the fine, but also the hyperfine (HF) structure of the atomic spectrum can be spectrally resolved.

2. PERTURBATION THEORY

The perturbation theory is valid in fairly weak \mathcal{E}_0 and $\bar{\mathcal{E}}_0$ fields. Depending on the relation between the spectral width, $\Delta\omega \gtrsim \tau^{-1}$, of the radiation and the characteristic scale of the HF splitting $\Delta E_{\text{HF}} \sim 10^{-3}$ cm⁻¹, we should (when $\Delta\omega \ll \Delta E_{\text{HF}}/\hbar$), or should not (when $\Delta\omega \gg \Delta E_{\text{HF}}/\hbar$), take into account the effect of the interaction of the electronic angular momentum *j* and the nuclear spin *I* on the spectrum and the wave functions of the atom. In the latter case the condition for resonance determines the value of the electronic-angular-momentum quantum number, *j'*, of the *P* multiplet, which is resonantly coupled to the ground state $S_{1/2}$. If, on the other hand, the spectral width of the radiation allows the resolution of the HF structure, then the condition for resonance also determines the values of the total-

angular-momentum ($F=j+I$) quantum numbers, F and F' , of the atom in the initial and resonance states.

We shall assume that the unexcited atoms are unpolarized, which is equivalent to the assumption that they are distributed with equal probability over the quantum numbers of the total-angular-momentum component, m_F , or of the electronic-angular-momentum component, m_j , along the z axis in the ground (S) state. Within the framework of perturbation theory, the probability for the production during resonance ionization of an electron propagating in a unit solid angle in the direction $\mathbf{n}=\mathbf{p}/|\mathbf{p}|$ and possessing a spin component along the z axis equal to $\mu=\pm 1/2$ can be found in the HF- and final-level-structure schemes from the formulas

$$\frac{dw_{\mu}^{\text{HF}}}{d\Omega} = \frac{2\pi(2m^2E)^{1/2}}{\hbar(2F+1)} \sum_{m_i, m_r} \frac{|\langle \psi_{p\mu} m_i | \bar{V} | P j' F' m_r + 1 \rangle|^2}{\Delta_{F'F}^2 + \Gamma^2/4} \times \left| \langle P j' F' m_r + 1 | V | S, j = \frac{1}{2}, F m_r \rangle \right|^2, \quad (1)$$

$$\frac{dw_{\mu}^{\text{F}}}{d\Omega} = \frac{\pi(2m^2E)^{1/2}}{\hbar} \sum_{m_i} \frac{|\langle \psi_{p\mu} | \bar{V} | P j' m_i + 1 \rangle|^2}{\Delta_{j'j}^2 + \Gamma^2/4} \left| \langle P j' m_i + 1 | V | S, j = \frac{1}{2}, m_i \rangle \right|^2 \quad (2)$$

where $\Delta_{F'F}$ and $\Delta_{j'j}$ are the resonance detuning with and without allowance for the corrections to the level energy due to the HF splitting; Γ is a phenomenologically introduced natural resonance-level width, which, by assumption, does not depend on m_F and m_j ; V and \bar{V} are the spatial parts of the operators of the interaction of the atom with the exciting and ionizing radiation,

$$V = 1/2 e \mathcal{E}_0 r e^{i\omega t} \sin \theta, \quad \bar{V} = 1/2 e \mathcal{E}_0 \bar{r} e^{i\omega t} \sin \theta,$$

$r, \theta,$ and φ are the spherical coordinates of the valence electron; $\psi_{p\mu}$ is the electron wave function in the continuum, possessing at large distances from the atom "a plane wave + an ingoing spherical wave" asymptotic form; \mathbf{p} and μ are the asymptotic values (for $r \rightarrow \infty$) of the momentum and the z component of the spin of the free electron; $E = p^2/2m$ is its kinetic energy, determined by the conservation law; and $|m_i\rangle$ is the final state's nuclear spin function corresponding to the z component, equal to m_i , of the nuclear spin. The limits of the summation over m_F and m_j in the formulas (1) and (2) are determined by the conditions

$$|m_F| \leq F, \quad |m_F + 1| \leq F'; \quad |m_j| = 1/2, \quad |m_j + 1| \leq j',$$

where $j' = 1/2$ or $3/2$.

The degree of polarization of the photoelectrons is defined as

$$P = \frac{w_{\mu} - w_{-\mu}}{w_{\mu} + w_{-\mu}}, \quad w_{\mu} = \int d\Omega \frac{dw_{\mu}}{d\Omega}. \quad (3)$$

The calculation of the degree of polarization from the formulas (1), (3) in the case of resonance at the $P_{1/2}$ level of the Cs atom, where the nuclear spin $I = 7/2$, yields

$$P = (1 + az^2)/(b + az^2), \quad (4)$$

TABLE I.

F'	F	a	b	$P(1/2)$	$P'(1/2)$
3	3	-1.25	-2.1	-0.29	0.25
	4	-0.63	-2.55	-0.31	0.16
4	3	-0.25	-1.75	-0.52	0.44
	4	-0.795	-1.93	-0.38	0.27

where a and b are constants that depend on F and F' ; $z = R_S(E)/R_D(E)$, $R_S(E)$ and $R_D(E)$ are the radial elements for the transitions from the resonance state $P_{1/2}$ into the $S_{1/2}$ and $D_{3/2}$ states, respectively, of the continuum.

