

# Polarization of photoelectrons in the ionization of unpolarized atoms

N. B. Delone and M. V. Fedorov

*P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow*  
Usp. Fiz. Nauk **127**, 651–681 (April 1979)

The physical effects which result in the polarization of the electrons produced in the ionization of unpolarized atoms by an electromagnetic field in the optical frequency range are discussed. Attention is focused on the Fano effect and resonance many-photon ionization. Basic equations for the angular and polarization properties of the photoelectrons are derived. A corresponding experiment is described, and the results are compared with the theoretical predictions. The outlook for the use of these processes to develop sources of polarized electrons is discussed.

PACS numbers: 32.80.Fb, 32.80.Kf

## CONTENTS

1. Introduction .....	252
2. Photoionization .....	253
a. Polarization and angular distributions of the photoelectrons .....	254
b. The Fano effect .....	255
c. Experimental procedure .....	257
d. Comparison of experimental and theoretical results .....	258
e. Fano effect induced by an intense external electromagnetic field .....	259
3. Resonance many-photon ionization .....	260
a. Ionization in a resonance with a fine-structure state .....	260
b. Ionization in a resonance with a hyperfine-structure state .....	262
c. Resonance ionization in a strong external field .....	264
d. Experimental procedure .....	264
e. Experimental results .....	265
f. Photoelectron polarization in the ionization of atoms with a selective population of fine-structure sublevels .....	267
4. Conclusion .....	267
5. References .....	268

## 1. INTRODUCTION

A polarized electron is one with a predominant spin orientation. Polarized photoelectrons can obviously be produced by ionizing polarized atoms, i.e., atoms in which the average projection of the electron spin onto some direction is not zero. In the present review we will analyze a different case: the production of polarized photoelectrons in the ionization of unpolarized atoms, i.e., atoms in which the average projection of the electron spin is zero.

Polarized electrons can of course be produced in the elastic scattering of unpolarized electrons by an unpolarized atomic target.<sup>1</sup> This effect, usually called "Mott scattering," results from the spin-orbit interaction in an electronic bound state in the atom. Mott scattering has been studied in detail<sup>2–5</sup> and is the principle underlying the well-known method for measuring the polarization of scattered electrons. A Mott detector operates by virtue of an asymmetry in the scattering (in the azimuthal direction) of electrons with spin perpendicular to the scattering plane. The very existence of this effect implies that the differential scattering cross section varies with the spin direction of

the incident electron.

Mott scattering experiment is an example of experiments in which the spin state of the electron is detected after a collision with an atom. The scattering of polarized electrons by atoms is of increasing importance in the physics of electron-atom collisions.<sup>5–7</sup> One reason for this interest is that experimental data on the scattering of unpolarized spinor particles by a target are incapable in principle of furnishing the entire scattering matrix. The elastic scattering matrix for an electron (or any other spin-1/2 particle) has three independent values. They can be, for example, the moduli of the probability amplitudes for finding the particle in states with definite spin projections on some direction and the relative phase of these states. In experiments in which only the final spin state of the electron is detected, only two of the three independent values of the scattering matrix can be measured. If any measurement is to be judged complete, it must include a detection of the spin state of the electron before the scattering; in other words, the experiment must involve the scattering of polarized electrons by atoms. Here there is an obvious analogy with the scattering of nucleons by nuclei.<sup>8</sup>

The practical use of polarized electron beams is not restricted to the problem of finding the complete scattering matrix for electron-atom collisions. As another example we can cite the promising outlook for the use of polarized-electron scattering to study the exchange interaction in atoms and in magnetic materials.<sup>5</sup> Magnetic materials seem to be among the most interesting subjects for experiments with polarized electron beams.<sup>9</sup> Slow electrons (in contrast with neutrons) do not penetrate deep into a solid because of the large Coulomb cross section. The scattering of low-energy polarized electrons is thus convenient for studying the crystal structure of thin films and surfaces.<sup>5</sup> This method thus supplements the use of polarized-neutron scattering for structural studies.<sup>8</sup> Fast (accelerated) electrons can be convenient for research in nuclear physics.<sup>5</sup> Finally, polarized electrons raise the possibility of selective impact excitation of atomic levels.<sup>6</sup>

This brief list of course does not exhaust the number of possible applications of polarized electrons, but it does clearly demonstrate the motivation for developing and optimizing methods for producing polarized electron beams.

Several approaches have been identified for producing such beams, making use of different physical principles. Some of these approaches have been pursued in practice and have led to working sources of polarized electrons.<sup>10</sup>

These methods can essentially be classed into three groups. The first group consists of Mott scattering.<sup>1-4</sup> The second consists of a variety of effects which arise in the emission of electrons from bound but previously oriented states. Included here are the photoionization<sup>11</sup> and Penning ionization<sup>12,13</sup> of polarized atoms, photoemission from semiconductors in which the electrons are oriented by circularly polarized light,<sup>14</sup> and emission from ferromagnets, i.e., from media with a fixed magnetization vector.<sup>9,15,16</sup> The third group consists of processes involving the photoionization of unpolarized atoms.<sup>17,18</sup> As basic measures of the usefulness of the various methods as sources of polarized electrons we can adopt two characteristics: the degree of polarization  $P$  (the relative number of electrons with a given spin direction) and the current  $I$  (or the number of polarized electrons in the pulse in the case of a pulsed method). The better methods of the first two of our three groups produce degrees of polarization up to  $P \approx 0.8-0.9$  at a current of  $I \approx 10^{-4}$  A (in a pulse 1  $\mu$ sec long) in the photoionization of polarized atoms<sup>11</sup> and at a current of  $I \approx 10^{-6}$  A in emission from ferromagnets.<sup>9</sup> We note that in the methods of the first two groups there is no apparent way to achieve complete polarization ( $P = 1$ ).

In this review we will be discussing two related methods which fall in the third group: photoionization (one-photon ionization) and resonance many-photon ionization by circularly polarized light. In the photoionization case we restrict the analysis to the polarization of the electrons in the case of the Fano effect, which has been predicted theoretically<sup>19</sup> and studied experimentally<sup>17</sup> for alkali metal atoms. The essential

physics of the Fano effect is the particular way in which the photoionization cross section varies with the electron kinetic energy for these atoms; specifically, there is a deep minimum near the continuum boundary. Near this minimum the spin-orbit interaction plays a role comparable to (or more important than) that of the Coulomb interaction, causing a substantial polarization of the photoelectrons. We will also discuss the Fano effect induced by an external field<sup>20</sup> as a promising extension of the method to a large number of atoms in a broad energy range. The basic physics of resonance ionization<sup>21</sup> as a method for producing polarized electrons is also directly related to the spin-orbit interaction. In this case, however, in contrast with the Fano effect, it is the effect of this interaction on the atomic spectrum, rather than on the magnitude of the cross section, which is of decisive importance. Because of the particular resonance conditions and selection rules, the intermediate excited states are populated nonuniformly in resonance ionization in a circularly polarized field; specifically, certain values of the electron angular momentum and the magnetic quantum number are predominant.

The atom in the intermediate state is thus polarized. Its subsequent ionization results in polarized electrons.

For both these methods (direct and resonance ionization), it is possible in principle to produce fully polarized electrons. These methods are in different stages of theoretical and experimental study at the present time; the Fano method has been studied quite thoroughly, but this is not the case with resonance ionization. On the basis of the experimental data currently available we can assert that, in terms of the electron current, both these methods can compete with the better methods of the first two groups. It is interesting to note that resonance ionization can also produce polarized nuclei,<sup>22</sup> through excitation of states of the spin hyperfine structure.

There is yet another method for producing polarized electrons in the ionization of unpolarized atoms, based on the excitation of autoionization states.<sup>5</sup> Formally, this method could be classified as a generalization of the Fano effect to complex atoms or as an analog of resonance ionization (if the autoionization state is assumed analogous to an excited intermediate level). However, the physics of this effect is completely different. In contrast with photoionization and resonance ionization, the excitation of an autoionization state and its subsequent ionization (the Auger effect) definitely do not constitute a one-electron transition. In this review we will thus not discuss in detail the method for producing polarized electrons through the excitation of autoionization states (see Ref. 5). We will restrict the analysis to one-electron transitions, primarily for monovalent atoms or ions.

## 2. PHOTOIONIZATION

The bound states of an electron in an atom are described by the principal quantum number  $n$  and the quantum numbers specifying the orbital angular momentum,  $l$ , the total angular momentum,  $j = l \pm 1/2$  ( $j > 0$ ), and

its projection onto the  $z$  axis,  $m_j$ . The electronic polarization of atoms is due to the orientation of the electron spin  $S$ : The atoms are polarized if the average projection of the electron spin onto some direction is not zero, or they are unpolarized if this projection is zero. The degree of polarization along the  $z$  axis is defined as the expectation value of the spin-projection operator  $S_z$ , divided by its maximum possible value:  $P_{at} = \langle S_z \rangle / S$ . For monovalent atoms with a distribution function which is diagonal in the  $|l j m_j\rangle$  representation, this definition gives us a polarization

$$P_{at} = \frac{\sum_{m_j} n_{m_j} \langle 2S_z | j m_j \rangle}{\sum_{m_j} n_{m_j}}, \quad (1)$$

where  $n_{m_j} / \sum n_{m_j}$  is the relative number of atoms with quantum numbers  $n, l, j, m_j$ . With  $l = 0$  (the  $S$  state),  $j = 1/2$ , and  $m_j = \pm 1/2$ , the polarization  $P_{at}$  takes its simplest form:

$$P_{at} = \frac{n_{1/2} - n_{-1/2}}{n_{1/2} + n_{-1/2}}. \quad (2)$$

If the atoms subjected to the light are unpolarized, we may ask whether the photoelectrons will have any polarization which results from the ionization process. In two independent cases, the answer to this question is negative.

1. The first case is the ionization of atoms by a linearly polarized field. No additional polarization can arise in the course of the ionization, since the selection rule  $\Delta m_j = 0$  (the  $z$  axis is parallel to the polarization) means that the  $m_j$  distribution of the electrons in the continuum is the same as that in the bound state. Then with  $P_{at} = 0$  the polarization of the photoelectrons found from an equation like (1) is also zero.

2. The second case is an arbitrary polarization of the ionizing radiation without a spin-orbit interaction in the atom. Again in this case, the photoelectrons do not acquire a polarization, since the interaction with the electromagnetic field, which is independent of the spin variables, does not alter the initial distribution in terms of the projection of the electron spin onto some specified direction.

In principle, therefore, the electrons can acquire a polarization in the course of ionization only by virtue of the circularly polarized field component when there is a spin-orbit interaction in the atom.

