Stark effect in hyperfine structure sublevels and splitting of $n^2S_{1/2}$ states of alkali atoms in a nonresonant optical field

N. L. Manakov and V. D. Ovsyannikov

Lenin Komsomol State University, Voronezh

(Submitted 24 February 1978)
Zh. Eksp. Teor. Fiz. 75, 803–815 (September 1978)

The change in the energy of an isolated atomic level in a nonresonant optical field is expressed in terms of scalar, tensor, and vector polarizabilities proportional to the amplitudes of the scalar, symmetric, and antisymmetric scattering of light. Expressions are obtained for the polarizabilities allowing for the hyperfine structure of levels and a study is made of the Stark effect in hyperfine structure sublevels in the field of a wave with an arbitrary polarization. It is shown that in the case of circular polarization the splitting of the ground states of alkali atoms may reach $\sim 1\ \mathrm{cm}^{-1}$ in a field of $\sim 10^6$ V/cm. The role of this effect in experiments on resonant multiphoton ionization is considered. It is shown that the splitting of the $S_{1/2}$ states gives rise to a high degree of polarization of photoelectrons and nuclei in the case of resonant multiphoton ionization of atoms. The degree of polarization of photoelectrons and nuclei is calculated for the four-photon ionization of cesium by neodymium laser radiation.

PACS numbers: 32.60. + i, 32.80.Fb, 31.30.Gs

1. INTRODUCTION

Perturbations of the atomic spectra by optical fields are being investigated intensively from the experimental and theoretical points of view (see the review by Delone *et al.*[1]). In most of the investigations of the shift and splitting of levels in a nonresonant field it is usual to ignore the hyperfine structure (hfs) of an atom. However, the recent rapid growth of nonlinear laser spectroscopy methods[2] makes it possible to investigate experimentally the Stark effect involving individual hfs sublevels.[3,4] We recall that the quadratic Stark effect in hfs levels in a static electric field was first measured in 1957 in cesium[5] and subsequently investigated in detail in other alkali atoms.[6] Experiments on the optical pumping revealed that the Stark effect in hfs sublevels was observed even in optical fields created by conventional sources because light was resonant.[7,8]

We shall consider the nonresonant Stark effect involving hfs levels of alkali atoms, consider the characteristics of this effect in the case of linearly and circularly polarized waves, and discuss the question of the splitting of the $n^2S_{1/2}$ states in the field of a wave with a finite degree of circular polarization, as well as possible manifestations of this effect in resonant multiphoton ionization. In particular, we shall discuss the possibility of generating polarized photoelectrons and nuclei by resonant ionization of field-split hfs sublevels of alkali atoms.

Since different definitions of the atomic parameters representing the level shifts and splittings have been used in calculations of the ac Stark effect (see, for example, Refs. 8–10), we shall use the technique of irreducible tensor operators to show generally (in Sec. 2) that a quantitative description of the perturbation of a spectrum in a monochromatic field is best given by introducing scalar, vector, and tensor dynamic polarizabilities of an atom, related closely to the parameters of the Rayleigh scattering of light and corresponding to the characteristics of the static Stark effect.

2. INVARIANT ATOMIC PARAMETERS GOVERNING LEVEL SHIFTS AND SPLITTING

We shall consider an isolated level $|n\rangle = |\gamma FM\rangle$ of an atomic multiplet formed by sublevels with different values of the total momentum F and we shall assume that this level is acted upon by the field of a wave with an electric vector

$$\mathbf{E}(t) = \mathscr{E}\,\mathrm{Re}\{\mathbf{e}e^{i(\mathbf{kr}-\omega t)}\}, \quad \mathbf{ee}^* = 1,$$

where M is the projection of the total momentum onto the quantization axis and γ are the other quantum numbers of the level. The momentum **F** may, depending on the type of the problem, be a sum of the electron **J** and nuclear **I** momenta (this applies to the Stark effect involving hfs) or it may be the electron momentum in the LS coupling: $\mathbf{F} \equiv \mathbf{J} = \mathbf{L} + \mathbf{S}$ (this applies to the Stark effect involving fine structure levels), etc.

Since we are interested in the range of optical frequencies ω, we shall describe the interaction of an atom with the field by the dipole approximation. Moreover, the frequency ω is assumed to be off resonance, so that the perturbation of the levels by the field is quadratic in $\mathscr{E}$. On application of the perturbation theory to quasienergies,[11,12] the change in the energy $\Delta E_n = E - E_n$ for a level $|\gamma F\rangle$ which is $(2F+1)$-fold degenerate in respect of M can be found from the secular equation (the atomic system of units is employed):[1)]

$$\det\|\Delta E_n \delta_{MM'} - {}^1/_4 \mathscr{E}^2 U_{MM'}\| = 0, \tag{1}$$

where

$$U_{MM'}(\omega; \mathbf{e}, \mathbf{e}^*) = \Big\langle \gamma FM \Big| \underset{n'}{S} \Big\{ \frac{(\mathbf{ed})|n'\rangle\langle n'|(\mathbf{e}^*\mathbf{d})}{\omega_{nn'} - \omega + i\eta} + \frac{(\mathbf{e}^*\mathbf{d})|n'\rangle\langle n'|(\mathbf{ed})}{\omega_{nn'} + \omega + i\eta} \Big\} \Big| \gamma FM' \Big\rangle \tag{2}$$

is the amplitude of the Rayleigh scattering[13] and **d** is the dipole moment operator. The symbol S denotes

0038-5646/78/090406-07$02.40

summation over the discrete spectrum and integration over the continuous spectrum of an atom n'. The correction $\eta=+0$ determines the rule for bypassing the poles in the integration over the continuous spectrum in the $\omega > |E_n|$ case, so that the imaginary part of ΔE_n associated with the photoionization probability results in decay of the level $|n\rangle$.