Notice that the resonance ionization that occurs according to the scheme $S_{1/2} \rightarrow P_{1/2} \rightarrow S_{1/2}$ in a circularly polarized field is, on the basis of the selection rules in the fine level structure approximation, forbidden. Such a transition becomes, however, possible when the interaction of the electron with the nuclear spin is taken into consideration, i.e., in the scheme of the HF level structure. The parameter z in the case of resonance at the $7^2P_{1/2}$ level of the Cs atom is close to the value $1/2$.^[5] In Table I we present the results of the computations of the constants a and b , which determine the degree of polarization, (3), of the photoelectrons and its $P(1/2)$ values, computed with $z = 1/2$, in the case of resonance ionization of the cesium atom under conditions of resonances at the sublevels of the HF structure of the $P_{1/2}$ term; $P(3/2)$ is the degree of polarization at resonance at the sublevels of the $P_{3/2}$ term.^[4] As can be seen from this table, the degrees of polarization in these two cases have different signs, i.e., the preferred direction of the electron spin is in the case of resonance at the sublevels of the $P_{1/2}$ term the direction antiparallel, and in the case of resonance at the $P_{3/2}$ sublevels the direction parallel, to the wave vector of the light.

The calculation of the degree of polarization in the fine-structure scheme shows that in the case of resonance at the $P_{1/2}$ and $P_{3/2}$ levels the degree of polarization has the values -0.6 and $+0.82$, respectively.^[6] A comparison of the values of the degree of polarization found under the conditions of applicability of the fine- and HF-structure schemes shows that allowance for the HF splitting worsens the polarization properties of the photoelectrons. This result is due to the fact that, in essence, the possibility of producing polarized photoelectrons in the process of resonance ionization is connected with the spectral properties of the radiation and with the selection rules involving the quantum number associated with the z component of the electron angular momentum m_j . Allowance for the interaction with the nuclear spin destroys to a certain extent the strict selection rules that are valid in the fine level structure scheme, and this leads to a reduction (in the absolute

TABLE II.

F'	F	a	b	P_f	P_i
3	3	10	16.4	0.19	-0.25
	4	-2.48	-3.48	-0.09	-0.37
4	3	-1.13	-1.94	-0.33	-0.50
	4	-1.18	-1.98	-0.31	-0.30

value) of the degree of polarization of the photoelectrons. It follows, in particular, from this that, from the standpoint of the production of highly polarized electrons in the process of resonance ionization, the optimal pulse width, τ , should satisfy the conditions $\hbar/\Delta E_{\text{HF}} \sim 10^{-8} - 10^{-9} \text{ sec} \gg \tau \gg 10^{-12} \text{ sec}$ (for $\tau \lesssim 10^{-12} \text{ sec}$ the spectral width $\Delta\omega \gtrsim 1/\tau$ overlaps the fine structure of the levels, and this also sharply reduces the degree of polarization).

Let us find the limits of applicability of the perturbation theory and the changes in the polarization properties of the electrons to which the growth of the intensities \mathcal{E}_0 and \mathcal{E}_0 leads. As applied to resonance ionization, the action of the external field on the atom has underlying it four distinguishable principal mechanisms, which lead to a reconstruction of the quasienergy spectrum of the atom; to wit: 1) the field resonance broadening^[9-11], which is a manifestation of the well-known shift of the quasienergy levels of a two-level system in a resonance field^[12], 2) resonance level mixing, which arises in the case when the field broadening covers several sublevels of the resonance-level structure, 3) ionization broadening, which is due to the ionization of the resonance states^[9,10,13], and 4) nonresonance mixing^[14], which arises when the corrections to the energy due to the quadratic dynamic Stark effect exceed the characteristic scale of the level structure. The first two of these phenomena are connected with the growth of the intensity, \mathcal{E}_0 , of the exciting field, while the last two are connected with the growth of \mathcal{E}_0 . Let us consider these two groups of effects separately.

3. FIELD BROADENING AND RESONANCE MIXING OF THE LEVELS

Let us consider the spectrum perturbation connected with the resonance interaction with the field \mathcal{E}_0 , neglecting the resonance level shift in this field (this shift being as small as the parameter $\mathcal{E}_0/\mathcal{E}$ at $\ll 1$, where $\mathcal{E}_{\text{at}} = 5 \times 10^9 \text{ V/cm}$ is the intratomic field), as well as the effect of the ionizing field on the spectrum, which is justified when $\mathcal{E}_0 \gg \mathcal{E}_0^2/\mathcal{E}_{\text{at}}$. If the natural width of the levels is small compared to the spectral width of the radiation, then the condition for the applicability of the results of the perturbation theory clearly has the form $\mathcal{E}_0 \ll \mathcal{E}_{\text{at}} \hbar/\tau \text{ Ry}$, where it has been assumed that $\tau^{-1} \sim \Delta\omega$. In this case, if $\tau \ll \hbar/\Delta E_{\text{HF}}$, then the fine level structure scheme operates, while if $\tau \gg \hbar/\Delta E_{\text{HF}}$, then the results obtained in the HF-structure scheme are valid. In the latter case, in the field range

$$\mathcal{E}_{\text{at}} \frac{\hbar}{\tau \text{ Ry}} \ll \mathcal{E}_0 \ll \mathcal{E}_{\text{at}} \frac{\Delta E_{\text{HF}}}{\text{Ry}} \sim 10^2 \frac{\text{V}}{\text{cm}}$$

there is realized the saturation regime in the HF-structure scheme, or, in terms of the theory of resonance ionization, the ionization of the atom via a single level under conditions of field broadening of the HF-structure sublevels. The problem of the computation of the ionization probability can be solved in this case if we allow for the resonance interaction V within the framework of a two-level system exactly^[12] and allow for the ionizing field \mathcal{E}_0 within the framework of (first-order) per-

turbation theory. As can easily be verified, in complete analogy to Refs. 9 and 10, the linear (with respect to the time) ionization regime is possible in this case, the probability $d w_\mu/d\Omega$ being determined by an expression of the type of the perturbation-theory formula (1), with, however, the natural width Γ replaced by the so-called field width

$$\Gamma_f(m_F) = 4 |\langle P'_{1/2} F' m_F + 1 | V | S'_{1/2} F m_F \rangle|. \quad (5)$$