### (a) Polarization and angular distributions of the photoelectrons

Let us examine the polarization and angular distributions of the photoelectrons produced through the ionization of unpolarized  $S$ -state atoms by a circularly polarized electromagnetic wave (for definiteness, let the wave be right-hand polarized). As the one-electron wave functions of the continuum we must use functions which have the following asymptotic behavior: a plane wave  $\exp(ipr/\hbar)$  + an incoming spherical wave  $(1/4) \exp(ipr/\hbar)$  (Ref. 23):

$$\psi_{p\mu} = (2m^2 E)^{-1/4} \sum_{j m_j} i^l C_{l, m_j - \mu; 1/2, \mu}^{j m_j} Y_{l, m_j - \mu}^* \left( \frac{\mathbf{p}}{|\mathbf{p}|} \right) R_{E l j}(r) |j m_j\rangle, \quad (3)$$

where

$$j = \frac{1}{2}; \frac{3}{2}, \dots; l = j \pm \frac{1}{2}; -j \leq m_j \leq j; \mu = \pm \frac{1}{2},$$

the  $C$ 's are the Clebsch-Gordan coefficients, the radial wave functions  $R_{E l j}$  are normalized to a  $\delta$ -function of the electron kinetic energy  $E = p^2/2m$ ,  $|j m_j\rangle$  is the spin-angular wave function of an electron with a total angular momentum  $j$  and a  $z$  projection  $m_j$ , the  $Y_{lm}$  are the spherical harmonics,  $\mathbf{p}$  is the electron momentum, and  $\mu$  is the  $z$  projection of the electron spin at a large distance from the atom.

The selection rules<sup>24</sup> tell us which states are excited in the course of the ionization, i.e., which terms in the sum in (3) actually contribute to the matrix element for the transition to the continuum. Figure 1 shows the transitions between the  $S$  and  $P$  states of an atom in a right-hand circularly polarized wave, in which case the selection rules are  $\Delta l = \Delta m_j = 1$ .

The spatial part of the interaction operator, which couples the wave functions  $\psi_{S\mu} = (4\pi)^{-1/2} R_0(r) u_\mu$  and  $\psi_{P\mu}$  in the case of a right-hand circularly polarized electromagnetic wave, is  $V = (1/2) e r \mathcal{E}_0 \sin\theta' e^{i\varphi'}$  where  $\mathcal{E}_0$  is the electric field amplitude of the wave, and  $\theta'$  and  $\varphi'$  are the spherical angles specifying the direction of the vector  $\mathbf{r}$ .

The transition probability matrix

$$\frac{dw_{\mu\mu_s}}{d\Omega} = \frac{2\pi}{\hbar} \sqrt{2m^3 E} |\langle \psi_{P\mu} | V | \psi_{S\mu_s} \rangle|^2 \quad (4)$$

can be written in the following form after the matrix elements are calculated:

	$\mu$	1/2	-1/2
$\mu_0$			
$\frac{\hbar}{e^2 \mathcal{E}_0^2} \frac{dw_{\mu\mu_s}}{d\Omega}$	1/2	$(1/8) R_3^2 \sin^2 \theta$	0
	-1/2	$(1/18) (R_1 - R_3)^2 \cos^2 \theta$	$[(2R_1 + R_3)^2 / 72] \sin^2 \theta$

(5)

Here  $d\Omega = \sin\theta d\theta d\varphi$ ,  $\theta$  and  $\varphi$  are the spherical angles specifying the direction of the photoelectron momentum  $\mathbf{p}$ , and  $R_{1,3}$  are the radial matrix elements of the transitions  $S \rightarrow P_{1/2}$  and  $S \rightarrow P_{3/2}$ .

For unpolarized atoms the average ionization probability with a definite value ( $\mu$ ) of the  $z$  projection of

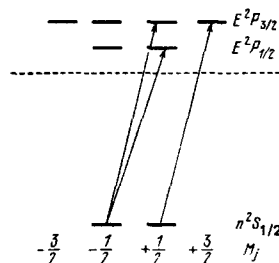


FIG. 1. The Fano effect. Transition scheme for right-hand polarized light. The dashed line is the continuum boundary.

the photoelectron spin is

$$\frac{dw_{\mu}}{d\Omega} = \frac{1}{2} \sum_{\mu_0} \frac{dw_{\mu\mu_0}}{d\Omega}. \quad (6)$$

The polarization along the  $z$  axis of the electrons which are moving at an angle  $\theta$  from this axis is defined as the expectation value of the operator  $2S_z$ , divided by the number of photoelectrons which are moving in this direction; it can be written

$$P(\theta, x) = \frac{(dw_{1/2}/d\Omega) - (dw_{-1/2}/d\Omega)}{\sum_{\mu} dw_{\mu}/d\Omega} = 2 \frac{\cos^2 \theta + x \sin^2 \theta}{1 + \cos^2 \theta + x^2 \sin^2 \theta}, \quad (7)$$

where  $x$  is the dimensionless parameter

$$x = \frac{2R_3 + R_1}{R_3 - R_1}, \quad (8)$$

whose magnitude and range will be discussed below.

The integral polarization of the electrons can be defined as twice the expectation value of the  $z$  projection of the electron spin,  $\langle 2S_z \rangle$ , integrated over all directions and divided by the total number of photoelectrons:

$$P = \frac{w_{1/2} - w_{-1/2}}{\sum_{\mu} w_{\mu}} = \frac{1 + 2x}{2 + x^2}, \quad (9)$$

where  $w_{\mu}$  is the total probability for photoionization with a definite value ( $\mu$ ) of the spin projection  $S_z$ .

We can also define the photoelectron polarization in the other directions ( $x$  and  $y$ ). The corresponding values,  $P_{x,y}$ , can be defined quite analogously as twice the expectation values of the spin projection operators,  $\langle 2S_{x,y} \rangle$ , divided by the total number of photoelectrons moving in the given direction. It is convenient to express  $P_{x,y}$  in terms of the quantities  $P_{\pm} = P_x \pm iP_y$ , which are calculated by analogy with the derivation of Eq. (10) for the polarization  $P_z(\theta, \varphi, x)$ ; the result is

$$P_{\pm}(\theta, \varphi, x) = 2(1-x) \frac{\cos \theta \sin \theta}{1 + \cos^2 \theta + x^2 \sin^2 \theta} e^{\pm i\varphi}. \quad (10)$$

In contrast with  $P$ , the polarizations along the directions perpendicular to the wave vector depend on the azimuthal angle  $\varphi$ . In the averaging over  $\theta$  or  $\varphi$ , the quantities  $P_{\pm}$  vanish; in other words, the spin vector can be oriented on the average (over the directions of the electrons) only along (or opposite) the direction of the light wave vector.

The total ionization cross section found with the help of transition matrix (5) is

$$\sigma = \int d\Omega \frac{d\sigma}{d\Omega} = \frac{4\pi\alpha\hbar\omega}{27} (\Delta R)^2 (2 + x^2), \quad (11)$$

where  $\Delta R = R_3 - R_1$ , and  $\alpha = e^2/\hbar c = 1/137$  is the fine structure constant.

If the spin-orbit interaction is neglected [with  $R_1 \approx R_3 = R(E)$ ], Eqs. (5) and (11) lead to the familiar expressions<sup>25-27</sup> for the differential and total ionization cross sections:

$$\frac{d\sigma}{d\Omega} = \frac{\pi\alpha\hbar\omega}{2} R^2(E) \sin^2 \theta, \quad \sigma = \frac{4\pi}{3} \alpha\hbar\omega R^2(E). \quad (12)$$

The first of these equations states that the directional pattern of the photoelectrons emitted by a circularly polarized field has a maximum in the plane perpendicular to the wave vector ( $\theta = \pi/2$ ).

Equations (5)-(7) completely determine the polarization and directional pattern of the photoelectrons for ionization from the  $S$  state of the atom when there is a spin-orbit interaction. Cherepkov<sup>28</sup> has studied the general case of ionization from states with an arbitrary orbital angular momentum  $l > 0$ .

## (b) The Fano effect

Various approaches<sup>29-31</sup> can be taken to calculate the radial wave functions and matrix elements which determine the photoionization cross sections and the polarization properties for real atoms. Without going into the details of the calculations (which are discussed in Refs. 25-27), we will examine the important question as to how the matrix elements vary with the photoelectron kinetic energy  $E$ . This behavior has been studied elsewhere, on the basis of both quantitative calculations and qualitative considerations.<sup>31-33</sup>

If we specify the sign of the radial wave functions by requiring that they be positive in the limit  $r \rightarrow 0$ , then, according to Fano and Cooper,<sup>33</sup> the matrix element  $R(nl, El')$  is positive at large  $E$  and falls off in proportion to  $E(-l+7/2)$ . At small  $E$  the sign of the matrix element  $R$  and its variation with  $E$  are very sensitive to the electronic configuration of the atom. The situation can be illustrated qualitatively for the inert gases neon and argon. Figure 2 shows the wave function of an electron in the outer  $p$  shell of these atoms, along with the  $d$  wave function of the continuum with an energy  $E=0$ . It is clear from an analysis of the regions of maximum overlap of these wave functions that for neon  $R$  is positive at  $E=0$  but negative for argon. With increasing energy, the oscillations of the continuum wave function become faster (the wave function gets "squeezed" against the origin). As a result, the matrix element  $R$  for neon increases slightly and then decreases, approaching an asymptotic value at large  $E$ . In contrast, the argon matrix element initially decreases in magnitude, vanishing at a certain  $E$ ; then it becomes positive, goes through a maximum, and only then approaches its high-energy asymptote (Fig. 3). This behavior of the matrix element as a function of the photo-

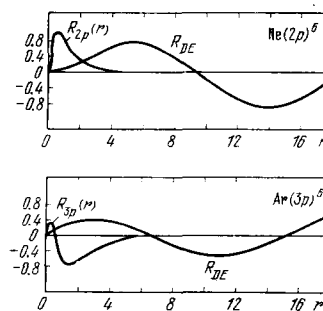


FIG. 2. Wave functions of the bound  $P$  and  $D$  states of the continuum with an energy  $E=0$ .

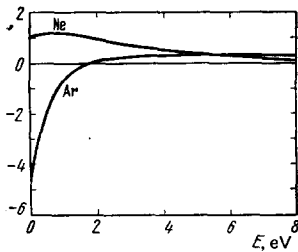


FIG. 3. Matrix elements for the transitions from the ground state to the continuum as a function of the photoelectron energy in argon and neon atoms.

electron energy is typical of many atoms and ions. According to the rules stated by Fano and Cooper,<sup>33</sup> there is an interval of  $E$  in which the matrix element is negative if (a)  $\Delta l = 1$  and (b)  $l < l_{\max} = n - 1$ . Condition a means that  $R$  can vanish only for transitions accompanied by an increase in the orbital angular momentum  $l$ . Over a broad range of  $E$  it is this photoionization pathway in governing.<sup>28-33</sup> From condition b we see that the bound-state wave function must also have antinodes. Conditions a and b tell us only whether an interval of negative  $R(E)$  can exist; they do not tell us where it is, and in fact it can be in different places for different atoms. In argon and krypton, we find  $R(E) = 0$  at  $E \sim 2$  Ry, while for the alkali elements we find  $R(E) = 0$  near the ionization threshold ( $E \sim 1$  eV). In the lithium atom the negative values of  $R(E)$  in fact lie below the ionization threshold.

In the case  $S - P$ , the vanishing of the matrix element  $R(E)$  when the spin-orbit interaction is neglected leads to a vanishing photoionization cross section [Eq. (12)]. Actually, however,  $\sigma(E)$  does not vanish, since the matrix elements  $R_1$  and  $R_3$ , which determine  $\sigma$  [Eqs. (8) and (11)], vanish at slightly different energies  $E$ . This difference, which is due to the spin-orbit interaction, is small, so that the ionization cross section has a deep minimum at these energies (Fig. 4).