Applying the technique of irreducible tensor operators,[14] we can conveniently transform $U_{MM'}$ to

$$U_{MM'}(\omega;\mathbf{e},\mathbf{e}^*)=\sum_{p=0,1,2} C^{FM'}_{FMpm}\alpha_p(\omega)\{\mathbf{e}\otimes\mathbf{e}^*\}_{pm}, \tag{3}$$

where

$$\{\mathbf{e}\otimes\mathbf{e}^*\}_{pm}=\sum_{i,k} C^{pm}_{1i1k}e_i(\mathbf{e}^*)_k$$

is the polarization tensor of the wave,[15] e_j are the spherical projections of $\mathbf{e}$ in the coordinate system with the z axis along the quantization axis of the atom, α_p is the combination of the reduced composite matrix elements:[2)]

$$\alpha_p(\omega)=\left(\frac{2p+1}{2F+1}\right)^{1/2}\sum_{F'=F,F\pm1}(-1)^{F+F'}\begin{Bmatrix}1&1&p\\F&F&F'\end{Bmatrix}$$
$$\times\mathop{\mathrm{S}}_{\gamma'}|\langle\gamma F\|\mathbf{d}\|\gamma'F'\rangle|^2\left\{\frac{1}{\omega_{nn'}-\omega+i\eta}+\frac{(-1)^p}{\omega_{nn'}+\omega+i\eta}\right\}, \tag{4}$$

$\begin{Bmatrix}a&b&c\\\alpha&\beta&\gamma\end{Bmatrix}$ is the $6j$ symbol.

The meaning of the parameters $\alpha_p(\omega)$ can be found from the cross section for the Rayleigh scattering of light by an atom in a state $|\gamma F\rangle$:

$$\frac{d\sigma}{d\Omega_{\mathbf{e}_2}}=\frac{1}{2F+1}\sum_{MM'}|U_{MM'}(\omega;\mathbf{e}_1,\mathbf{e}_2^*)|^2(\alpha\omega)^4,\quad \alpha\approx\frac{1}{137},$$

which, subject to Eq. (3), can be represented in the form

$$\frac{d\sigma}{d\Omega_{\mathbf{e}_2}}=\sum_p\frac{|\alpha_p(\omega)|^2}{2p+1}\sum_m|\{\mathbf{e}_1\otimes\mathbf{e}_2^*\}_{pm}|^2(\alpha\omega)^4$$
$$=\{{}^1/{}_3|\alpha_0|^2|\mathbf{e}_1\mathbf{e}_2^*|^2+{}^1/{}_6|\alpha_1|^2(1-|\mathbf{e}_1\mathbf{e}_2|^2)$$
$$+{}^1/{}_{10}|\alpha_2|^2(1+|\mathbf{e}_1\mathbf{e}_2|^2-{}^2/{}_3|\mathbf{e}_1\mathbf{e}_2^*|^2)\}(\alpha\omega)^4, \tag{5}$$

where $\mathbf{e}_{1,2}$ are the polarization vectors of the incident and scattered photons. A comparison of Eq. (5) with Eq. (6.17) in the monograph of Berestetskiĭ *et al.*[13] shows that $\alpha_p(\omega)$ with $p=0$, 1, and 2 represents, respectively, the amplitudes of the scalar, antisymmetric, and symmetric scattering.[3)] Thus, the scattering processes and the atomic spectrum are governed by the same set of three invariant atomic parameters α_p in Eq. (3), which depend only on ω and the quantum numbers γ and F. Then, an investigation of the spectral characteristics allows us to find not only the absolute value but also the sign of α_p.

If $\omega=0$, the parameters $\alpha_0(0)$ and $\alpha_2(0)$ can be expressed in terms of the scalar α^S and tensor α^T polarizabilities, which define the Stark effect in a static field:[17]

$$\alpha^S=\frac{\alpha_0(0)}{3^{1/2}},\quad \alpha^T=-\alpha_2(0)\left[\frac{2F(2F-1)}{3(F+1)(2F+3)}\right]^{1/2}.$$

These relationships can be applied conveniently also to the ac effect.

The parameter α_1 corresponding to $\omega=0$ vanishes and it has a finite value only in an ac field. Then, the contribution of the term with α_1 to ΔE_n differs from zero only for a finite degree of circular polarization of the incident wave, because in Eq. (3) we have

$$\{\mathbf{e}\otimes\mathbf{e}^*\}_1=2^{-1/2}[\mathbf{e}\times\mathbf{e}^*]=2^{-1/2}A\mathbf{n},$$

where $\mathbf{n}$ is the direction of propagation of the wave and A the degree of its circular polarization.[13]

By analogy with α^S and α^T, it is convenient to introduce the parameter

$$\alpha^a(\omega)=-\alpha_1(\omega)\left(\frac{2F}{F+1}\right)^{1/2},$$

which can naturally be called the vector (or antisymmetric) polarizability. The final expressions for the polarizabilities in terms of the reduced matrix elements

$$\mathfrak{M}^{(\pm)}_{F,F'}=-\mathop{\mathrm{S}}_{\gamma}|\langle\gamma F\|\mathbf{d}\|\gamma'F'\rangle|^2\left\{\frac{1}{\omega_{nn'}+\omega+i\eta}\pm\frac{1}{\omega_{nn'}-\omega+i\eta}\right\}$$

have the form

$$\left.\begin{aligned}\alpha^S(\omega)&=\frac{1}{3(2F+1)}\sum_{F'=F,F\pm1}\mathfrak{M}^{(+)}_{F,F'},\\ \alpha^a(\omega)&=\frac{1}{(F+1)(2F+1)}[(F+1)\mathfrak{M}^{(-)}_{F,F-1}+\mathfrak{M}^{(-)}_{F,F}-F\mathfrak{M}^{(-)}_{F,F+1}],\\ \alpha^T(\omega)&=-\frac{1}{3(2F+1)}\left[\mathfrak{M}^{(+)}_{F,F-1}-\frac{2F-1}{F+1}\mathfrak{M}^{(+)}_{F,F}+\frac{F(2F-1)}{(F+1)(2F+3)}\mathfrak{M}^{(+)}_{F,F+1}\right].\end{aligned}\right\} \tag{6}$$

Using the expansion (3), we can easily check that the matrix $U_{MM'}$ is diagonal for the linear and circular wave polarizations if the quantization axis of an atom is selected to be, respectively, the direction of the polarization vector $\mathbf{e}$ and the direction of propagation $\mathbf{n}$. The corresponding expressions for ΔE_n then become

$$\Delta E_n=-\frac{1}{4}\mathscr{E}^2\left\{\alpha_{\gamma F}{}^S(\omega)+\alpha_{\gamma F}{}^T(\omega)\frac{3M^2-F(F+1)}{F(2F-1)}\right\} \tag{7}$$

(for a linearly polarized wave) and

$$\Delta E_n=-\frac{1}{4}\mathscr{E}^2\left\{\alpha_{\gamma F}{}^S(\omega)-A\alpha_{\gamma F}{}^a(\omega)\frac{M}{2F}-\alpha_{\gamma F}{}^T(\omega)\frac{3M^2-F(F+1)}{2F(2F-1)}\right\} \tag{8}$$

(for a circularly polarized wave), where $A=\pm1$ applies to the right-handed (and left-handed) polarizations.