The dependence of the field width Γ_f on the quantum number m_F changes the degree of polarization P . In particular, in contrast to the case of a weak \mathcal{E}_0 field, the degree of polarization P depends on the magnitude of the detuning $\Delta_{F'F}$ (i.e., it can vary appreciably as the frequency ω is varied in an interval $\sim \Gamma_f/\hbar$). At exact resonance ($|\Delta_{F'F}| \ll \Gamma_f$) the expression for the probability $d w_\mu/d\Omega$ assumes the extremely simple form:

$$\frac{d w_\mu}{d\Omega} = \frac{\pi(2m^2 E)^{\text{th}}}{2\hbar(2F+1)} \sum_{m_F} |\langle \psi_{\mu, m_F} | V | P'_{1/2} F' m_F + 1 \rangle|^2. \quad (6)$$

The computation of the degree of polarization P with the aid of the formulas (3) and (6) for the case of resonance at the sublevels of the $P_{1/2}$ multiplet of the Cs atom ($I = 7/2$) allows us to again reduce the expression for P to the form (4), where the constants a and b , as well as the values of the degree of polarization P_f for $z = 1/2$ are given in Table II. A comparison with the results obtained within the framework of perturbation theory (Table I) shows that an increase in the field intensity \mathcal{E}_0 and the transition to the field-broadening (saturation) regime lead in all cases to a decrease (in the absolute value) of the degree of polarization of the photoelectrons. In the region of stronger fields, i.e., for $\mathcal{E}_0 \gg \mathcal{E}_{\text{at}} \Delta E_{\text{HF}}/\text{Ry} \sim 10^2 \text{ V/cm}$, the field broadening exceeds the scale of the HF structure of the levels, and this leads to resonance mixing of the sublevels with different F and m_F . A consequence of the resonance mixing in this case is the transition from the HF structure scheme to the fine-level-structure scheme with allowance for the field broadening of the levels.

We can convince ourselves of the validity of this assertion with the aid of the following argument. If the field broadening is small in comparison with the characteristic scale of the fine structure, i.e., if

$$\mathcal{E}_0 \ll \mathcal{E}_{\text{at}} |E_{F+1} - E_{F-1}|/\text{Ry} \sim 5 \cdot 10^3 \text{ V/cm},$$

then in the fine-structure scheme the resonance interaction V couples only the states $|S'_{1/2} m_j\rangle$ and $|P'_{1/2} m_j + 1\rangle$, where j' is fixed by the condition for resonance. Consequently, without allowance for the HF splitting, the interaction with the resonance field is realized for each j' , m_j within the framework of a two-level system, whose quasienergies are well known.^[12] Since the matrix elements $\langle P'_{1/2} m_j + 1 | V | S'_{1/2} m_j \rangle$ are different for different m_j , the resonance interaction completely removes the degeneracy with respect to m_j in the set of functions participating in the resonance-ionization process, the

quasienergy-level spacing being characterized by the quantity $Ry \mathcal{E}_0 / \mathcal{E}_{at} \gg \Delta E_{HF}$. Therefore, further allowance for the interaction of the electronic angular momentum with the nuclear spin leads only to small corrections $\sim (\Delta E_{HF} / Ry) (\mathcal{E}_{at} / \mathcal{E}_0) \ll 1$, which can be neglected.

The solution of the problem of resonance ionization in the field range

$$\mathcal{E}_{at} \frac{\Delta E_{HF}}{Ry} \ll \mathcal{E}_0 \ll \mathcal{E}_{at} \frac{|E_{P_{3/2}} - E_{P_{1/2}}|}{Ry} \sim 5 \cdot 10^5 \frac{V}{cm}$$

can thus be carried out as a result of the allowance for the field \mathcal{E}_0 within the framework of a two-level system in the basis of the functions of the fine structure and the allowance for the \mathcal{E}_0 in first-order perturbation theory. As in Refs. 9 and 10, in this case the ionization probability can be found with the aid of the perturbation theory formula (2), where, however, the natural width Γ should be replaced by the field width

$$\Gamma_f(m_j) = 4 |\langle P'_{j'} m_{j'} + 1 | V | S^{1/2} m_j \rangle|.$$

The dependence of Γ_f on the quantum number m_j does not change the degree of polarization (which is equal to -0.6) in the case of resonance at the level $P_{1/2}$, since in this case only the term with $m_j = -1/2$ remains in the sum over m_j in (2). On the other hand, in the case of resonance at the $P_{3/2}$ level the dependence of Γ_f on m_j is important. The degree of polarization in this case turns out to be dependent on the detuning $\Delta_{j'j}$. At exact resonance (i.e., for $|\Delta_{j'j}| \ll \Gamma_f$) at $P_{3/2}$ level the probability $dw_\mu/d\Omega$ assumes the form

$$\frac{dw_\mu^{(h)}}{d\Omega} = \frac{\pi}{4\hbar} (2m^2 E)^h \sum_{m_j} \left| \langle \psi_{m_j} | V | P_{3/2} m_{j'} \rangle \right|^2 \quad (7)$$

The computation of the probability $dw_\mu/d\Omega$ with the aid of this formula in the case when the effect of the spin-orbit interaction on the magnitudes of the matrix elements is neglected yields

$$\begin{aligned} \frac{dw_{-1/2}}{d\Omega} &= \frac{e^2 \bar{\mathcal{E}}_0^2 R^2(E) \sin^2 \alpha}{128\hbar} \\ \frac{dw_{1/2}}{d\Omega} &= \frac{e^2 \bar{\mathcal{E}}_0^2 R^2(E) \sin^2 \alpha}{128\hbar} (3 \sin^2 \alpha + 4 \cos^2 \alpha), \end{aligned} \quad (8)$$

where $R(E)$ is the radial matrix element of the transition from the resonance state into the continuum, α is the angle between the direction, \mathbf{n} , of propagation of the electron and the wave vector (the z axis). The degree of polarization P , found with the aid of the formulas (3) and (8), is equal in this case to 0.6 (instead of $P = 0.82$ found in the fine-level-structure scheme in a weak field).