The quantity  $\Delta R = R_3 - R_1$ , which determines the absolute value of the ionization cross section in (11), is non-zero because of the spin-orbit interaction, and it varies comparatively slowly (the characteristic energy interval of the variation is of the order of a rydberg). Near the  $\sigma(E)$  minimum we can thus assume  $\Delta R \approx \text{const}$ . In this case Eq. (11) describes a parabolic behavior of the cross section as a function of the energy  $E - E_0 \sim x$  near

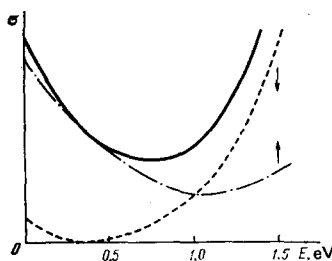


FIG. 4. Photoionization cross section as a function of the photoelectron energy. Solid curve—total cross section; dashed and dot-dashed curves—cross sections for the production of photoelectrons with opposite spin orientations.

the minimum, which is reached at  $x(E_0) = 0$  or  $R_1 + 2R_3 = 0$ .

This deep minimum in the ionization cross section is responsible for several interesting and unusual polarization properties of the photoelectrons (the Fano effect<sup>19,34</sup>). Generally speaking, the spin-orbit interaction in an atom is small in comparison with the basic Coulomb-interaction energy. Thus the polarization of the electrons due to this interaction is also small under ordinary conditions. It is not small, however, if the photoionization probability becomes anomalously low because of interference effects, as it does near the  $\sigma(E)$  minimum. In this case the photoelectron polarization can be high and can even reach 100%.

Figure 5 shows the polarization as a function of  $x$  and thus as a function of  $E$  (the parameter  $x$  usually decreases with increasing  $E$ ). The maximum value,  $P_{\max} = 1$ , is reached at  $x = 1$ . Interestingly, the polarization reaches its maximum at  $x = 1$  regardless of the direction in which the electron is moving, so that  $P_{\max}(\theta) = P_{\max} = 1$ . The minimum of  $P(\theta, \varphi, x)$ , in contrast, does depend on  $\theta$  and is reached at values of  $x$  which do depend on the direction in which the electrons are moving:

$$P_{\min}(\theta, \varphi, x) = P(\theta, \varphi, x) = -\frac{1 + \cos^2 \theta}{\sin^2 \theta} = -\frac{\sin^2 \theta}{1 + \cos^2 \theta}. \quad (13)$$

We obviously have  $P_{\min}(\theta, \varphi, x) = -1$  at  $\theta = \pi/2$ ; i.e., those photoelectrons which are moving in the plane perpendicular to the wave vector of the electromagnetic wave,  $\mathbf{k}$ , are completely polarized in the  $-\mathbf{k}$  direction at  $x = -1$ . The minimum value of the integral polarization  $P$  is  $-1/2$  and is reached at  $x = -2$ .

We thus conclude that the photoelectrons can be highly polarized in certain atoms, because of the interference compensation of the photoionization cross section. The most favorable photoelectron energies here are those which correspond to a unit value of the parameter  $x$ , since in this case the polarization can be nearly 100%, regardless of the observation direction and the angular aperture of the detector. The most convenient objects for observing and making practical use of the Fano effect are the alkali metal atoms. Since the cross section has a minimum near the threshold, the minimum is deep and clearly defined, not masked by other effects such as other transitions and autoionization peaks in the absorption coefficient.<sup>33</sup>

Equations (9) and (11) for the integral polarization  $P(x)$  and ionization cross section can be generalized to the case of arbitrarily polarized atoms and a radiation

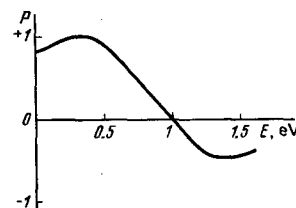


FIG. 5. Degree of photoelectron polarization as a function of the photoelectron energy.

field with an arbitrary degree of circular polarization<sup>35</sup>  $P_f$ :

$$P_{el} = \frac{(2x+1)P_f + x^2 P_{at}}{x^2 + 2 + (2x-1)P_f P_{at}}, \quad (14)$$

$$\sigma = \frac{4\pi\alpha\hbar\omega}{27} (\Delta R)^2 [2 + x^2 + (2x-1)P_f P_{at}]. \quad (15)$$

In addition to the parameter  $P = (P_{el}/P_f)|_{P_{at}=0}$ , defined in (9), we can introduce several other polarization parameters when they are more convenient for analyzing experimental data. We denote by  $\sigma_+$  the photoionization cross section at some particular value of the product  $P_f P_{at}$  and by  $\sigma_-$  the same cross section but corresponding to the opposite sign of this product (but for the same  $|P_f P_{at}|$ ). Then the parameter

$$Q = \frac{\sigma_+ - \sigma_-}{P_f P_{at}(\sigma_+ + \sigma_-)} = \frac{2x-1}{x^2+2} \quad (16)$$

is obviously independent of the particular values of  $P_f$  and  $P_{at}$ .

Since the polarization parameters  $P(x)$  and  $Q(x)$  depend on only the single variable  $x(E)$ , two independent experiments can be designed. The quantity  $Q$  can be found as the function of the photoelectron kinetic energy in experiments in which the total ionization cross sections  $\sigma_+$  are measured for various polarizations of the atoms,  $P_{at}$ , and the field,  $P_f$ . The results can be used to determine the function  $x(E)$ . Knowing  $x(E)$ , we can use (13) to construct the  $E$  dependence of the electron polarization parameter  $P(x) = P[x(E)]$ , which can be checked experimentally,<sup>35,36</sup> through direct measurements of the photoelectron polarization.

### (c) Experimental procedure

An experiment to observe the Fano effect involves subjecting alkali metal atoms to ionizing radiation in the UV range, extracting the electrons produced, and analyzing their polarization.

The number of photoelectrons produced,  $N_e$ , is obviously governed by  $N_e = n_a l \sigma N_f$ , where  $n_a$  is the density of neutral atoms,  $\sigma$  is the cross section for the interaction of the radiation with the atoms,  $l$  is the length of the interaction volume, and  $N_f$  is the total number of photons. Since the electron polarization depends on the frequency of the radiation, the optimum frequency and the corresponding photoionization cross section are given quantities. Then the only way the electron yield can be increased is to increase the target density, the length of the interaction volume, and the radiation intensity.

The need to use UV light is a substantial complication. Both high-pressure inert-gas lamps and lasers have been used as UV sources. The laser is the better choice when the photoelectron yield must be maximized. Lasers of various designs currently available can produce the necessary UV intensities at certain frequencies.<sup>17,36,37</sup> Although high-pressure lamps cannot furnish radiation with a spectral brightness as high as that from lasers, the lamps have the advantage for research purposes that, when combined with a monochromator, they represent a simple means of producing

UV radiation over a broad frequency range with tunable frequency.

The targets can be either atomic vapors<sup>36</sup> or atomic beams.<sup>17</sup> With vapors it is simple to arrange a long interaction volume, but there are the usual difficulties resulting from contamination of the working medium by various parts of the apparatus. If the light beam strikes an atomic beam at an angle of 90° these difficulties are avoided, but it is difficult to arrange a long interaction volume: Even if a large-aperture multichannel collimator is used to form the beam, the beam diameter is still no more than a few millimeters. In some experiments, several parallel atomic beams have in fact been used to increase the interaction length.

From data on the various characteristics of the experiment it is simple to estimate the efficiency of the electron production from the equation given above. In particular, if we assume that the photoionization cross section is of order  $\sigma \sim 10^{19}$  cm<sup>2</sup>, that the density of atoms is  $n_a \sim 10^{12}$  cm<sup>-3</sup>, and that the interaction length is  $l \sim 1$  cm, then with an energy per pulse of  $Q \sim 1$  mJ ( $\sim 10^{16}$  photons) the number of electrons is  $N_e \sim 10^{10}$ .

Only at the optimum frequency of the ionizing radiation, however, such that  $x=1$  and such that the polarization is maximized, will all the electrons produced have the same polarization. For other values of  $x$  the polarizations will be different for electrons with different emission angles  $\theta$  [Eq. (7)]. Thus only some of the electrons will have the given degree of polarization,<sup>36</sup>

The electrons extracted from the atomic target are focused and accelerated to an energy of the order of 10<sup>2</sup> keV. This high energy is necessary for measurements of the degree of polarization by the Mott-scattering method.<sup>1</sup> This method makes use of the particular nature of electron scattering by nuclei: In large-angle scattering by a heavy nucleus, an unpolarized electron acquires a certain degree of polarization in the plane perpendicular to the scattering plane. For the effect to occur, the electron energy must be  $\approx 60$  keV, the atomic number of the scatterer must be high, and the scatterer itself must not contain so many nuclei that repeated scattering occurs. The transverse polarization factor [ $N_e(\dagger)$  is the number of electrons with a given polarization-vector direction],

$$S = \frac{N_e(\dagger) - N_e(\ddagger)}{N_e(\dagger) + N_e(\ddagger)}, \quad (17)$$

is known very accurately (within  $\sim 1\%$ ) as a function of the atomic number of the scatterer, the electron energy, and the scattering angle.<sup>38,39</sup> If the incident electrons are polarized, an asymmetry appears in the number of electrons scattered through angles  $\pm \theta$ . This asymmetry can be expressed in terms of  $P$  and  $S$ :

$$\frac{N_+}{N_-} = \frac{1+PS}{1-PS}. \quad (18)$$

Knowing  $S$ , and measuring the ratio  $N_+/N_-$ , we can thus determine the degree of polarization of the electrons.

If the apparatus for extracting, accelerating, and energy-analyzing the electrons is such that the spin is

the direction in which the electrons are moving, then a Wien filter is used. This device rotates the electron spin through  $90^\circ$ , so that the electrons turn out to be polarized in the direction perpendicular to their motion.

The scatterer is usually a gold ( $Z=79$ ) foil about  $10^2$  mg/cm<sup>2</sup> thick, and the electrons are observed at about  $120^\circ$ . A detailed analysis of the accuracy of this method<sup>17,36</sup> has shown that the degree of polarization can be found within 4%-7%, depending on the particular experimental details.

Figure 6 shows a typical experimental arrangement, as described in Ref. 36. The first electron-optics system images the emission from the interaction chamber onto a limiter. The retarding-potential method is used for energy analysis of the electrons and to eliminate background electrons. The second electron-optics system focuses the electrons onto the accelerator input. The electron current is measured with a Faraday cup.

A control experiment on observation of the Fano effect involves measuring the electron polarization as a function of the polarization of the ionizing radiation. The polarization of the radiation can be changed without affecting other characteristics of the radiation by changing the orientation of a quarter-wave plate in the laser beam. The electron polarization is maximized by circularly polarized light, and it can be adjusted continuously by changing the eccentricity. It vanishes if the light is linearly polarized.

#### (d) Comparison of experimental and theoretical results

Experimental data are available for two atoms: cesium<sup>17,35,36</sup> and rubidium.<sup>36,37</sup> For a comparison with the theory, the results of the first experiment, by Baum et al.,<sup>35</sup> are the most interesting. They used a high-pressure xenon lamp and a double monochromator. In this arrangement it was possible to obtain experimental data over a broad range of light frequencies. From the practical standpoint, Granneman's results<sup>36</sup> were better. The results of the various experiments are summarized in Fig. 7, where they are compared with the calculations from Ref. 35. There is a good agreement.