Thus, the scalar polarizability determines the shift of the center of gravity of a level $|\gamma F\rangle$ (averaging of M eliminates the terms with α^a and α^T), whereas α^a and α^T represent splitting into sublevels with different values of M. It is interesting to note that the "triangle rule" for the $6j$ symbol in Eq. (4) shows that for $F=\frac{1}{2}$ we have $\alpha^T_{F=1/2}\equiv0$ and, generally speaking, $\alpha^a_{F=1/2}$ differs from zero and—in contrast to the static field case—causes splitting of the level with $F=\frac{1}{2}$ into sublevels with $M=\pm\frac{1}{2}$.

The projection M is not conserved in the general case of elliptic polarization. The diagonal elements of Eq. (1) in the coordinate system with the quantization axis along $\mathbf{n}$ have the form of Eq. (8) with $|A|<1$ and the nondiagonal elements differ from zero for $M=M\pm2$ and are given by

$$U_{M,M\pm2}=-\frac{3}{4}l\alpha_{\gamma F}{}^T(\omega)\frac{[(F\pm M+1)(F\pm M+2)(F\mp M)(F\mp M-1)]^{1/2}}{F(2F-1)}, \tag{9}$$

where l is the degree of linear polarization ($A^2+l^1=1$). A detailed analysis of the Stark effect in an elliptically polarized field is given by Zon.[10]

In the range of frequencies lower than the ionization potential ($\omega<|E_\gamma|$) the summation over the intermediate states in $\mathfrak{M}_{F,F'}$ can be carried out using the expression for the Green function of an atom in the one-electron approximation. The most convenient for this purpose is the Green function obtained in the model potential method,[18,19] which represents a further development and generalization of the well-known semiempirical quantum defect method applied to the problem of calculation of complex spectral sums in the perturbation theory. Calculations indicate that for $\omega<|E_\gamma|$ the quantities $\alpha^{S,a,T}$ exhibit a strong frequency dependence and the relationship between them can vary within wide limits.

If $\omega>|E_\gamma|$ (excited states), the expressions in Eq. (6) are practically unsuitable for calculations because of the mutual compensation of the terms $\mathfrak{M}^{(\pm)}$ with $+\omega$ and $-\omega$, and one has to use the formula for $\alpha_p(\omega)$ expressed in terms of the matrix elements of the momentum. Then, the sum over the virtual states is calculated by direct summation over the discrete spectrum and integration over the continuous spectrum of an atom. An analysis shows that at high frequencies we have

$$\alpha^s\propto-1/\omega^2,\ \alpha^a\propto\omega^{-11/2},\ \alpha^T\propto\omega^{-6},$$

so that $\alpha^{a,T}$ decreases rapidly in the range $\omega\gg|E_\gamma|$ and α^S corresponds to a change in E_γ by an amount equal to the average vibrational energy in the presence of the wave. The qualitative behavior of the parameters $\alpha(\omega)$ is shown in Fig. 1 for the $2P$ state of hydrogen.

3. STARK EFFECT IN HYPERFINE STRUCTURE

The action of a field on hfs multiplets not only shifts the center of gravity of the spectrum by

$$\Delta E_{\gamma J}=-{}^1/_4\alpha_{\gamma J}{}^s(\omega)\mathscr{E}^2$$

(**J** is the electron momentum; **the total atomic momentum F** governing the hfs includes the nuclear spin **I**:**F** =**I** +**J**), but also alters the interval between the hfs sublevels by an amount $\delta f=\Delta E_{\gamma JF}-\Delta E_{\gamma JF'}$ and causes splitting of the magnetic sublevels of the level F:

$$\delta\nu_{MM'}=\Delta E_{FM}-\Delta E_{FM'}.$$

Clearly, because of the smallness of the hfs parameter $\alpha^2\mu_I$ (μ_I is the nuclear magnetic moment), the quantities δf and $\delta\nu$ are several orders of magnitude smaller than $\Delta E_{\gamma J}$. In view of the smallness of the hyperfine splitting $\Delta_{\rm hf}$ of the atomic levels, we find that in the case of the level with $J>\frac{1}{2}$ belonging to an hfs multiplet the nondiagonal matrix element $\frac{1}{4}\mathscr{E}^2U_{\gamma JF,\gamma JF'}$ of the (2) type is proportional to $\frac{1}{4}\mathscr{E}^2\alpha_{\gamma J}{}^T(\omega)$ and determines the mixing between the hfs sublevels with different values of F and the field-induced suppression of the hfs becomes of the order of $\Delta_{\rm hf}$ even infields of a few hundreds of volts per centimeter. This makes it impossible to investigate the Stark effect of isolated hfs levels with $J>\frac{1}{2}$ in strong fields and makes it much more difficult to detect this effect experimentally in weak fields because of the smallness ($\sim\mathscr{E}^2$) of the absolute values of δf and $\delta\nu$. However, for states with $J=\frac{1}{2}$, we have $\alpha^T_{n,1/2}=0$, so that $U\approx\alpha^2\mu$ and the mixing of the $F=I\pm\frac{1}{2}$ doublet does not occur right up to fields $\mathscr{E}\lesssim\mathscr{E}_{\rm at}$. This is the situation which occurs in hfs doublets of the ground $n^2S_{1/2}$ states of alkali atoms and hydrogen for which the values of δf and $\delta\nu$ in a static field $\mathscr{E}\approx10^4-10^5$ V/cm have been measured with a high accuracy by the rf spectroscopic method (for bibliography see Ref. 6). In view of this, we shall consider only the ac-field-induced shift and splitting of the hfs sublevels of the $n^2S_{1/2}$ state with $|\gamma F\rangle\equiv|n,\ J=\frac{1}{2},\ L=0,\ F=I\pm\frac{1}{2}\rangle$.