Finally, let us consider the region of still stronger fields

$$\mathcal{E}_0 \gg \mathcal{E}_{at} |E_{P_{3/2}} - E_{P_{1/2}}| / Ry \sim 5 \cdot 10^5 V/cm$$

where the field broadening exceeds the distance between the terms $P_{1/2}$ and $P_{3/2}$ and the resonance mixing of these terms becomes possible. If the initial state of

the atom is $|S^{1/2} 1/2\rangle$, then the resonance interaction does not, as before, fall outside the framework of the two-level system ($S^{1/2} 1/2$) and ($P_{3/2} 3/2$). This ionization channel makes a contribution to $dw_{1/2}/d\Omega$, and the corresponding expression for the probability at exact resonance is determined by the formula (7) with the m_j -summation sign dropped and m_j set equal to $+1/2$.

If, on the other hand, initially the atom is in the state $|S^{1/2}, -1/2\rangle$, then the resonance interaction is realized within the framework of the three-level system:

$$(S^{1/2}, -1/2), (P^{1/2} 1/2), (P^{3/2} 1/2).$$

The quasienergy wave function of such a system, which takes the interaction with the field \mathcal{E}_0 exactly into account in the resonance approximation, can be represented in the form

$$\Psi_{\pm} = e^{-iE_{\pm}t/\hbar} \left(e^{i\omega t} A_s \left| S \frac{1}{2}, -\frac{1}{2} \right\rangle + \sum_{j=1/2, 3/2} A_j \left| P_j \frac{1}{2} \right\rangle \right), \quad (9)$$

where E is the quasienergy and A_s and A_j are constant coefficients. The allowed quasienergy values are, as can easily be verified by substituting (9) into the Schrödinger equation, determined by the secular equation

$$(E - E_s - \hbar\omega) (E - E_{P_{1/2}}) (E - E_{P_{3/2}}) - |\langle P^{1/2} 1/2 | V | S^{1/2}, -1/2 \rangle|^2 (E - E_{P_{3/2}}) - |\langle P^{3/2} 1/2 | V | S^{1/2}, -1/2 \rangle|^2 (E - E_{P_{1/2}}) = 0, \quad (10)$$

which in the strong-field asymptotic limit

$$\mathcal{E}_0 \gg \mathcal{E}_{at} |E_{P_{3/2}} - E_{P_{1/2}}| / Ry, \mathcal{E}_{at} |\Delta| / Ry$$

has two solutions that increase in proportion to \mathcal{E}_0 :

$$\begin{aligned} E_{\pm} &= \frac{E_s + \hbar\omega}{2} + \sum_j E_{P_j} \left| \langle P_j \frac{1}{2} | V | S \frac{1}{2}, -\frac{1}{2} \rangle \right|^2 \\ &\times \left[2 \sum_j \left| \langle P_j \frac{1}{2} | V | S \frac{1}{2}, -\frac{1}{2} \rangle \right|^2 \right]^{-1} \\ &\pm \left[\sum_j \left| \langle P_j \frac{1}{2} | V | S \frac{1}{2}, -\frac{1}{2} \rangle \right|^2 \right]^h \end{aligned} \quad (11)$$

The third quasienergy eigenvalue, $E \sim (\mathcal{E}_0)^0$, is not important for the solution of the problem with the initial condition

$$\Psi(t=0) = |S^{1/2}, -1/2\rangle.$$

The wave function that satisfies this initial condition has the form

$$\begin{aligned} \Psi &= \frac{1}{2} \sum_{\pm} \exp\left(-\frac{iE_{\pm}t}{\hbar}\right) \left\{ e^{i\omega t} \left| S \frac{1}{2}, -\frac{1}{2} \right\rangle \right. \\ &\pm \sum_j \left| P_j \frac{1}{2} \right\rangle \langle P_j \frac{1}{2} | V | S \frac{1}{2}, -\frac{1}{2} \rangle \\ &\times \left[\sum_j \left| \langle P_j \frac{1}{2} | V | S \frac{1}{2}, -\frac{1}{2} \rangle \right|^2 \right]^{-1/2} \left. \right\}. \end{aligned} \quad (12)$$

Taking now into account the ionizing field $\bar{\mathcal{E}}_0$ within the framework of perturbation theory, we can also easily determine the contribution of the ionization channels

under consideration to the probability

$$\frac{dw_{\mu}}{d\Omega} = \sum_{\pm} \left\{ \frac{\pi}{4\hbar} (2m^2 E)^{1/2} \left| \sum_j \langle \psi_{\mu} | \nabla | P_j \frac{1}{2} \rangle \langle P_j \frac{1}{2} | \nabla | S \frac{1}{2}, -\frac{1}{2} \rangle \right|^2 \right. \\ \left. \times \left[\sum_j \left| \langle P_j \frac{1}{2} | \nabla | S \frac{1}{2}, -\frac{1}{2} \rangle \right|^2 \right]^{-1} \right\}. \quad (13)$$

All the quantities in (13) are evaluated at $E = E_{\pm} + \hbar\bar{\omega}$.