It should be noted that the calculated results shown in Fig. 7 are based on an independent experiment in which the parameter  $Q$  [Eq. (16)] was measured. The measurements of the parameter  $Q$ , in Ref. 35, served

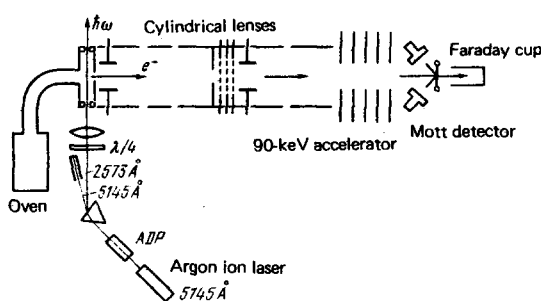


FIG. 6. Experimental arrangement of Ref. 36 to observe the polarized electrons produced in the Fano effect.

as a test of whether the spin-orbit interaction in alkali atoms was taken into account correctly. Experiments to measure  $Q$  can be carried out more accurately than experiments to measure  $P$ , because ions are detected, so that the problems of suppressing the background electrons can be avoided, and the detection of the asymmetry in the ions is extremely effective. The arrangement in this experiment was typical of research on the photoionization of polarized atoms. Intersecting light and atomic beams were used. In this case the light beam was polarized light from a xenon lamp, filtered with a monochromator, and the atomic beam was a beam of polarized atoms produced by passage through a hexapole magnet. The ions were detected with an electron multiplier. The number of ions was measured as a function of the light polarization. The direct result of the experiment was a measurement of  $Q$  as a function of the light frequency for three alkali atoms: potassium, rubidium, and cesium. These measurements were used along with (16) to find the function  $\chi(\omega)$ , which was used in turn to find the function  $P(\omega)$ , shown in Fig. 7. The good agreement between the measured and calculated values gives us confidence in the measurements of  $P$  and  $Q$ , since the parameter  $\chi$  satisfies two independent experiments.

Through the use of the Fano effect, it was possible at the time to produce  $3 \cdot 10^3$  electrons in a pulse with a degree of polarization  $90\% \pm 7\%$  (Ref. 17). The density of atoms was  $1 \cdot 10^{12}$  cm<sup>-3</sup>, the interaction length was 80 mm, the pulse repetition frequency of the laser was 3 min<sup>-1</sup>, and the energy of the light pulse was  $1.5 \cdot 10^{-3}$  J.

A detailed analysis of the experimental capabilities of modern UV sources and atomic beams shows that the atomic density and the UV energy can be increased by a factor of no more than two or three, while it would be very difficult to arrange a severalfold increase in the interaction length. Then the only significant possibility which remains is to increase the pulse repetition frequency of the laser. For example, if the second harmonic is taken from dye laser pumped by a neodymium-glass laser, it is possible to achieve a pulse repetition frequency of 50 Hz with a substantial energy

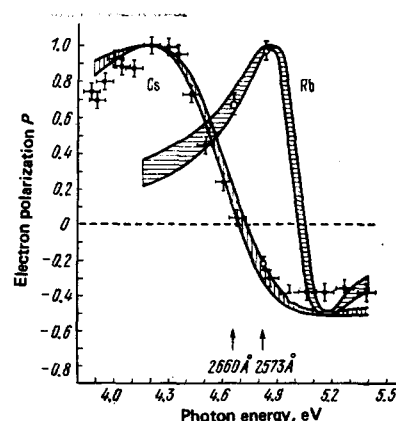


FIG. 7. Experimental data and calculated results (hatching) on the degree of polarization of the electrons produced in the Fano effect. Hatching—Ref. 35; ●—Ref. 34; □—Ref. 37; ○—Ref. 36.

per pulse. In conclusion, we should point out that the pulse repetition frequency in the irradiation of the atomic target is restricted by the construction of the apparatus in which polarized electrons are used.

### (e) Fano effect induced by an intense external electromagnetic field

It is clear from the discussion of the physical nature of the Fano effect that the effect is not universal: It occurs in by no means all atoms, and then only at certain strictly determined light frequencies. These limitations can be somewhat overcome by switching to the induced Fano effect, i.e., a modification of the effect which can be arranged by illuminating an atom with a second intense laser beam.<sup>20</sup> To explain the conditions under which this modified effect occurs, we appeal to the level scheme in Fig. 8. The field with frequency  $\omega$ , and electric field amplitude  $\mathcal{E}_1$  is strong, while the field  $\omega_0, \mathcal{E}_0$  is weak (the corresponding quantitative conditions are given below). The frequencies  $\omega_1$  and  $\omega_0$  must satisfy

$$E_1 + \hbar\omega_1 \approx E_0 + \hbar\omega_0 > 0, \quad (19)$$

where  $E_1$  is the energy of some arbitrary excited discrete level of the atom, and  $E_0$  is the ground state energy. The photon energies  $\hbar\omega_1$ , and  $\hbar\omega_0$  must exceed the thresholds for ionization from levels  $E_1$  and  $E_0$ , respectively. For simplicity we assume that the orbital angular momentum is  $l=0$  in the ground and excited states (S states), and we assume that the fields  $\mathcal{E}_1$  and  $\mathcal{E}_0$  are right-hand polarized.

This ionization process was described theoretically by Armstrong et al.<sup>40</sup> and Geller and Popov,<sup>41</sup> who ignored spin effects. The ionization probability is

$$w \sim \frac{(\Delta - a)^2}{\Delta^2 + (\Gamma/4)}, \quad (20)$$

where  $a$  and  $\Gamma$  are certain constants satisfying  $a \sim \Gamma \sim E_{at} (\mathcal{E}_1 / \mathcal{E}_{at})^2$ ,  $\mathcal{E}_{at}$  and  $E_{at}$  are the intraatomic field and the characteristic atomic energy, and  $\Delta = E_0 + \hbar\omega_0 - E_1 - \hbar\omega_1$  is the detuning from the "resonance" (Fig. 9).

When spin is taken into account, we find that a polarization of the photoelectrons is possible in this photoionization scheme, by analogy with the Fano effect, because of the deep minimum in the function  $w(\Delta)$ . In taking spin into account we must, as usual, distinguish between the radial matrix elements  $R_1^{(0,1)}$  and  $R_3^{(0,1)}$  for

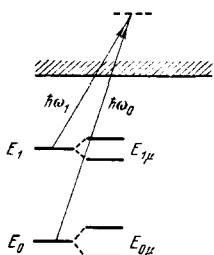


FIG. 8. Level and transition scheme for photoionization of an atom by two fields, of frequencies  $\omega_0$  and  $\omega_1$ .

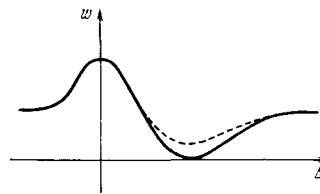


FIG. 9. The atomic ionization probability  $w$  as a function of the detuning from "resonance,"  $\Delta$ . Dashed curve—With the spin-orbit interaction and Stark level splitting.

transitions from the  $(0, 1)$  states to the continuum states  $P_{1/2}$  and  $P_{3/2}$ . Furthermore, since the field  $\mathcal{E}_1$  is assumed strong, it is necessary to take into account the splitting of the  $E_0$  and  $E_1$  levels due to the dynamic Stark effect (Fig. 8):  $E_1 \rightarrow E_{1\mu}$ ,  $E_0 \rightarrow E_{0\mu}$ ,  $\mu = \pm \frac{1}{2}$ . Although this splitting, which is governed by the vector polarizabilities of the  $E_0$  and  $E_1$  levels, is small in comparison with the average shift of these levels (governed by the scalar polarizabilities), it can still compete with the effect of the spin-orbit interaction on the size of the matrix elements. When both these factors are taken into account, the minimum value of the probability  $w$  becomes nonzero, and the following equation can be derived for the degree of polarization  $P(\Delta)$  near the minimum<sup>20</sup>:

$$P(\Delta) = \frac{A - 2(\Delta_0 + B)x}{A - (1/2)(\Delta_0 + B)^2 + 2x^2}, \quad (21)$$

where

$$x = \frac{1}{2} \sum_{\mu} (E_{0\mu} + \hbar\omega_0 - E_{1\mu} - \hbar\omega_1),$$

$$\Delta_0 = E_{1, 1/2} - E_{0, 1/2} - E_{1, -1/2} + E_{0, -1/2},$$

and  $A$  and  $B$  are small constants governed by the difference between the matrix elements  $R_1^{(0,1)}$  and  $R_3^{(0,1)}$ . The parameter  $\Delta_0$  is governed by the splitting of the  $E_0$  and  $E_1$  levels. The function  $P(x)$  (Fig. 10) is completely analogous to the corresponding function in the case of the ordinary Fano effect [Fig. 5; Eq. (9)]. The maximum value of  $P(x)$  is reached at  $x = -(\Delta_0 + B)/2$ ; as before,  $P_{\max} = 1$ . The minimum value of the probability in this case is not  $-\frac{1}{2}$ , in contrast with the ordinary Fano effect. The minimum value  $P_{\min}$  is reached at

$$x = \frac{\Delta_0 + B}{2} - \frac{A}{\Delta_0 + B},$$

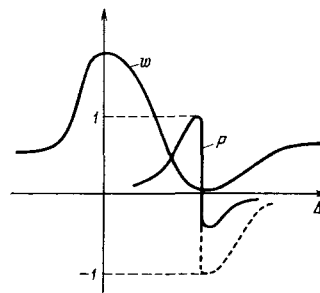


FIG. 10. The ionization probability  $w$  and the degree of polarization  $P$  as functions of  $\Delta$ . Dashed curve—Case in which the Stark splitting is more important than the spin-orbit interaction.



and is  $P_{\min} = -(\Delta_0 + B)^2 [2A + (\Delta_0 + B)^2]^{-1}$ .

The function  $P(x)$  simplifies substantially if the splitting of the  $E_0$  and  $E_1$  levels is larger than the spin-orbit effect:  $|\Delta_0| \gg \sqrt{A}$ ,  $B$ . In this case,

$$P(x) = -\Delta_0 \frac{x}{x^2 + (\Delta_0/4)}, \quad P_{\max} = P_{\min} = P\left[\left(\pm\right) \frac{\Delta_0}{2}\right] = \left(\pm\right)^4. \quad (22)$$

In this case the  $P(x)$  curve becomes antisymmetric:  $P(x) = -P(-x)$  (Fig. 10).

We can find how strong the field  $\mathcal{E}_1$  must be by noting that the characteristic interval for a substantial change in the degree of polarization  $P(x)$  must be large in comparison with the radiation level width  $\Gamma_r$ ; then with  $\Gamma_r = 10^{-3} \text{ cm}^{-1}$  and  $\Delta_0 \sim \sqrt{A} \sim B \sim \alpha E_{\text{at}} (\mathcal{E}_1 / \mathcal{E}_{\text{at}})^2$  ( $\alpha$  is the fine structure constant) we find  $\mathcal{E}_1 \sim 5 \cdot 10^6 \text{ V/cm}$ . This is not an overly severe restriction for modern lasers.

In summary, this ionization scheme with illumination by an intense field  $\mathcal{E}'$  can produce a high degree of photoelectron polarization, and it is more generally applicable. There is no restriction on the choice of level  $E_1$ , nor is there any restriction on the choice of frequencies  $\omega_0$  and  $\omega_1$  satisfying condition (19), since the resonance can occur at any energy in the continuum. The induced Fano effect can thus occur in essentially any atom over broad ranges of the frequencies  $\omega_0$  and  $\omega_1$ .