Calculating $\mathfrak{M}_{F,F'}$ in Eq. (6), assuming the vector coupling of the momenta **I** and **J**, and allowing for the hyperfine interaction in accordance with the perturbation theory, we find that the parameters $\alpha^{S,a,T}$ can be represented in the form

$$\begin{gathered}\overset{s}{\alpha}_{n,F}(\omega)=\overset{s}{\alpha}_{n,J=1/2}(\omega)+\alpha^2\mu_I(-1)^{F-I-1/2}\frac{2I+2}{2F+1}\beta_n{}^s(\omega),\\ \alpha_{n,F}(\omega)=(-1)^{F-I-1/2}\frac{2F}{2I+1}\overset{a}{\alpha}_{n,J=1/2}(\omega)+\alpha^2\mu_I\frac{2F}{2F+1}\frac{2I+2}{2I+1}\beta_n{}^a(\omega)\\ \overset{T}{\alpha}_{n,F}(\omega)=(-1)^{F-I+1/2}\alpha^2\mu_I\frac{F(2F-1)}{I(2I+1)}\beta_n{}^T(\omega).\end{gathered}\tag{10}$$

Here $\alpha^{S,a}_{n,J=1/2}$ are the scalar and vector polarizabilities in the absence of hfs and the parameters $\beta_n(\omega)$ are independent of the quantum numbers F and I but are governed by the structure of a specific atom and by ω. We shall now give the values of $\alpha^{S,a}_{n,J=1/2}$ for rubidium and cesium at the frequencies of ruby ($\omega_R=14\,400$ cm^{-1}) and neodymium ($\omega_N=9440$ cm^{-1}) lasers:

	Cs : ω_N	Cs : ω_R	Rb : ω_N	Rb : ω_R
α^S :	1220	—720	706	—1146
α^a :	210	101	29	112

The parameters β_n of these atoms are $\approx10^5$ atomic units (a.u.). For hydrogen, we have $\alpha^S\approx4.5$, $\alpha^a\approx10^{-6}$, and $\beta\approx20$. The numerical calculations were carried out by a technique similar to that used in the static field case.[6]

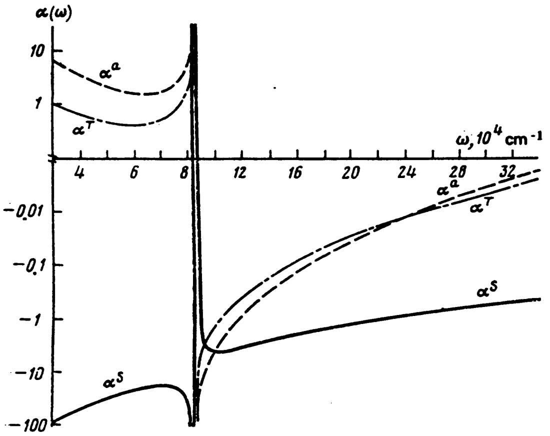

FIG. 1. Frequency dependences of the polarizabilities α^S, α^a, and α^T of the $2P$ level of the hydrogen atom.

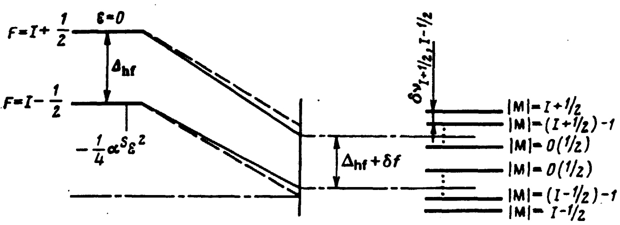

FIG. 2. Qualitative splitting of the hfs doublet $F=I\pm 1/2$ in a linearly polarized field.

We shall now consider separately the cases of linear and circular polarization.

A. Linearly polarized wave. It follows from Eqs. (7) and (10) that in a linear field the spectrum can be represented qualitatively as shown in Fig. 2. Since $|\beta_n^T|\ll|\beta_n^S|$, the change in the frequency of a hyperfine transition in the field is

$$\delta f_{F=I+1/2,F'=I-1/2}\approx-\mu_I\varkappa\frac{2I+1}{4I}\beta_n^S(\omega)\mathcal{E}^2. \tag{11}$$

(If δf is measured in hertz, $\mathcal{E}$ in volts per centimeter, and μ_I in nuclear magnetons, the conversion factor on the right-hand side is $\varkappa=0.7207\times10^{-11}$ Hz·V^{-1}·cm.) For example, for the hydrogen atom (with the hyperfine transition wavelength $\lambda=21$ cm) in the field of ruby laser radiation with $\mathcal{E}=10^6$ V/cm, we have $\delta f=436$ Hz. For cesium in the same field, we find that $\delta f=10.1$ MHz, which differs considerably from the result in a static field[20]: $\delta f_{st}(\mathcal{E}=10^6\text{ V/cm})=2.25\pm0.05$ MHz.

The frequency of $\delta\nu$ of the transitions between the magnetic sublevels is governed by the value of β_n^T and it is approximately two or three orders of magnitude less than δf. In particular, for the $M=F\rightleftarrows M=F-1$ transitions involving the $F=I+\frac{1}{2}$ level, investigated experimentally in a static field,[21] we have

$$\delta\nu_{F,F-1}=\varkappa'\frac{\mu_I}{2I+1}\beta_n^T(\omega)\mathcal{E}^2, \tag{12}$$

where $\varkappa'=0.108\,106\times10^{-10}$ Hz·V^{-2}·cm^2. For example, the frequency of the $(F=4, M=4\rightleftarrows F=4, M'=3)$ transition in cesium is

$$\delta\nu_{4,3}(\mathcal{E}=10^6\text{ V/cm}\quad\omega_R)=10.9\text{ kHz}. \tag{13}$$

It should be noted that the signs of $\delta\nu$ and δf are the same at all frequencies.