The final computation of the ionization probability $dw_{\mu}/d\Omega$ in the strong-field asymptotic limit with allowance for both ionization channels from the formulas (7) and (13) yields

$$\frac{dw_{\mu}}{d\Omega} = \frac{3}{64\hbar} e^2 \bar{\mathcal{E}}_0^2 \sin^4 \alpha R_{31}^2(E_0 + \hbar\bar{\omega}) + \frac{e^2 \bar{\mathcal{E}}_0^2 \cos^2 \alpha \sin^2 \alpha}{32\hbar} \frac{1}{R_1^2 + 2R_2^2} \\ \times \sum_{\pm} \left[R_1 R_{31}(E_{\pm} + \hbar\bar{\omega}) + \frac{1}{5} R_3 (R_{33}(E_{\pm} + \hbar\bar{\omega}) - 6R_{53}(E_{\pm} + \hbar\bar{\omega})) \right]^2, \quad (14)$$

$$\frac{dw_{-\mu}}{d\Omega} = \frac{e^2 \bar{\mathcal{E}}_0^2 \sin^4 \alpha}{128\hbar (R_1^2 + 2R_2^2)} \sum_{\pm} [2R_1 R_{31}(E_{\pm} + \hbar\bar{\omega}) \\ + \frac{1}{5} R_3 (2R_{33}(E_{\pm} + \hbar\bar{\omega}) + 3R_{53}(E_{\pm} + \hbar\bar{\omega}))], \quad (15)$$

where R_1 and R_3 are the radial matrix elements for the transitions, $S \rightarrow P_{1/2}$ and $S \rightarrow P_{3/2}$, between the discrete levels; $R_{31}(E)$, $R_{33}(E)$, and $R_{53}(E)$ are respectively the radial matrix elements for the transitions $P_{1/2} \rightarrow D_{3/2}$, $P_{3/2} \rightarrow D_{3/2}$, and $P_{3/2} \rightarrow D_{5/2}$ into the continuum.

The arguments of the radial matrix elements determine the value of the energy corresponding to the radial wave function, $R_{i,jE}$, of the continuous spectrum

$$E_0 = \frac{1}{2}(E_S + E_{P_{\mu}} + \hbar\bar{\omega}) - \text{sign} \Delta_{\mu, \mu} \cdot e \bar{\mathcal{E}}_0 R_3 / \sqrt{6}, \\ E_{\pm} = \frac{1}{2}(E_S + \hbar\bar{\omega}) + \frac{2R_1^2 E_{P_{\mu}} + R_3^2 E_{P_{\mu}}}{2(2R_1^2 + R_3^2)} \pm \frac{e \bar{\mathcal{E}}_0}{3 \cdot 2^{1/2}} (2R_1^2 + R_3^2)^{1/2}. \quad (16)$$

The difference between the matrix elements R_1 and R_3 and between R_{31} , R_{33} , and R_{53} is determined by the effect on their magnitudes of the spin-orbit interaction. If we neglect this small difference and set $R_1 \approx R_3$, $R_{31} \approx R_{33} \approx R_{53} \equiv R(E)$, and also neglect the small (on a scale $\sim \text{Ry}$) difference between E_0 , E_+ , and E_- , then the formulas (14) and (15) yield

$$\frac{dw_{\mu}}{d\Omega} = \frac{3e^2 \bar{\mathcal{E}}_0^2}{64\hbar} R^2(E) \sin^4 \alpha. \quad (17)$$

In these approximations the degree of polarization of the photoelectrons is equal to zero. Consequently, the resonance mixing of the $F_{1/2}$ and $F_{3/2}$ terms leads to a sharp decrease in the degree of polarization of the photoelectrons. The only exception may be the frequency region where the total probability

$$w = \sum_{\mu} w_{\mu}$$

is anomalously small. In the vicinity of the minimum of $w(\bar{\omega})$ the small difference between $w_{1/2}$ and $w_{-1/2}$ can lead to the appreciable degree of polarization (3). In the case of direct single-photon ionization this phenomenon is known as the Fano effect.^[15] It is interesting to

note that, according to (14)–(16), in the case of resonance ionization in a strong field, appreciable polarization effects are possible in the vicinity of the minimum $w(\bar{\omega})$ not only on account of the difference in the magnitudes of the matrix elements, as in the normal Fano effect, but also on account of the difference in the arguments of $R(E)$, i.e., on account of the difference in the quasienergies of the wave functions participating in the ionization process.

In the absence of such type of interference phenomena, which are possible only at a specially selected value of the frequency $\bar{\omega}$, the optimum conditions for the production of highly polarized electrons are realized in the case of short pulses, i.e., pulses of width $\tau < \hbar/\Delta E_{\text{HF}} \sim 10^{-8}$ sec, and not too high intensities, i.e., $\mathcal{E}_0 < \mathcal{E}_{\text{at}} (\hbar/\tau \text{ Ry}) \sim 5 \times 10^5$ V/cm. The raising of the intensity \mathcal{E}_0 to values $\sim 5 \times 10^5$ V/cm leads almost to the total disappearance of the polarization effects and is from this point of view absolutely inadmissible.