### 3. RESONANCE MANY-PHOTON IONIZATION

Many-photon resonance ionization opens up numerous opportunities for producing polarized photoelectrons.<sup>21</sup> At a resonance of the frequency of the external field with the energy of a transition to an intermediate bound state we can make the same assertions as above: In the case of unpolarized atoms, the polarization of the photoelectrons is due to the circularly polarized part of the radiation, and it is ultimately governed by the spin-orbit interaction of the atomic electrons. In the case of resonance ionization, however, the mechanism by which the spin-orbit interaction affects the polarization and angular distribution of the photoelectrons is radically different from that responsible for the Fano effect. In the case of resonance ionization, the possibility of producing polarized photoelectrons is due primarily to the spectral properties of the radiation and the atoms. When the spin-orbit interaction is taken into account, atomic levels with given quantum numbers  $n$  and  $l$  split into sublevels  $E_j$  corresponding to different values of the total electron angular momentum  $j = l \pm \frac{1}{2}$  ( $j = \frac{1}{2}$  for S levels). There is still a degeneracy with respect to the  $z$  projections  $m_j$  of the total angular momentum. If the external electromagnetic field has a narrow frequency spread and is not too intense, it is possible to excite the sublevels corresponding to a particular value  $j'$  of the angular momentum of the resonance state by adjusting the frequency of the external field. If the excitation is with a circularly polarized wave, the  $m_j'$  distribution of the atoms will not be symmetric even if there is a uniform distribution with respect to  $m_j$  in the ground state (unpolarized atoms). The nonuniformity of the  $j'$  and  $m_j'$  distributions means that the excited atoms have a certain degree of polariza-

tion. Then it is evidently possible to produce polarized photoelectrons through a transition from the resonance state to the continuum.

#### (a) Ionization in a resonance with a fine-structure state

The fine structure in the spectrum of bound electron states is known to be due to an interaction between the spin and orbital angular momentum of the atomic electron. In alkali atoms this interaction results in a doublet structure in the spectrum. The energy splitting of the doublet is extremely large: many reciprocal centimeters of the low-lying excited levels. Then there are evidently no experimental difficulties in arranging the resonance excitation of a certain fine-structure state in alkali atoms by means of laser beams, where desired. We recall that the fine-structure states have not only the quantum numbers  $n$ ,  $l$ , and  $j$  but also  $m_j$ , which determines the number of possible orientations of the electron angular momentum with respect to some particular direction.

We begin by examining the characteristic physical features of the polarization and angular distributions of photoelectrons in resonance ionization for the case of an  $S \rightarrow P \rightarrow D$  transition (Fig. 11). We assume that the external field is weak enough that we can ignore the perturbation of the atomic spectrum.

We assume that the atom is initially in the state  $|S; j = \frac{1}{2}; m_j\rangle$  ( $m_j = \pm \frac{1}{2}$ ) and that the frequency  $\omega$  is at resonance with the transition to some state  $|P; j'; m_j'\rangle$ :

$$|\Delta_{j'}| \equiv |E_{Pj'} - E_S - \hbar\omega| \ll |E_{P3/2} - E_{P1/2}|.$$

The general equation for the probability density matrix for transitions to the continuum with a certain  $z$  projection of the electron spin according to second-order perturbation theory is

$$\frac{d\rho_{\mu\mu_0}}{d\Omega} = \frac{2\pi}{\hbar} \sqrt{2m^3 E} \frac{1}{\Delta_{j'}^2 + (\Gamma/4)^2} \left| \sum_{m_j'} \langle \psi_{\mu\mu_0} | \tilde{V} | Pj'm_j' \rangle \times \langle Pj'm_j' | V | S, j = \frac{1}{2}; m_j = \mu_0 \rangle \right|^2, \quad (23)$$

where  $\Gamma$  is the natural width of the resonance level, introduced on a phenomenological basis. It is assumed to be independent of the quantum number  $m_j'$ . Here  $V$  and  $\tilde{V}$  are the spatial parts of the operators representing the interaction of the atom with the exciting and ionizing waves, for which the frequencies can in general be different ( $\omega$  and  $\tilde{\omega}$ ). If the two waves are propagating in the same direction (along  $z$ ) and if both are right-hand polarized, then  $V_{\pm} = (e\mathcal{E}_0/2)r \sin\theta e^{\pm i\varphi'}$ ,

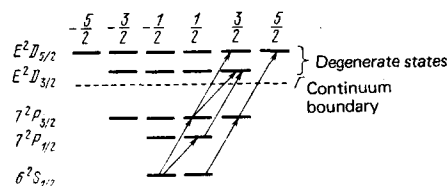


FIG. 11. Transition scheme for resonance ionization through a  $P$  state.

$\vec{V} = (e \mathcal{E}_0/2) r \sin \theta e^{i\varphi}$ . In addition,  $E = p^2/2m$  is the electron kinetic energy, which is governed by the conservation law  $E = E_S + \hbar(\omega + \bar{\omega}) \approx E + \hbar\bar{\omega}$ .

We first assume  $V \approx \vec{V} = V_+$  (a right-hand polarized monochromatic field). Then the selection rule  $\Delta m_j = 1$  singles out a single term, with  $m'_j = m_j + 1$ , in the sum over the intermediate states  $m''_j$ . If the resonance level is a  $P_{1/2}$  level, then by calculating the matrix elements in (23) we find the probability for ionization of the unpolarized atoms to be

$$\frac{dw}{d\Omega} = \frac{1}{2} \sum_{\mu\mu_s} \frac{dw_{\mu\mu_s}}{d\Omega} = \frac{e^4 \mathcal{E}_0^2 \bar{\mathcal{E}}_0^2}{24\hbar} \frac{\mathcal{R}_{PS}^2 \mathcal{R}_{DP}^2(E)}{\Delta_{1/2}^2 + (1/4)\Gamma^2} \sin^2 \theta, \quad (24)$$

where  $R_{PS}$  and  $R_{DP}$  are the radial matrix elements of the transitions  $S \rightarrow P$  and  $P \rightarrow D$  (Fig. 11). (We are ignoring the effect of the spin-orbit interaction on  $R_{PS}$  and  $R_{DP}$ .) The angular distribution of the electrons in this case is analogous to that of the photoelectrons in one-photon direct ionization, (12). We note that this result is a specific property of the resonance at the  $P_{1/2}$  level and occurs because the electron distribution in the excited state is isotropic in this case. This point is easily shown by writing out explicitly the wave function for the excited state:  $\psi = \sqrt{3/4\pi} (\sin \theta' e^{i\varphi'} u_{-1/2} + \cos \theta' u_{1/2})$ . We see immediately that  $\psi^* \psi$  is independent of the angles  $\theta'$  and  $\varphi'$ ; i.e., the distribution of excited electrons is isotropic. Then from the standpoint of the angular distribution the process of resonance ionization through an intermediate  $P_{1/2}$  level is equivalent to the photoionization (direct one-photon ionization) of an atom with an isotropic distribution of bound-state electrons. The integral degree of polarization  $P$  in this case is 60% (Ref. 42). The degree of polarization of the photoelectrons moving at an angle  $\theta$  from the wave vector is easily shown to be given by the elementary expression

$$P(\theta) = \cos 2\theta. \quad (25)$$

In the case of a resonance at a  $P_{3/2}$  level, analogous calculations lead to the following expressions for the ionization probability for unpolarized atoms:

$$\frac{dw}{d\Omega} = \frac{e^4 \mathcal{E}_0^2 \bar{\mathcal{E}}_0^2}{144\hbar} \frac{R_{PS}^2 R_{DP}^2(E)}{\Delta_{3/2}^2 + (1/4)\Gamma^2} \sin^2 \theta (5 \sin^2 \theta + 2 \cos^2 \theta). \quad (26)$$

Also, the degree of polarization for electrons moving at an angle  $\theta$  from the  $z$  axis is

$$P(\theta) = 2 \frac{2 \sin^2 \theta + \cos^2 \theta}{5 \sin^2 \theta + 2 \cos^2 \theta}. \quad (27)$$

The integral degree of polarization is 9/11, i.e., about 82% (Ref. 42).

Resonance ionization in the scheme  $S \rightarrow P \rightarrow D$  has been observed experimentally,<sup>43</sup> and the observed degree of electron polarization agrees very well with the results above. The achievement of a polarization less than 100% in this simple case is consequence of the possible appearance of electrons with spin projections of opposite signs. It is possible, however, to choose more complicated ionization schemes in which the electron can have only a certain value of the spin projection, and in

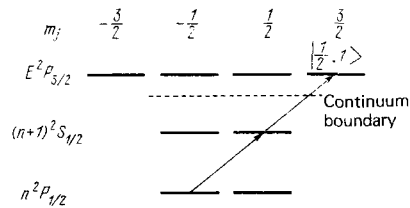


FIG. 12. Transition scheme for resonance ionization from an initial  $P$  state.

this case a 100% degree of polarization can be obtained.

One such scheme is based on the use of atoms in a  $P_{1/2}$  ground state.<sup>44</sup> This situation arises in atoms with three valence electrons (e.g., Ga, In, and Te). The selection rules incorporated in the transition scheme in Fig. 12 lead in this case to the appearance of photoelectrons only in states with  $l=1$ ,  $m_j=j=3/2$ , that is, in the  $\psi_{p_{1/2}}$  state. Electrons with the opposite spin projection do not arise in this resonance ionization process, and for this reason it is possible to achieve a 100% degree of polarization.

Another scheme, in which the same final state is reached (and in which a 100% degree of polarization can be achieved), is the three-photon resonance ionization of atoms with an  $|S_{1/2}\rangle$  ground state.<sup>45</sup> Figure 13 shows the corresponding transition scheme in the sodium atom under the influence of left-hand polarized light at the frequency of the first transition and right-hand polarized light at the frequencies of the second and third transitions.

At present, ionization through these last two schemes has not yet been achieved experimentally, but we see no serious obstacles here.

Up to this point it has been assumed that the energy of the exciting photon is approximately equal to the energy of transition to one of the levels  $P_{1/2}$  or  $P_{3/2}$  and that the effect of nonresonance levels could be ignored. A continuous transition from one of these resonances to the other through a nonresonance region has also been discussed.<sup>46</sup> The results show that the degree of polarization (Fig. 14) reaches 100% at certain frequencies in the region between the resonances. The experimental results on the frequency variation of the polarization agree qualitatively with these theoretical calculations. It should be kept in mind, however, that in the nonresonance region the probability itself (and thus the total number of photoelectrons) is sharply lower. For this

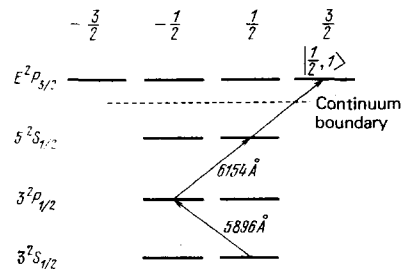


FIG. 13. Transition scheme for three-photon resonance ionization with opposite (circular) polarizations of the radiation.