B. Circularly polarized wave. In the case of circular polarization of the electromagnetic field, the results are basically different from case *A* because the level $n^2S_{1/2}$ splits into levels with $M=\pm\frac{1}{2}$ and even hfs is ignored. This splitting is given by $\alpha^a_{n,J=1/2}(\omega)$ [see Eq. (10)] and it is due to different admixtures of the virtual k^2P_J states to the wave functions of the state $n^2S_{1/2}$ with $M=\frac{1}{2}$ and $-\frac{1}{2}$. For example, for a field with the right-handed polarization ($A=1$) the selection rules show that correction of the first order in $\mathcal{E}$ to the function $\Psi(n^2S_{1/2}, M=+\frac{1}{2};\mathbf{r})$ is governed only by an admixture of virtual k^2P_J states characterized by $J=\frac{3}{2}$, whereas the correction to $\Psi(n^2S_{1/2}, M=-\frac{1}{2})$ is governed by the admixture of states with $J=\frac{1}{2}$ and $\frac{3}{2}$. We can estimate $\alpha^a_{n,J=1/2}$ simply allowing for the kP_J level nearest to $n^2S_{1/2}$. In this approximation the value of α_n^a can be expressed, by means of a simple transformation, in terms of the scalar polarizability

$$\alpha_n^a(\omega)\approx\frac{4}{3}\alpha_n^S(\omega)\frac{\omega\delta_{s0}}{\omega_{kn}^2-\omega^2}. \tag{14}$$

Here, $\delta_{s0}=E_{k3/2}-E_{k1/2}$ is the spin–orbit splitting of the k^2P_J doublet. Consequently, α_n^a can reach a high value for heavy atoms, where δ_{s0} amounts to hundreds of reciprocal centimeters (for cesium, its value is $\delta_{s0}\approx550$ cm^{-1}). The numerical values of $\alpha^a_{n,J=1/2}$ given above agree very accurately with estimates based on Eq. (14).

If allowance is made for the hfs, the shift and splitting of the $F=I\pm\frac{1}{2}$ levels are given by Eq. (8) with $\alpha^{S,a,T}$ given by the expressions in Eq. (10). Calculations indicate that practically all the atoms satisfy the inequalities

$$|\alpha^a_{n,J=1/2}|\gg\alpha^2\mu_I|\beta_n^a(\omega)|\gg\alpha^2\mu_I|\beta_n^T(\omega)|.$$

Therefore, the main effect of the action of the field reduces to the splitting of each of the hfs $F=I\pm\frac{1}{2}$ levels into $2F+1$ magnetic sublevels, the splitting being governed mainly by the value of $\alpha^a_{n,J=1/2}$. A change in the frequency of a transition between the central sublevels with $M=0$ of the upper and lower Zeeman multiplets is given by the same expression (11) as in a linearly polarized field. For sublevels of the upper and lower multiplets with $|M|=F$ the change in the frequency again depends on α_n^a:

$$\delta f_{F=I+1/2,M=F;\,F'=I-1/2,M'=F'}=-\frac{\mathcal{E}^2}{4}\left\{\alpha^2\mu_I\frac{2I+1}{I}\beta_n^S-A\frac{2I}{2I+1}\alpha_n^a\right\}.$$

The magnetic sublevels are equidistant in each of the $\{F, M\}$ multiplets. Therefore, the action of a circularly polarized field is equivalent to a weak magnetic field and the effective Landé factors of the upper and lower multiplets differ by an amount $\sim\alpha^2\mu_I\beta_n^a(\omega)$.

Level crossing occurs in fields $\mathcal{E}\gtrsim5\times10^5$ V/cm and this gives rise to the same effects as level crossing in magnetic fields.[22]

The splitting of magnetic sublevels in a circular field, governed by the vector polarizability $\alpha^a_{n,J=1/2}(\omega)$, does not contain the small parameter $\alpha^2\mu_I$ and in fields $\mathcal{E}\gtrsim10^6$ V/cm it may exceed considerably the value of Δ_{hs}. For example, in the case of cesium (see Fig. 2) the splitting of the $F=M=4$ and $F=M=3$ sublevels is ~0.7 cm^{-1} in a field of 10^6 V/cm and it is of the same order of magnitude as the splitting of excited states whose orbital momentum is $L>0$. Therefore, allowance for the splitting of the ground states in a field with a finite degree of circular polarization may be very important in studies of nonresonant perturbation of the atomic spectra in strong optical fields, particularly in experiments involving resonant multiphoton ionization.

Delone *et al.*[23] observed a dispersion dependence of the probability of the four-photon ionization of cesium by elliptically polarized radiation from a neodymium laser ($\mathcal{E}\approx10^6$V/cm;$A=0.5$, $l=0.87$) in the presence of a three-photon resonance with the 6^2F level. The observed resonance structure is interpreted as the splitting of the upper level 6^2F and the splitting of the ground state $6^2S_{1/2}$ is ignored. A calculation gives the following values of the parameters α for the 6^2F level at the neodymium laser frequency [Eqs. (8) and (9) with $F=l=3$]:

$$\alpha_{6F}^{S}=-627+i20.9;\ \alpha_{6F}^{a}=198-i38.0:\ \alpha_{6F}^{T}=66.7-i9.7.$$

Hence, it follows that the splitting of the 6^2F level is $\sim 0.4\ \text{cm}^{-1}$. The splitting of the ground state in an elliptically polarized field is given by the same formula (8) with $A<1$ as in the circular polarization case because the nondiagonal matrix element (9) is $\sim\alpha^2\mu_I\mathscr{E}^2$ and it is negligible compared with $\alpha^a_{n,J=1/2}\mathscr{E}^2$. For $\mathscr{E}=10^6$ V/cm and $A=0.5$, we have

$$E_{F=4,\,M=4}-E_{F=3,\,M=3}=0.5\ \text{cm}^{-1}.$$

It follows that in the case of the $6^2S_{1/2}$ state the splitting may be of the same order of magnitude as for the 6^2F level. Therefore, from our point of view, multipeak resonance dependence of the ionization probability found experimentally[23] has a complex structure associated with the splitting of the ground and excited resonant levels. In a field of $\mathscr{E}=10^6$ V/cm the theoretically calculated width of the resonance structure is almost four times less than the value found experimentally by the Delone *et al.*[23] and it agrees4) with the latter for $\mathscr{E}\approx 2\times 10^6$ V/cm. One of the reasons for this difference may be the errors in the experimental determination of the effective field ($\mathscr{E}_{\text{exp}}\sim 10^6$ V/cm) associated, for example, with the inhomogeneity of the distribution of the radiation in the ionization region.