4. POLARIZATION BROADENING AND NONRESONANCE MIXING OF THE LEVELS

Let us now assume that the exciting field, \mathcal{E}_0 , is weak, i.e., that $\mathcal{E}_0 \ll \mathcal{E}_{\text{at}}^2/\mathcal{E}_{\text{at}}$, and, as in the preceding section, consider the changes in the electron polarization to which the growth of the ionizing-field intensity \mathcal{E}_0 leads. It should then be borne in mind that, generally speaking, in the case of single-photon transitions from the resonance states into the continuum the magnitude of the level shift due to the dynamic Stark effect has the same order of magnitude as the level broadening due to the ionization. Nevertheless, sometimes the polarizabilities can be anomalously small, and this allows us to neglect the level shift. On the other hand, if the transition from the resonance states into the continuum is accomplished as a result of the absorption of several photons ($k', k' > 1$), then the ionization width is small in the ratio $(\mathcal{E}_0/\mathcal{E}_{\text{at}})^{2(k'-1)} \ll 1$ in comparison with the quadratic level shift. Let us first consider the case of long pulses (i.e., pulses of width $\tau \gg \hbar/\Delta E_{\text{HF}}$), which allow the resolution of the HF structure. The perturbation theory in the HF level structure scheme is valid when

$$\bar{\mathcal{E}}_0 \ll \mathcal{E}_{\text{at}} (\hbar/\tau \text{ Ry})^{1/2}.$$

In the field range

$$\mathcal{E}_{\text{at}} (\hbar/\tau \text{ Ry})^{1/2} \ll \bar{\mathcal{E}}_0 \ll \mathcal{E}_{\text{at}} (\Delta E_{\text{HF}}/\text{Ry})^{1/2} \sim 10^8 \text{ V/cm}$$

there is realized a regime of ionization via a single level of the HF structure under conditions of ionization broadening. As in Refs. 9 and 10, the ionization probability in this case is determined by the formula (1), in which the natural width, Γ , should be replaced by the ionization width

$$\Gamma_+(m_{\mu}) = 2\pi (2m^2 E)^{1/2} \int d\Omega \sum_{\mu, \mu'} |\langle \psi_{\mu, m_{\mu}} | \nabla | F' m_{\mu} + 1 \rangle|^2, \quad (18)$$

and the level shift should be allowed for in the detuning $\Delta_{F', F}$:

$$\Delta_{F',F} \rightarrow \Delta_{F',F}(m_F) = E_{F',F} - E_{SF} - \hbar\omega, \quad (19)$$

$$E_{F',F} = E_{F',F} - 1/4 \alpha'(m_F) \mathcal{E}_0^2, \quad E_{SF} = E_{SF} - 1/4 \alpha(m_F) \mathcal{E}_0^2,$$

where $\alpha'(m_F)$ and $\alpha(m_F)$ are the dynamic polarizabilities of the levels $(Pj'F'm_F + 1)$ and $(S1/2Fm_F)$ at the frequency ω .

The dependence on m_F of the width Γ_i and the detuning $\Delta_{F',F}$ is the reason why in the general case the degree of polarization depends, under ionization-broadening conditions, on the frequency (varies on a scale $\sim \Gamma_i$).

The calculation of the ionization width for the sub-levels of the $P_{1/2}$ multiplet of the Cs atom ($I=7/2$) yields

$$\Gamma_i(m_F) = \frac{\pi}{48} e^2 \mathcal{E}_0^2 R_{31}^2(E) \begin{cases} 11 - m_F, & F'=3 \\ 13 + m_F, & F'=4 \end{cases} \quad (20)$$

If the range of variation of the polarizability difference $\alpha' - \alpha$ as m_F is varied (but at fixed F and F') is substantially less than $e^2 R_{31}^2(E)$, then the detuning $\Delta_{F',F}$ can be assumed to be independent of m_F , and this allows us to choose the frequency ω such that it is a near-resonance frequency for all m_F simultaneously. Let us emphasize that, in essence, here we have in mind the polarizability difference, which can be small even at not too small values of the polarizabilities, provided the range of variation of the shift of the resonance frequency (with varying m_F) is substantially smaller than the characteristic shift value alone.

At exact resonance the ionization probability, $dw_\mu/d\Omega$, for the atom is determined by the expression

$$\frac{dw_\mu}{d\Omega} = \frac{4\pi(2m^2E)^{\hbar}}{h} \sum_{m_F m_F'} \frac{1}{\Gamma_i^2(m_F)} \times |\langle \psi_{Pj'm_F} | \nabla | P_{1/2} F' m_F + 1 \rangle \langle P_{1/2} F' m_F + 1 | \nabla | S F m_F \rangle|^2. \quad (21)$$

The results of the calculation of the degree of polarization P_i with the aid of the formulas (20), (21) for $z=1/2$ are given in Table II. A comparison with the results presented in Table I shows that the divergence from the degree of polarization in the HF structure scheme in a weak field is relatively small, which can be explained by circumstances of numerical character.

In the region of fields of higher intensities, i.e., for

$$\mathcal{E}_0 \gg \mathcal{E}_{at} (\Delta E_{HF}/Ry)^{\hbar} \sim 10^8 \text{ V/cm},$$

the shift of the levels and their ionization broadening exceed the scale of the HF splitting ΔE_{HF} , as a result of which the description of the resonance-ionization process in the fine level structure scheme with allowance for the ionization broadening of the levels evidently becomes adequate (no rigorous proof of this assertion exists at present). Before proceeding to the characteristics of this field region, let us note that the above-considered cases are not realized at all in the case of short pulses, i.e., pulses with $\tau \ll \Delta E_{HF}/\hbar$. In this case the perturbation theory in the fine level structure scheme is valid when $\mathcal{E}_0 \ll \mathcal{E}_{at} (\hbar/\tau Ry)^{1/2}$, while in the range of fields