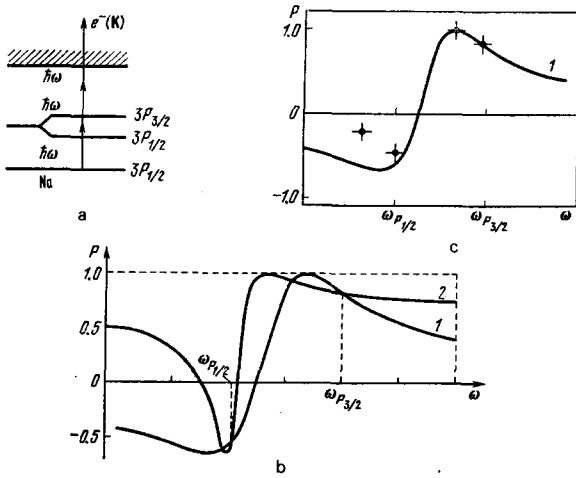


FIG. 14. Electron polarization with a detuning of the intermediate resonance state. a: Transition scheme. b: Calculations. 1—Identical contributions of the matrix elements for transitions through the  $P_{1/2}$  and  $P_{3/2}$  states; 2—the transition through the  $P_{3/2}$  states is dominant. c—Experimental data, normalized to a unit maximum polarization.

reason we conclude that nonresonance many-photon ionization cannot compete successfully with either the resonance process of the Fano effect in terms of the number of polarized electrons.

#### (b) Ionization in a resonance with a hyperfine-structure state

The bound-state spectrum of atomic electrons may of course contain, under certain conditions, a hyperfine structure as well as a fine structure, because of an interaction of the nuclear spin  $I$  and the total electron angular momentum  $J$ . Classically, the hyperfine splitting of levels can be described as the result of a precession of the vectors  $I$  and  $J$  with respect to one other, with a period  $T \sim \hbar/\Delta E_{HF}$ . The hyperfine splitting of the first few levels is of the order of  $\Delta E_{HF} \sim 10^{-3} \text{ cm}^{-1}$ .

In this case there are some stringent requirements on the exciting light:

- (1) Its spectral width must be smaller than the hyperfine splitting; i. e.,  $\Delta\omega \ll \Delta E_{HF}/\hbar$ .
- (2) The tuning error at resonance must be of the same order of magnitude as the width of the resonance state.
- (3) The exciting and ionizing fields must be weak enough that the field-induced perturbation of the resonance state is smaller than the hyperfine splitting (in the case of one-photon transitions, this condition is satisfied with  $\mathcal{E}_0 < 10^2 \text{ V/cm}$ ).
- (4) The pulse length of the exciting field must be larger than the precession time  $T$  (the typical value  $\Delta E_{HF} \sim 10^{-3} \text{ cm}^{-1}$  corresponds to  $t > 10^{-8} \text{ sec}$ ).

If any of these conditions is not met, the interaction of the nuclear spin with the electron spin will not be exhibited, so that only a fine-structure state can be excited. From the standpoint of experimental design,

these conditions do not run into any special difficulties, and modern lasers can furnish beams with the necessary characteristics. Certain hyperfine states have been excited in several experiments, including some carried out expressly to produce polarized electrons.<sup>18,43</sup>

Before going into the details of the transitions through a hyperfine resonance state, we should point out that the very fact that the electron angular momentum  $j$  interacts with the nuclear spin leads to a mixing of states with different  $z$  projections of the electron angular momentum,  $m_j$ . There are thus deviations from the  $m_j$  selection rules, so that the degree of polarization of the electrons should be smaller than in the case of a transition through a fine-structure state.

Let us assume that all the conditions above have been met, and let us examine the changes in the polarization properties of the photoelectrons which are caused by the hyperfine structure of the ground and resonance levels. We shall consider the effect of the hyperfine interaction on the atomic spectrum only, ignoring its effect on the matrix elements. This simplification allows us, in particular, to again use the wave functions  $\psi_{p\mu}$  in (3) as the continuum wave functions. Figure 15 shows the level scheme of the hyperfine structure of the ground ( $S$ ) and resonance ( $P$ ) states. Stationary states in the hyperfine level scheme have the quantum numbers  $n, l, j$  and also the quantum numbers of the nuclear spin  $I$ , the total angular momentum of the atom,  $F(F=I+j)$ , and the  $z$  projection of this angular momentum,  $m_F$ . In the absence of external fields, there is a degeneracy with respect to  $m_F$  in the hyperfine level scheme.

We assume that the atom is initially in the state  $|S; j=1/2; F; m_F\rangle$  and that the frequency  $\omega$  is at resonance with the transition to some level ( $P; j'; F'; m'_F$ ):

$$\Delta_{F'P} \equiv |E_{Pj'F'} - E_{SF} - \hbar\omega| \ll \Delta E_{HF}.$$

Let us use second-order perturbation theory to calculate the ionization probability density for a certain  $z$  projection of the electron spin,  $\mu$ . Assuming unpolarized atoms, we average the probability over the quantum number  $m_F$ , finding, by analogy with (23),

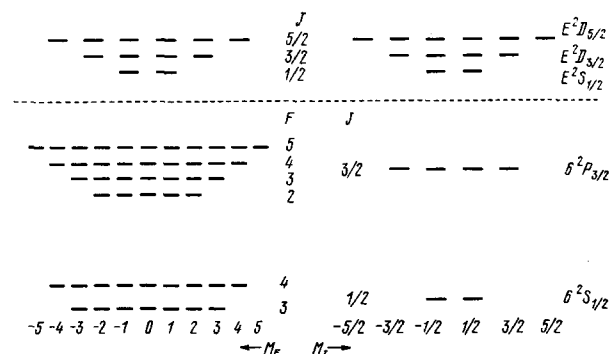


FIG. 15. States of cesium atoms which can occur when a hyperfine structure (left) and a fine structure (right) is displayed. The dashed line is the continuum boundary.

$$\frac{dw_{\mu}}{d\Omega} = \frac{2\pi/\hbar}{2F+1} \frac{\sqrt{2m^3E}}{\Delta_{F,F}^2 + (1/4)\Gamma^2} \times \sum_{m_I, m_F, m'_F} \left| \sum_{m'_I} \langle \Psi_{p_{\mu}, m_I} | \tilde{V} | Pj'F'm'_F \rangle \langle Pj'F'm'_F | V | SFm_F \rangle \right|^2 \quad (28)$$

where  $m_I$  is the  $z$  projection of the nuclear spin in the final state. If the resonance field is right-hand (left-hand) circularly polarized, then only a single term should remain in the sum over  $m'_F$ , specified by the selection rule  $\Delta m_F \equiv m'_F - m_F = \pm 1$ .

We first consider a resonance at the hyperfine sublevels of the  $P_{1/2}$  term in the case in which the exciting and ionizing waves are right-hand polarized. Expanding the  $|Fm_F\rangle$  and  $|F'm'_F\rangle$  wave functions in the wave functions of the fine-structure levels,  $|jm_j\rangle$  and  $|j'm'_j\rangle$ , and calculating the corresponding matrix elements, we find that the degree of polarization can be written

$$P_R = \frac{1 + aZ^2}{b + aZ^2}, \quad (29)$$

where  $Z = R_{11}/R_{31}$  is the ratio of the radial matrix elements  $R_{11}$  and  $R_{31}$  of the transitions from the resonance state to the continuum,  $P_{1/2} \rightarrow S_{1/2}$  and  $P_{1/2} \rightarrow D_{3/2}$ , respectively. The ratio  $Z$  must be calculated or found experimentally. For the  $7^2P_{1/2}$  level of the cesium atom,  $Z$  is approximately 1/2 (Ref. 18). Table I lists the constants  $a$  and  $b$  calculated for the cesium atom ( $I = 7/2$ ).

Comparing the degrees of polarizations shown in Table I for the various transitions with the degree of polarization for a transition through a  $P_{1/2}$  fine-structure state,  $P = -0.6$  (Subsection 32), we see that this particular example confirms our assertion regarding the advantages of transitions through fine-structure states.

Through analogous calculations, we can find the ionization probabilities for the case in which the resonance level is excited by right-hand polarized light and ionized by left-hand polarized light. The degree of polarization of the photoelectrons in this case,  $P_L$ , can again be written in the form in (29). The corresponding constants  $a$  and  $b$  are listed along with the values of  $P_L$  for  $Z = 1/2$  in Table II.

In the case of a resonance at hyperfine sublevels of a  $P_{3/2}$  state, the calculations from (28) are more involved, but there are no fundamental difficulties. The results of the calculations of Refs. 36 and 51 show that, as in the preceding case, the degree of polarization is smaller than the corresponding value for a transition

TABLE I. The parameters  $a$  and  $b$  in Eq. (29) and the degree of electron polarization  $P_R$  in the case of resonance at hyperfine-structure states of the cesium atom. The exciting and ionizing fields are right-hand polarized.  $Z = \frac{1}{2}$ .

$F'$	$F$	$a$	$b$	$P_R$
3	3	-1.25	-2.1	-0.29
	4	-0.63	-2.55	-0.31
4	3	-0.25	-1.75	-0.52
	4	-0.79	-1.93	-0.38

TABLE II. The same as Table I, but for left-hand polarization.

$F'$	$F$	$a$	$b$	$P_L$
3	3	-0.79	-1.93	-0.38
	4	-1.97	-2.36	-0.18
4	3	-3.25	-2.78	-0.05
	4	-1.27	2.11	-0.29

through a fine-structure state.

Up to this point we have made no distinction between the pulse lengths of the exciting and ionizing fields. There are some interesting features in the cases of very different pulse lengths and in which the fields are applied at different times.<sup>43</sup>

1. Let us first assume  $\tau \gg \hbar/\Delta E_{CT} \gg \tau$  (a short exciting pulse and a long ionizing pulse). In this case the large spectral width of the exciting field ( $\Delta\omega \geq 1/\tau$ ) overlaps the hyperfine structure of the ground and resonance states, so that corrections should be made to the detuning from resonance in Eq. (28) for the hyperfine interaction  $\Delta_{F,F} \rightarrow \Delta_j$ . The resonance levels do not single out any values of  $F$  and  $F'$ , so that the probability  $d\omega/d\Omega$  in (28) must be summed over  $F'$  and averaged over  $F$ . This result occupies an intermediate position between the results for ionization in the fine-structure and hyperfine-structure schemes. A field  $\mathcal{E}(\tau)$  with a broad spectrum excites the fine-structure states  $|j'm'_j\rangle$ , but after a long time  $\tau$  these states decay to hyperfine sublevels with a different energy  $E_{PF}$ . The ionization probability is found through an averaging over the beats between these states. A calculation of the degree of polarization in this scheme under conditions corresponding to a resonance at a  $P_{1/2}$  level yields  $P = -0.33$ .

2. We now assume that both pulses are short ( $\tau, \tau \ll \hbar/\Delta E_{HF}$ ) but that the ionizing pulse is delayed a time  $t \gg \hbar/\Delta E_{HF}$  with respect to the exciting pulse. It is easy to show that the ionization probability in this case can be written schematically as

$$\frac{dw_{\mu}}{d\Omega} \sim \left| \sum_{F'} \left[ \exp\left(-iE_{PF'} \frac{t}{\hbar}\right) \right] A_{F'} \right|^2, \quad (30)$$

where the  $A_{F'}$  are constants governed by the transition matrix elements.

It can be seen from (30) that the ionization probability oscillates as a function of the delay time  $t$ . This is a consequence of quantum beats, i. e., oscillations in the wave function of an atom which is initially excited to fine-structure states, which then decay (during the long time  $t$ ) to hyperfine states with different energies.