4. POLARIZATION OF PHOTOELECTRONS IN RESONANT IONIZATION IN A CIRCULARLY POLARIZED FIELD

The question of the photoelectron polarization has begun to attract attention because of the feasibility of obtaining polarized electron beams by resonant multiphoton ionization of atoms (see the review of Delone and Fedorov[24]). Various mechanisms[25-28] have been suggested to account for the polarization of photoelectrons in the course of ionization of unpolarized atoms. In most cases the polarization appears in excited resonant states and one has to provide two light sources: a pump source populating the resonance levels and an "ionizing" field.

The splitting of the ground states in a circular field, discussed in the preceding section, allows us to obtain polarized photoelectrons using just one laser because then the polarization appears even in the ground state. The qualitative pattern is easily understood if we bear in mind that the split sublevels with $M=\pm\frac{1}{2}$ correspond to the electron spin projection $\pm\frac{1}{2}$ onto the direction of propagation of a wave. Since the spin projection is not affected by electric dipole transitions, it follows that tuning of the laser frequency to resonance between one of the split sublevels and some high excited atomic state in which the spin-orbit interaction is weak may ensure a high degree of polarization of photoelectrons. Obviously, ions will also be polarized in the direction parallel or opposite to the direction of polarization of photoelectrons. The degree of polarization increases on reduction of the ratio of the width Γ of the excited level to the splitting of the ground state.

In quantitative calculations of the degree of polarization we shall consider the probability $W^{(N)}_{m_s}$ of N-photon ionization of an atom with a fixed projection m_s of the photoelectron spin in the presence of an intermediate n-photon resonance ($N\geq 3, n\geq 2$). The initial state of an atom is described by the wave function $|FM\rangle$ subject to allowance for the hfs ($F=I+J$). The selection rule $\Delta M=A$ for the dipole transitions in a circular field shows that the orbital momentum of an electron in the final state is $l=N\geq 3$ and, therefore, the effects of the hyperfine and fine structure of the continuous spectrum are unimportant and the final state of an atom is described by the wave function

$$|f\rangle=\left(\frac{(2\pi)^3}{p}\right)^{1/2}\sum_{l,m=0}^{\infty} i^l\exp[-i\sigma_l(p)]Y_{lm}^*(\mathbf{p})Y_{lm}(\mathbf{r})R_{El}(r)\chi^{1/2}_{m_s}\Phi_{IM_I},$$

where $E=p^2/2$, σ_l is the partial phase of electron scattering by a residual ion, and Φ is the nuclear spinor.

If we assume an equiprobable population of the hfs sublevels of the ground state, we find that

$$W^{(N)}_{m_s}=2\pi\sum_{F=I\pm 1/2}\frac{1}{4I}\sum_{M_I,M_F}\int|A_{M_F,f}|^2\frac{d\mathbf{p}}{(2\pi)^3},\tag{15}$$

where $A_{M_F,f}$ is the amplitude of the ionization governed by the composite matrix element of order N with a resonance denominator allowing for the shift and splitting of the ground and resonance levels $|k,l=n,m=\pm n\rangle$ in the field; k is the principal quantum number; n is the resonance order; $m=+n$ $(-n)$ for the right-hand (left-hand) field polarization. The final expression for W can be written in the form5)

$$W^{(N)}_{m_s}=c_N\mathscr{E}^{2N}\sum_{F=I\pm 1/2}\frac{1}{4I}\sum_{M_I,M_F}\frac{(C^{FM_F}_{IM_I1/2m_s})^2}{\Delta^2_{FM_F}+\Gamma^2/4}.\tag{16}$$

Here,

$$\Delta_{F=I\pm 1/2,\,M_F}=\pm{}^1/_2\Delta_{ns}+\Delta_{FM_F}(\mathscr{E})=\pm{}^1/_2\Delta_{ns}+\Delta^0+\Delta E_{F=I\pm 1/2,\,M_F}-\operatorname{Re}\Delta E_{klm},$$

$$\Delta^0=(E^0_{I+1/2}+E^0_{I-1/2})/2+n\omega-E_{kl}^0$$

is the unperturbed detuning from resonance measured from the center of gravity of the hyperfine structure of the ground state; $\Delta_{FM_F}(\mathscr{E})$ is the dynamic detuning which allows for the level shift and splitting; $\Gamma=\Gamma_{sp}+2\operatorname{Im}\Delta E_{klm}$ is the sum of the spontaneous and ionization widths of a resonant level. The constant c_N is governed by the radial matrix elements and it is independent of F or M_F.

It should be noted that Eq. (16) gives also the probability $W^{(N)}_{M_I}$ of ionization for a specific polarization state of the nucleus if the summation over M_I is replaced with summation over $m_s=\pm\frac{1}{2}$. The degree of photoelectron polarization

$$P_e=\frac{W^{(N)}_{1/2}-W^{(N)}_{-1/2}}{W^{(N)}_{1/2}+W^{(N)}_{-1/2}}\tag{17}$$

is of the form

$$P_e=\frac{2}{2I+1}\left\{\sum_{M=-(I+1/2)}^{I+1/2}\frac{M}{\Delta^2_{F=I+1/2,M}+\Gamma^2/4}-\sum_{M=-(I-1/2)}^{I-1/2}\frac{M}{\Delta^2_{I-1/2,M}+\Gamma^2/4}\right\}\left\{\sum_M\frac{1}{\Delta^2_{F=I+1/2,M}+\Gamma^2/4}+\sum_M\frac{1}{\Delta^2_{I-1/2,M}+\Gamma^2/4}\right\}^{-1}$$

that defines P_e as a function of the field frequency and intensity.