$$\mathcal{E}_{at} (\hbar/\tau Ry)^{\hbar} \ll \mathcal{E}_0 \ll \mathcal{E}_{HF} (|E_{P_{1/2}} - E_{P_{3/2}}|/Ry)^{\hbar} \sim 5 \cdot 10^7 \text{ V/cm}$$

there is realized a regime of ionization via the isolated levels of the fine structure under conditions of their ionization broadening. The ionization probability can then be found with the aid of the formula (2), in which the ionization width of the $(Pj'm_j + 1)$ level,

$$\Gamma_i(j'm_j) = 2\pi(2m^2E)^{\hbar} \int d\Omega \sum_{\mu} |\langle \psi_{Pj'm_j} | \nabla | Pj'm_j + 1 \rangle|^2, \quad (22)$$

should be used in place of Γ and the Stark shift should be allowed for in the detuning $\Delta_{j'j}$:

$$\Delta_{j'j} \rightarrow \Delta_{j'j} = \Delta_{j'j} - 1/4 \mathcal{E}_0^2 (\alpha'(m_j) - \alpha(m_j)), \quad (23)$$

where $\alpha'(m_j)$ and $\alpha(m_j)$ are the dynamic polarizabilities of the $(Pj'm_j + 1)$ and $(S, j=1/2, m_j)$ levels of the fine structure.

In the case of resonance at the $P_{1/2}$ level the modification of the resonance denominator in the formula (2) does not affect the degree of ionization, which is equal to $\sim 60\%$, since only the term with $m_j = -1/2$ is retained in the sum over m_j . If, on the other hand, the $P_{3/2}$ level is the resonant level, then the dependence of $\Delta_{j'j}$ and Γ_i on m_j significantly affects the degree of polarization P , which now depends on the magnitude of the detuning. If the polarizability difference $\alpha' - \alpha$ computed with $m_j = -1/2$ differs little (in comparison with $e^2 R^2(E)$) from this difference found with $m_j = +1/2$, then the frequency can be chosen such that it is resonant with respect to both transitions (from the $m_j = -1/2$ and $m_j = +1/2$ sub-levels) simultaneously. Under these conditions (exact resonance), the computation of the probabilities $dw_\mu/d\Omega$ yields

$$\frac{dw_{-1/2}}{d\Omega} = \frac{25}{256\pi^2 \hbar} \left(\frac{R_3 \mathcal{E}_0}{R(E) \mathcal{E}_0} \right)^2 \sin^4 \alpha, \quad \frac{dw_{1/2}}{d\Omega} = \frac{25}{64\pi^2 \hbar} \left(\frac{R_3 \mathcal{E}_0}{H(E) \mathcal{E}_0} \right)^2 \sin^2 \alpha. \quad (24)$$

The degree of polarization in this case is equal to 67% , instead of the 82% in the weak-field case in the fine level structure scheme.

Let us turn, finally, to the region of very strong fields, $\mathcal{E}_0 > 5 \times 10^7 \text{ V/cm}$, in which the nonresonance mixing of the $P_{1/2}$ and $P_{3/2}$ terms can be realized. If the initial state of the atom is $|S1/2 1/2\rangle$, then, as before, the process of resonance ionization occurs via the single level $(P3/2 3/2)$, which is characterizable by the shift due to the dynamic Stark effect and the ionization width $\Gamma_i(3/2 1/2)$, (22). This ionization channel makes a contribution to the probability for the production of a photoelectron with spin component $\mu=1/2$, this probability being equal in the strong-field asymptotic limit (or in the vicinity of exact resonance where $|\Delta| \ll \Gamma_i \sim Ry(\mathcal{E}_0/\mathcal{E}_{at})^2$) to

$$\frac{dw_{1/2}}{d\Omega} = \frac{25}{64\pi^2 \hbar} \left(\frac{\mathcal{E}_0 R_1}{\mathcal{E}_0 R(E)} \right)^2 \sin^4 \alpha. \quad (25)$$

If, on the other hand, initially the atom is in the state $|S1/2, -1/2\rangle$, then the picture of the interaction with the intense external field looks more complicated. The normal approach to the description of virtual nonresonance effects consists in the separation of the fast and

slow motions in the Schrödinger equation,^[16] which allows us, in principle, to write down the matrix, $Q_{jj'}(m_F)$, that brings about the reconstruction of the multiplets (the nonresonance mixing).^[14]

With allowance for the ionization, the matrix $Q_{jj'}$ is non-Hermitian. To solve the problem of resonance ionization, it is necessary to diagonalize this matrix, the complex eigenvalues of which determine the positions and widths of the quasienergy levels in the external periodic field. The corresponding eigenfunctions of the Q matrix are quasistationary, quasienergy functions of the atom. The ionization probability can be determined after this if allowance is made within the framework of perturbation theory for the transitions from the ground state into the found quasienergy resonance states under the action of the weak exciting field. Actually, such a procedure can be carried out with the aid of a method based on the application of the Laplace transformation^[17] to the time-dependent amplitudes of the expansion of the quasienergy wave functions in terms of the components of the multiplets of the free atom if we limit ourselves to the quadratic nonresonance Stark effect. Omitting the details of these calculations, in view of their unwieldiness, let us only point out that in the strong-field asymptotic limit

$$\tilde{\mathcal{E}}_0 \rightarrow \tilde{\mathcal{E}}_{at} (|E_{P_n} - E_{P_n}|/Ry)^{1/2}, \tilde{\mathcal{E}}_{at} (|\Delta|/Ry)^{1/2}$$

the ionization channel corresponding to the initial state $|S1/2, -1/2\rangle$ leads to the appearance of electrons with $\mu = -1/2$; moreover,

$$dw_{-1/2}/d\Omega = dw_{1/2}/d\Omega, \quad (26)$$

where $dw_{-1/2}/d\Omega$ is determined by Eq. (25) if the ionization broadening predominates over the shift and splitting of the quasienergy levels. It follows from the formula (26) that the polarization of the electrons vanishes in this case. Physically, this result is understandable, and is connected with the fact that the level broadening (in the present case, ionization broadening) is greater than the multiplet-component spacing. Under these conditions we can neglect completely the spin-orbit interaction. Since it is precisely the spin-orbit interaction that is the mechanism determining the possibility of producing polarized electrons, it is natural that the polarization effects should turn out to be suppressed under conditions of strong broadening.