A detailed analysis of resonance ionization through a hyperfine-structure state thus confirms the qualitative conclusion that transitions through fine-structure states have advantages when it is desired to maximize the degree of electron polarization. Correspondingly, when practical use is made of resonance ionization as a source of polarized electrons, the laser pulse length should not be extremely long, and the spectral width should not be extremely small. The optimum values of

$\tau$  and the spectral width  $\Delta\omega$  are governed by

$$10^{-8} \text{ sec} \sim \frac{\hbar}{\Delta E_{\text{HF}}} > \tau > \frac{1}{\Delta\omega} > \frac{\hbar}{\Delta E_T} \sim 10^{-11} \text{ sec}.$$

Finally, we note that in the selective excitation of hyperfine sublevels or in resonance ionization through these sublevels a certain part of the angular momentum of the absorbed photons is acquired by the nuclear spin. Then a certain nuclear polarization arises in such processes.<sup>22</sup>

### (c) Resonance ionization in a strong external field

Intensification of the external field (or fields, if there are two, for excitation and ionization) increases the ionization probability and thus increases the number of polarized electrons only up to the point at which the field intensification begins to perturb the initial atomic states. There are three distinct effects here: resonance mixing in a two-level system\* with a ground and excited state,<sup>21</sup> the dynamic polarizability of the ground and excited resonance state,<sup>21,47</sup> and the ionizational broadening of the excited state. For the resonance transition to the continuum with which we are concerned, the resonance mixing (a shift of quasienergy levels) takes the form of a broadening of the resonance dependence of the ionization probability on the detuning of the exciting-field frequency from resonance. This broadening is customarily referred to as "field broadening," in distinction from ionizational broadening.

The relative importance of the various effects during resonance ionization varies with the intensities of the exciting and ionizing fields and with the degree of non-linearity of the corresponding transitions<sup>48</sup> (the degree to which these transitions are of a many-photon nature). Let us consider those effects which can reduce the degree of electron polarization as the external field is intensified. This effect has been observed experimentally.<sup>18,46</sup> As an example we can use the experimental data of Ref. 18 on the polarization in the two-photon ionization of the cesium atom through a hyperfine-structure state. The influence of these perturbations of the atomic spectrum on the photoelectron polarization was analyzed in Ref. 49. Without going into a detailed discussion of the various aspects of resonance ionization and the polarization of electrons in strong fields (see Ref. 49), we can estimate the optimum intensities of the exciting and ionizing fields for efficient sources of polarized electrons. By "optimum" here we mean those field intensities at which the perturbation of the spectrum does not yet significantly affect the degree of polarization but at which the number of photoelectrons is near the maximum possible value, i. e., the number in the interaction volume.

As before, we consider the case of resonance ionization in two fields: exciting and ionizing. To optimize the process it is evidently sufficient that the exciting field be at the saturation threshold  $\mathcal{E}_0 \sim \mathcal{E}_{\text{st}}(\hbar/\tau Ry)$ , while the ionizing field satisfies the condition of com-

plete ionization of the atom during the pulse,  $\mathcal{E}_0 \sim \mathcal{E}_{\text{st}} \sqrt{\hbar/\tau Ry}$ . The pulse length  $\tau$  in the case of a single-mode laser is unambiguously related to the laser spectral width:  $\Delta\omega \sim 1/\tau$ . According to the results in Subsection 3b the degree of electron polarization can be maximized if  $\hbar\Delta\omega$  is smaller than the scale interval of the level fine structure but larger than the hyperfine splitting. The corresponding pulse lengths  $\tau$  turn out to lie between  $10^{-8}$  and  $10^{-11}$  sec; then optimum conditions can be arranged, for example, with  $\tau = 10^{-9}$  sec.

The optimum intensities of the exciting and ionizing fields are then estimated to be  $\mathcal{E}_0 \sim 5 \times 10^2$  V/cm and  $\mathcal{E}_0 \sim 10^6$  V/cm.

### (d) Experimental procedure

Various specific schemes for resonance ionization have been discussed in Refs. 42, 44, 46, and 50 from the standpoint of producing polarized electrons. Detailed experiments have recently been reported on the polarization of the electrons produced in the resonance ionization of the cesium atom.<sup>18,43,51</sup> Some experimental results are also reported in the reviews in Refs. 42 and 46. In all cases, the atoms involved were alkali atoms, which have the lowest ionization potentials. For the most part, therefore, the ionization processes studied were two-photon processes.

As mentioned earlier, several characteristics of the laser beams (the spectral width, the intensity, and the pulse length) determine the states which are coupled by the resonance which results in the excitation of the atom. These can be either spin hyperfine states or fine-structure states (Fig. 15). It should be kept in mind that we are talking about the first excited states, not highly excited states. In order of magnitude, the hyperfine splitting (due to the nuclear magnetic moment) is  $10^{-3}$  of the fine-structure splitting (due to the magnetic moment of the atomic electron). The fine-structure splitting for the first excited states of the alkali atoms varies substantially for different atoms, from  $0.34 \text{ cm}^{-1}$  for lithium to  $554 \text{ cm}^{-1}$  for cesium. As the principal quantum number  $n$  is increased, the splitting falls off in proportion to  $n^3$  (for hydrogen-like states). It is clear from these figures that for only a few of the first excited states of several atoms is the hyperfine splitting greater than the natural level width, so that transitions between hyperfine states can in principle be arranged. It must be kept in mind here that the best modern lasers can furnish an output with a line width  $\Delta\omega$  as small as  $10^{-4} \text{ cm}^{-1}$ , with a frequency stability of the same order. Where necessary, however, radiation with a spectral width as large as  $10^2 \text{ cm}^{-1}$  can be used. Then various states can in principle be excited by choosing the appropriate atom, transition, laser frequency, and laser spectral width. We note that an electromagnetic wave is never perfectly circularly polarized, and at very high intensities the small part of the radiation which has the "wrong" polarization can be quite substantial in magnitude. Then there may be an effective ionization by this "wrongly" polarized radiation, with the consequence of a serious lowering of the degree of electron polarization.<sup>46</sup> With two la-

\*The terms "optical Autler-Townes effect" and "resonance Stark effect" are sometimes used in the literature.

sers in use, the experimental possibilities are much more extensive, since it is possible in principle to change the polarization of the ionizing radiation and to adjust the time interval between the excitation and the ionization in addition to optimizing the two intensities. By changing the polarization one can change the quantum numbers of the final state of the free electron. By changing the time interval between the exciting and ionizing pulses it is possible in principle to observe quantum beats between hyperfine states.

As a particular experiment we consider the two-photon resonance ionization of the cesium atom described in Ref. 18.

The general experimental arrangement is shown in Fig. 16. The method of intersecting light and atomic beams is used here. The output from a gallium arsenide semiconductor laser, used for excitation, and that from an argon ion laser, used to ionize the excited atoms, are combined into a common light beam. The pulsed semiconductor laser operates with a pulse length  $\sim 1 \mu\text{sec}$ . It emits linearly polarized light, which is converted into circularly polarized light by an external polarizer. During the pulse, the heating of the working medium changes the light frequency,  $\sim 11700 \text{ cm}^{-1}$ , by about  $1 \text{ cm}^{-1}$ , so that it is possible to excite two transitions from initial states with quantum numbers  $F=3$  and  $4$ , with an energy separation of  $\Delta E \approx 3 \times 10^{-1} \text{ cm}^{-1}$ . The time of the excitation of the transition to  $6^2P_{3/2}$  states with quantum numbers  $F'=2, 3, 4, 5$  (width  $\sim 10^{-4} \text{ cm}^{-1}$ ) is of the order of  $10^{-8} \text{ sec}$ . The time interval between the excitation of the initial states is  $150 \text{ nsec}$ . The pulse repetition frequency of the argon laser ( $\bar{n}\omega \sim 22000\text{--}20000 \text{ cm}^{-1}$ ) is the same as that of the semiconductor laser. The two lasers can be synchronized so precisely that it is possible to arrange ionization in the excitation of either the first state or the second state (or of both states at the same time).

The electrons produced in the volume in which the light and atomic beams intersect are accelerated in a static field, focused on the entrance of a magnetic analyzer, passed through a Wien filter, which rotates the spin through  $90^\circ$ , accelerated to about  $100 \text{ keV}$ , and sent to a Mott analyzer. The magnetic analyzer strongly suppresses the electron background from the residual gas and the walls of the interaction chamber (the rest

of the apparatus is described in some detail in Subsection 2c). The  $\text{Cs}^+$  ions are detected as well as the electrons, by an electron multiplier.

There are various systematic errors in the measurement of the degree of polarization, due to the Wien filter, the extraction of the electrons from the interaction volume, the asymmetry function  $S$  of the Mott analyzer (Subsection 2c), and the absolute calibration of this analyzer. The various sources of error were analyzed in detail in Ref. 36, and quantitative estimates were made.

### (e) Experimental results

Extremely little experimental information is available on the properties of the photoelectrons from resonance ionization. Only in the case of the two-photon ionization of the cesium atom<sup>18,43,51</sup> do we have reliable experimental data. In one case the cesium atoms were excited from initial  $6S_{1/2}$  states ( $F=3, 4$ ) by right-hand circularly polarized light to various hyperfine states of the  $6^2P_{3/2}$  level ( $F'=2, 3, 4, 5$ ). As mentioned in Subsection 3d, cesium atoms can be excited from either  $6S_{1/2}$  ( $F=3$ ) or  $6S_{3/2}$  ( $F=4$ ) states or from both states. The ionization from the excited states was caused by circularly polarized light of a different frequency; the polarization could be either right-hand or left-hand. The electron taken from the atom could thus be in several final states:  $S_{1/2}$ ,  $D_{3/2}$ , and  $D_{5/2}$  (Fig. 15). The transitions which can occur under such conditions were discussed in Subsection 3b.

The measurements were relative: The measured quantities depended on the direction of the circular polarization of the ionizing beam (right-hand or left-hand) through the radial parts of the matrix elements and the populations of the intermediate states. Specifically, these measured quantities are (1) the asymmetry in the ion yield,  $A_i = I_R/I_L$ , i.e., the ratio of the yields for the cases of right-hand (R) and left-hand (L) circular polarizations of the ionizing beam; (2) the degree of electron polarization for the right-hand circularly polarized ionizing beam,  $P_R$ ; and (3) the same, but for left-hand polarization,  $P_L$ .