In experimental investigations the cesium atom is most interesting because in this case it is possible to observe three-photon ionization in the field of a ruby

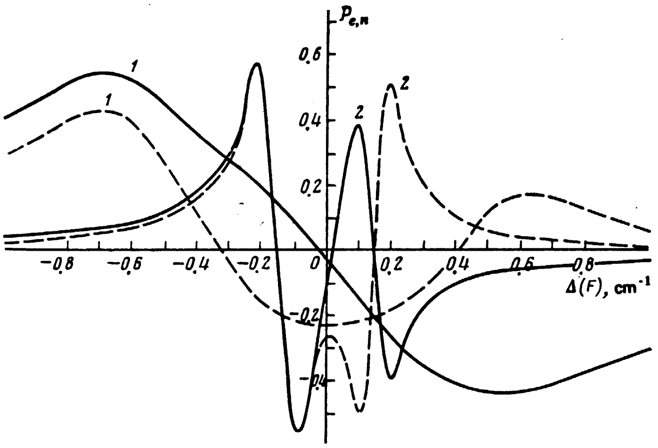

FIG. 3. Dependences of the degree of polarization of photoelectrons (continuous curves) and nuclei of ions (dashed curves) on the detuning of three-photon resonance with the 6^2F level in the case of four-photon ionization of cesium by circularly polarized radiation from a neodymium laser: 1) $\mathscr{E}=1.5\times10^6$ V/cm; 2) $\mathscr{E}=5\times10^5$ V/cm.

laser with two-photon $6^2S_{1/2}-9^2D$ resonance and four-photon ionization with three-photon $6^2S_{1/2}-6^2F$ resonance.[23] Figure 3 shows also the dependences $P(\omega)$ for electrons and nuclei obtained for various values of $\mathscr{E}$ in the case of four-photon ionization of cesium. For convenience, the ordinate gives the dynamic detuning from resonance $\Delta_F=\Delta^0-4.11\mathscr{E}^2$ (A is in reciprocal centimeters and $\mathscr{E}$ is in megavolts per centimeter). The width Γ is governed by the ionization broadening of the 6^2F state and for $\mathscr{E}=10^6$ V/cm, we have $\Gamma\approx0.09$ cm^{-1}. It is clear from Fig. 3 that in weak fields $\mathscr{E}$ the dependence $P(\omega)$ has two oscillations corresponding to resonances of the $F=I\pm\frac{1}{2}$ levels. On increase of $\mathscr{E}$, the field splitting exceeds the hfs interval Δ_{hs}, the hfs effects become unimportant, and $P(\omega)$ has only one oscillation corresponding to resonances of magnetic sublevels of the $6^2S_{1/2}$ state with $M=\pm\frac{1}{2}$. It follows from Eq. (17) that the degree of polarization is very sensitive to the width Γ. Therefore, for a resonance with a level from which one-photon ionization is impossible and whose width is governed by the spontaneous decay the value of P can be much higher than in the example considered above.

We shall conclude by pointing out that the proposed method allows us to consider also the splitting of the S_J levels with $J>\frac{1}{2}$ in a nonresonant field, for example, the metastable states 2^3S_1 of helium and 7^3S_1 of mercury. As in the case of alkali atoms, the splitting is governed by the fine structure of the excited $P_{J'}$ states except that for $J>\frac{1}{2}$ the splitting appears in a field with linear polarization because the tensor polarizability differs from zero. The tensor polarizability of the 2^3S_1 state of helium in a static electric field was determined experimentally by Player and Sandars[29]: $\alpha^T(2^3S_1, 0)=3.53\pm0.14)\cdot10^{-3}$ a.u. Our calculation of the polarizability at the neodymium laser frequency gave $\alpha^a(2^3S_1, \omega_N)=-12.1$, $\alpha^T(2^3S_1, \omega_N)=4.04$ a.u. Thus, the splitting of a triplet metastable level of helium in an optical field should be between three and four orders of magnitude greater than the splitting in a static field of the same intensity.

The authors are grateful to N. B. Delone for valuable discussions.

1) The secular equation for ΔE_n is obtained by a different method by Zon.[10]

2) Expansions into irreducible parts similar to Eq. (3) have been obtained earlier[8,9] for the effective Stark operator which is given by the expression in the braces on the right-hand side of Eq. (2).

3) The formula (5) is valid also for the Raman scattering in which α_p is replaced by the corresponding "Raman" parameters $\alpha_p^{\text{Ram}}(\omega)$ (Ref. 16).

4) It should be noted that Delone *et al.*,[23] used an incorrect numerical value of the parameter $v(1/6)\alpha^a_{6F}$, which resulted in a qualitative agreement between the theory and experiment.

5) If $n=2$, Eq. (16) applies if the field broadening is less than the ionization width of the level $|klm\rangle$, because otherwise we have to allow for the mixing of the resonance levels in the field.

---

[1] N. B. Delone, B. A. Zon, V. P. Kraĭnov, and V. A. Khodovoĭ, Usp. Fiz. Nauk 120, 3 (1976) [Sov. Phys. Usp. 19, 711 (1976)].

[2] V. S. Letokhov and V. P. Chebotaev (Chebotayev), Printsipy nelineĭnoĭ lazernoĭ spektroskopii, Nauka, M., 1975 (Nonlinear Laser Spectroscopy, Springer-Verlag, Berlin, 1977).

[3] P. F. Liao and J. E. Bjorkholm, Phys. Rev. Lett. 34, 1 (1975).

[4] R. T. Hawkins, W. T. Hill, F. V. Kowalski, A. L. Schawlow, and S. Svanberg, Phys. Rev. A 15, 967 (1977).

[5] R. D. Haun and J. R. Zacharias, Jr., Phys. Rev. 107, 107 (1957).

[6] N. L. Manakov, V. D. Ovsyannikov, and L. P. Rapoport, Opt. Spektrosk. 38, 424 (1975) [Opt. Spectrosc. (USSR) 38, 242 (1975)].

[7] W. Happer, Rev. Mod. Phys. 44, 169 (1972).

[8] C. Cohen-Tannoudji and J. Dupont-Roc, Phys. Rev. A 5, 968 (1972).

[9] W. Happer and B. S. Mathur, Phys. Rev. 163, 12 (1967).

[10] B. A. Zon, Opt. Spektrosk. 36, 838 (1974) [Opt. Spectrosc. (USSR) 36, 489 (1974)].