The assumption made above that the ionization width predominates over the Stark shift and the splitting of the levels can be realized at definite frequencies where the polarizabilities α and α' are small. As a rule, however, it is the opposite situation that obtains: the real part of the matrix elements $Q_{jj'}$ is usually large compared to the ionization widths of the levels. In the $k' = 1$ case this is a numerical effect; in the $k' > 1$ case, however, the relative smallness of the ionization width is characterized by the parameter

$$(\tilde{\mathcal{E}}_0/\tilde{\mathcal{E}}_{at})^{2(k'-1)} \ll 1.$$

The correct description of the reconstruction of the spectra of the multiplets on account of the nonresonance

Stark effect in very strong fields ($\mathcal{E}_0 > 10^7$ V/cm) can be extremely difficult. This is connected with the fact that in such fields the perturbation-series expansion may no longer be valid. As a result, the allowance for only the quadratic Stark effect may not be sufficient. The matrix $Q_{jj'}(m_F)$ may be determined by quantities of higher order in \mathcal{E}_0 , which in the case of isolated levels is equivalent to the necessity for allowance for the hyperpolarizabilities.^[14] However, we can, without performing a detailed quantitative analysis, draw some qualitative conclusions about the characters of the spectrum, the resonance-ionization process, and the polarization phenomena in so strong fields. In view of the smallness of the ionization broadening in comparison with the distance between the quasienergy levels (the eigenvalues of the matrix Q), the frequency ω can be chosen to be resonant with respect to the transition into one of the exact quasienergy states. In other words, the resonance-multiplet structure cannot be neglected in this case, but this structure is determined not by the spin-orbit interaction, but by the dynamic Stark effect in the strong ionizing field.

Since the exciting field feels this multiplet structure, the production of polarized electrons is also possible in this case. Thus, for example, in the S - P - D transition scheme the frequency ω can be chosen to be resonant with respect to the transition to the $|P3/2\ 3/2\rangle$ state, i.e., in the quadratic-Stark-effect approximation

$$\hbar\omega \approx E_{P_n} - E_s - \frac{1}{2}(\alpha'(\frac{1}{2}) - \alpha(\frac{1}{2}))\tilde{\mathcal{E}}_0^2. \quad (27)$$

In this case the resonance-ionization process proceeds only via the $(P3/2\ 3/2)$ level. All the rest of the quasienergy sublevels are in this case nonresonant and are not excited by the field \mathcal{E}_0 . Under these conditions the degree of polarization is, within the framework of the assumed idealizations, equal to 100%. This result confirms the fact that by itself the nonresonance mixing of the levels (without broadening) does not, generally speaking, lead to the loss of photoelectron polarization; on the contrary, it can even increase the degree of polarization by tuning out the group of "extraneous" levels from resonance.

Let us draw attention to the profound difference, which becomes apparent in these conclusions, between the resonance and nonresonance mixings. Although both of these phenomena can be regarded as a rearrangement and shifting of the quasienergy levels in the external field, their roles in the resonance-ionization process are quite different. Whereas resonance mixing can, under suitable conditions, manifest itself as a broadening of the resonance dependence of the ionization probability on the detuning, nonresonance mixing does not possess such a property. It leads to a shift in the position of the resonances and to a regrouping of the wave functions, but it is not a mechanism determining the resonance width.

5. CONCLUSION

Thus, the above-performed analysis allows us to identify two main factors as the most dangerous from

the standpoint of the use of the resonance ionization of atoms as a source of polarized electrons, to wit: resonance mixing of the spectral terms and their ionization broadening. In the case of two-photon resonance ionization the corresponding limitations on the magnitudes of the exciting- and ionizing-field intensities have the form $\mathcal{E}_0 \ll 5 \times 10^5$ V/cm, $\tilde{\mathcal{E}}_0 \ll 5 \times 10^7$ V/cm. The optimum pulse width should in this case be confined to the interval $10^{-9} - 10^{-8} < \tau < 10^{-11} - 10^{-12}$. The limitation on the ionizing-field intensity $\tilde{\mathcal{E}}_0$ is not absolute, and is, in fact, nonexistent if the broadening of the levels is accompanied by a substantial shift of them on account of the dynamic Stark effect. The most typical in this respect is the ionization process in which the transition from the resonance states into the continuum is accompanied by the absorption of several ($k' > 1$) $\hbar\tilde{\omega}$ quanta. The smallness of the ionization width ($\sim \tilde{\mathcal{E}}_0^{2k'}$) compared to the quadratic shift leads in this case to the possibility of resonance excitation of the individual quasienergy sublevels of the atom in the strong field $\tilde{\mathcal{E}}_0$ and to large values of the degree of polarization P . The latter result is, evidently, very important, since it indicates the possibility in principle of the production of highly polarized photoelectrons in very strong fields $\tilde{\mathcal{E}}_0 \gtrsim 5 \times 10^7$ V/cm, such as are actually commonly used in experiments on the multiphoton ionization of atoms.^[18]

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