[11] V. I. Ritus, Zh. Eksp. Teor. Fiz. 51, 1544 (1966) [Sov. Phys. JETP 24, 1041 (1967)].

[12] N. L. Manakov, V. D. Ovsyannikov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. 70, 1697 (1976) [Sov. Phys. JETP 43, 885 (1976)].

[13] V. B. Berestetskiĭ, E. M. Lifshitz, and L. P. Pitaevskiĭ, Relyativistskaya kvantovaya teoriya, ch. 1, Nauka, M., 1968 (Relativistic Quantum Theory, Part I, Pergamon Press, Oxford, 1968).

[14] D. V. Varshalovich, A. N. Moskalev, V. K. Khersonskiĭ, Kvantovaya teoriya uglovogo momenta (Quantum Theory of Angular Momentum), Nauka, M., 1975).

[15] M. I. D'yakonov, Zh. Eksp. Teor. Fiz. 47, 2213 (1964) [Sov. Phys. JETP 20, 1484 (1965)].

[16] N. L. Manakov, V. D. Ovsyannikov (Ovsiannikov), and S. Kielich, Acta Phys. Pol. A 53, 581 (1978).

[17] J. R. P. Angel and P. G. H. Sandars, Proc. R. Soc. London Ser. A 305, 125 (1968); A. Khadjavi, A. Lurio, and W. Happer, Phys. Rev. 167, 128 (1968).

[18] N. L. Manakov and V. D. Ovsyannikov (Ovsiannikov), J. Phys. B 10, 569 (1977).

[19] N. L. Manakov, V. D. Ovsyannikov, and L. P. Rapoport, Opt. Spektrosk. 38, 206 (1975) [Opt. Spectrosc. (USSR) 38, 115 (1975)].

[20] J. R. Mowat, Phys. Rev. A 5, 1059 (1972).

[21] H. Gould, E. Lipworth, and M. C. Weisskopf, Phys. Rev. 188, 24 (1969).

[22] M. P. Chaĭka, Interferentsiya vyrozhdennykh atomnykh

sostoyaniĭ (Interference of Degenerate Atomic States), Izd. LGU, 1975.

[23] G. A. Delone, B. A. Zon, and K. B. Petrosyan, Pis'ma Zh. Eksp. Teor. Fiz. **22**, 519 (1975) [JETP Lett. **22**, 253 (1975)].

[24] N. B. Delone and M. V. Fedorov, Preprint No. 188, Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow (1975).

[25] P. S. Farago and D. W. Walker, J. Phys. B **6**, L280 (1973).

[26] P. Lambropoulos, J. Phys. B **7**, L33 (1974).

[27] E. H. A. Granneman, M. Klewer, and M. J. Van der Wiel, J. Phys. B **9**, 2819 (1976).

[28] E. H. A. Granneman, M. Klewer, M. J. Van der Wiel, and G. Nienhuis, J. Phys. B **10**, 1625 (1977).

[29] M. A. Player and P. G. H. Sandars, Phys. Lett. A **30**, 475 (1969).

Translated by A. Tybulewicz

Spontaneous bremsstrahlung of an electron in the field of an intense electromagnetic wave

R. V. Karapetyan and M. V. Fedorov

P. N. Lebedev Physics Institute, USSR Academy of Sciences

(Submitted 1 March 1978)
Zh. Éksp. Teor. Fiz. **75**, 816–826 (September 1978)

The spectral intensity and total power are found for the spontaneous bremsstrahlung of an electron in scattering by ions in the field of an intense electromagnetic wave (of frequency ω_0). A resonance structure of the radiation is observed at frequencies close to $n\omega_0$, where $n = 1,2,3,\ldots.$ The width of the resonance curve, the locations of the peaks, the number of resonance peaks, and the values of spectral intensity at the peaks are determined. The maximum number of quanta of the intense field which can be absorbed in spontaneous bremsstrahlung is found. It is shown that the maximum energy of the spontaneously radiated photon is determined by the energy of the oscillations of the electron in the field of the wave. The nonresonance part of the spectral radiation intensity, which falls off with increase of the pumping wave intensity E_0 as E_0^{-1}, is determined.

PACS numbers: 41.70. + t

1. INTRODUCTION

In recent years definite attention in the literature has been devoted to the theoretical study of induced bremsstrahlung of electrons in scattering by ions in the presence of an intense electromagnetic wave[1-3] (see also the references cited in our earlier article[2]). The interest in phenomena of this type is apparently due primarily to the development of the physics of laser plasmas. The theory of induced bremsstrahlung has treated both induced multiphoton radiation and absorption at the frequency of the intense wave and induced bremsstrahlung at other frequencies $\omega \neq \omega_0$ which arises on passage of a second, weak (probing) wave. It should be noted that the strong field of the pumping wave affects not only induced bremsstrahlung processes, but also the spontaneous bremsstrahlung of the electron, which may present independent interest. Borisov and Zhukovskiĭ[4] discuss the effect of an external electromagnetic wave on the spontaneous bremsstrahlung of ultrarelativistic electrons. With regard to a laser plasma, however, greater interest is presented by the case of nonrelativistic electrons, which is considered in the present work.

In discussing the pumping wave, we have in mind those values of field strength E_0 at which the velocity of the electron's oscillations in the wave v_E exceeds the velocity of its translational (or thermal) motion v:

$$v_E = eE_0/m\omega_0 \gg v. \tag{1}$$

Plasma states of this type are nonequilibrium and can exist only for a finite length of time less than the time of heating of the electrons to a temperature corresponding the velocity v_E. The processes of induced and spontaneous bremsstrahlung in this case are multiphoton processes in the field of a strong pumping wave. For example, in spontaneous radiation the electron can absorb or radiate n quanta $\hbar\omega_0$ and radiate one proton $\hbar\omega$, where n is an arbitrary integer. In view of these circumstances there is no direct and known beforehand relation between the induced and spontaneous bremsstrahlung processes such as exists between the Einstein coefficients in the absence of a pumping wave. Therefore in spite of the fact that the solution of the problem of induced bremsstrahlung at arbitrary frequencies ω for any values of E_0 is known,[2,3] the study of spontaneous bremsstrahlung for $v_E \gg v$ is an independent problem, to solution of which the present article is devoted.

0038-5646/78/090412-07$02.40