All the data were obtained for the two initial states with quantum numbers  $F=3$  and  $4$ . In the  $F=4$  case, measurements were carried out with various excitation intensities (Fig. 17). These measurements showed that at an exciting field intensity  $\mathcal{E}_0 \approx 50 \text{ V/cm}$  the function  $A_i(\mathcal{E}_0^2)$  begins to become nonlinear, and the degree of polarization decreases. These deviations are evidently due to a mixing of resonance states. All the results discussed below were obtained at a low excitation intensities, at which these effects did not occur. The three measured quantities found under these conditions for both initial states are listed in Table III. The indicated errors are the statistical error and the error in the calibration of the Mott analyzer. Using the equations above and angular-momentum algebra, we can calculate values of the measured quantities. The results of these calculations agree well with the measured values if it is assumed that the matrix elements for transitions from the resonance state to various continuum states are

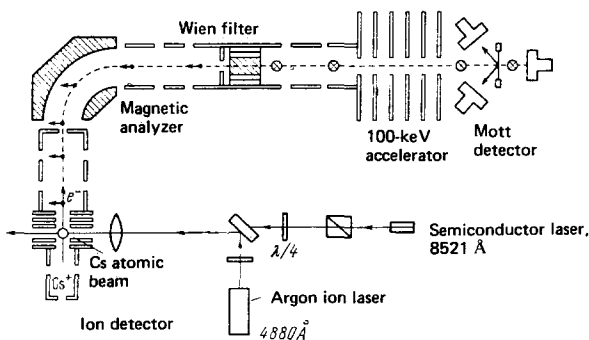


FIG. 16. Experimental arrangement for observing the polarized electrons produced in the resonance two-photon ionization of the cesium atom.<sup>18</sup>

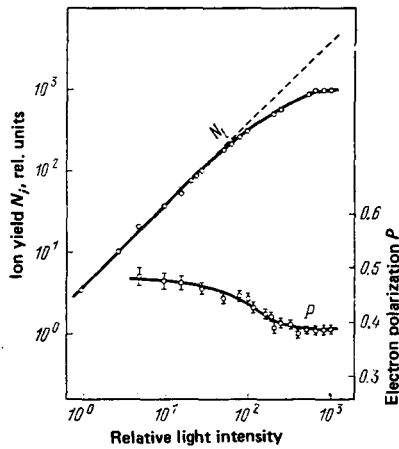


FIG. 17. Decrease in the degree of polarization with increasing intensity of the external field.

identical and that all transitions to intermediate states allowed by the selection rules occur. Interestingly, the calculated results indicate that the degree of polarization depends on the quantum number of the hyperfine structure of the intermediate state (within a factor of  $\sim 2$ ).

The same quantities were calculated under the assumption that transitions occur between fine-structure states and are governed by the selection rules for the orbital quantum number. When this assumption is used, the calculated results do not even agree qualitatively with experiment. Although we would not, on the basis of general considerations, expect transitions between fine-structure states to play a role in this experiment, the experimental confirmation that transitions do occur between hyperfine-structure states is of obvious importance.

Interesting experimental data on two-photon ionization of the cesium atom through the  $7^2P_{1/2}$  and  $7^2P_{3/2}$  intermediate resonance states were found in Ref. 43 (Fig. 18). Here a dye laser pumped by a nitrogen laser was used. The wavelength of the dye laser could be tuned from 4555 to 4593 Å, and the spectral width of the output was 12 Å. The pulse length was 1.5 nsec and the repetition period 100 Hz. The laser beam was used to both excite the cesium atoms and ionize the exciting atoms. In order to prevent mixing of the resonance states, and since the ionization probability was quite high, the radiation frequencies were chosen such that the excitation was caused by a small part of the radiation in the far wing of the output line (obviously, ionization was caused by all the radiation).

TABLE III. Experimental and calculated data on the degree of electron polarization  $P$  and the asymmetry in the ion emission  $A_i$  for the transitions  $6^2S_{1/2} (F=3, 4) \rightarrow 6^2P_{3/2} (F=2, 3, 4; 3, 4, 5) \rightarrow E^2S_{1/2}, E^2D_{3/2}, E^2D_{5/2}$  for right-hand (R) and left-hand (L) circular polarization of the ionizing radiation.

	Experiment	Calculation		Experiment	Calculation
$A_i (3 \rightarrow 4)$	$1.338 \pm 0.008$	1.472	$P_R (3)$	$0.390 \pm 0.020$	0.381
$P_R (4)$	$0.488 \pm 0.025$	0.520	$P_L (3)$	$0.018 \pm 0.014$	0.016
$P_L (4)$	$0.185 \pm 0.014$	0.225			

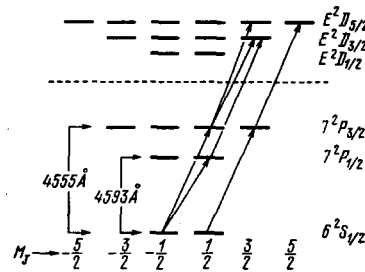


FIG. 18. Transition scheme for two-photon ionization through the  $7P$  multiplet in the cesium atom.

The exciting pulse was shorter than the precession period for the  $7^2P_{3/2}$  hyperfine states ( $F=2, 3, 4, 5$ ) in the range 5–20 nsec and comparable in magnitude to the precession period of the  $7^2P_{3/2}$  states: 2.7 nsec. In the transition through the  $7^2P_{3/2}$  states, the hyperfine interaction thus should not be important. The experiments yielded a degree of polarization  $P=0.8$  for the transition through the  $7^2P_{3/2}$  state and  $P=-0.6$  for the transition through the  $7^2P_{1/2}$  state (Fig. 19). The same values were found in the calculation of Ref. 52 for transitions through fine-structure states.

Data on the polarization of electrons produced in the resonance ionization of cesium through the  $7^2P_{3/2}$  states are also given in Ref. 42.

Some data on the electron polarization in the three-photon ionization of the sodium atom in a single-photon resonance with the  $3P_{1/2}$  and  $3P_{3/2}$  states are reported in Ref. 46.

An average current of polarized electrons up to  $10^{-11}$  A was achieved in Ref. 51 (of the order of  $10^4$  ions were formed in each pulse of the exciting laser, which operated at a repetition frequency of 20 kHz). This current is of the same order as the average polarized-electron current obtained in the Fano effect (Subsection 2c). In contrast with photoionization, however, the method of resonance many-photon ionization is far from being optimized. Various refinements, both quantitative and qualitative, can be suggested. Among the qualitative refinements is the possibility of arranging the resonance ionization of essentially any atom by intense visible light. This approach would of course bring into

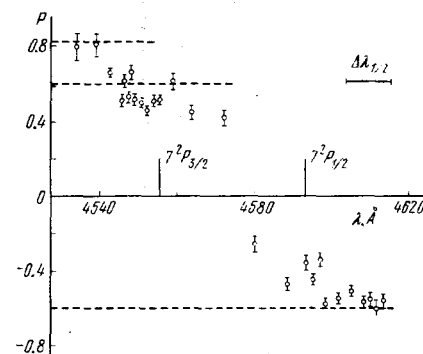


FIG. 19. Experimental data on the degree of polarization as a function of the frequency of the exciting light in a resonance with states of the  $7P$  multiplet in the cesium atom.

play not only the two- and three-photon processes but also many-photon processes in the strict sense of the term.

At first glance it might appear that there is a contradiction here: To drive processes with a high degree of nonlinearity effectively we need to use a high field intensity, but then there would be a strong perturbation of the atomic spectrum. The key is that a nonresonance perturbation does not broaden the atomic levels but simply changes the spectral structure; i.e., it does not prevent selective excitation of fixed states.

Turning to the quantitative refinements, we note that the major hope is to use visible-range lasers in resonance ionization. In the visible range there are many possibilities for selecting more intense lasers, for increasing the volume in which ionization can occur, and thus for increasing the number of target atoms and the number of ions formed.

On the basis of all the data, obtained in a variety of experiments, it can be suggested that a degree of ionization of atoms of the order of 100% can be achieved in a volume on the order of  $10^{-1}$  cm<sup>3</sup> with a beam density up to  $10^{12}$  cm<sup>-3</sup> through the use of high-power pulsed lasers with a repetition frequency of the order of  $1$  min<sup>-1</sup>.

At a higher repetition frequency, the interaction volume will be substantially smaller, since the beam must be focused at a lower laser power. It may be possible to arrange an interaction volume of this size by using multichannel collimators to form atomic beams or several parallel beams, as in Ref. 17.

#### f) Photoelectron polarization in the ionization of atoms with a selective populating of fine-structure sublevels

It is clear from the description of the process by which electrons become polarized in resonance ionization that one of the basic conditions for this effect is the selective excitation of fine-structure sublevels of the spectrum of atomic bound states. In principle, this selection can be performed by a different mechanism: through a nonuniform population of these levels. This situation obviously can be arranged only in atoms which have a very large fine-structure splitting. An example is the Tl atom, in which the energy difference between the ground level ( $P_{1/2}$ ) and the excited level ( $P_{3/2}$ ) is 0.96 eV, i.e., much larger than  $kT$ . Under equilibrium conditions, the Tl atoms are thus unpolarized, but their distribution with respect to electron angular momentum  $j$  is nonuniform. This situation is sufficient for the appearance of polarized photoelectrons in the direct ionization of these atoms by a circularly polarized field.<sup>53</sup> This effect is equivalent to the polarization during resonance ionization. Less predictable and more interesting is another feature of the photoionization of atoms from states with a given  $j$  which was observed in Ref. 54. It turns out that the mechanism for the polarization of photoelectrons in this case can be the additional selection in terms of emission direction. The photoelectrons can be polarized independently of the polarization of the ionizing radiation. In particular, polarization occurs

even when the atoms are ionized by linearly polarized or unpolarized radiation. Without going into the details of the calculations, which are carried out in Ref. 54, we give the equations for the degree of polarization of an atom with a  $P_{1/2}$  ground state when it is ionized by linearly polarized and unpolarized radiation:

$$P_{\text{lin}} = -6 \frac{R_S R_D \sin \delta}{R_S^2 - 2R_D^2} \frac{(\mathbf{s}[\mathbf{e}\boldsymbol{\kappa}])(\mathbf{e}\boldsymbol{\kappa})}{1 - (\beta/2)[3(\boldsymbol{\kappa}\mathbf{e})^2 - 1]}, \quad (31)$$

$$P_{\text{unpol}} = 3 \frac{R_S R_D \sin \delta}{R_S^2 - 2R_D^2} \frac{(\mathbf{s}[\mathbf{k} \times \boldsymbol{\kappa}](\mathbf{k}\boldsymbol{\kappa}))}{1 - (\beta/4)[3(\boldsymbol{\kappa}\mathbf{k})^2 - 1]}, \quad (32)$$

where  $\mathbf{s}$  and  $\boldsymbol{\kappa}$  are unit vectors along the directions of the electron spin and the electron momentum,  $\mathbf{k}$  and  $\mathbf{e}$  are unit vectors along the propagation and polarization directions of the radiation,  $\beta$  is the so-called asymmetry coefficient (which can be expressed in terms of the radial matrix elements for the transitions to the  $S$  and  $D$  states of the continuum,  $R_S$  and  $R_D$ ), and  $\delta = \delta_S - \delta_D$  is the difference between the phase shifts for scattering to the  $S$  and  $D$  states. Let us examine the basic features of the photoelectron polarization described by Eqs. (31) and (32).

1. In both cases the polarization direction  $\mathbf{s}$  is perpendicular to the scattering plane,  $(\boldsymbol{\kappa}, \mathbf{e})$  or  $(\boldsymbol{\kappa}, \mathbf{k})$ .

2. In contrast with the case of circular polarization, the electrons can become polarized only through selection in terms of emission direction. An averaging of Eqs. (31) and (32) over the direction of the vector  $\boldsymbol{\kappa}$  causes these equations to vanish.

3. The degree of polarization  $P$  is governed by  $\delta$ , the difference in scattering phase shifts. This difference is generally not small [in contrast with the phase shift difference  $\delta_{lj}$  with given values of  $l$  and different values of  $j$  in the Fano effect (Subsection 2b)].

Experimental observation of electron polarization under these conditions can be interesting in its own right and also for studying the characteristics of the photoionization process: the matrix elements  $R_S$  and  $R_D$  and the difference  $\delta$ .

Finally, we note that Cherepkov<sup>54</sup> has given a complete description of the polarization of photoelectrons with a given direction of motion for the photoionization of any atoms with any bound-state quantum numbers, for various relations among the populations of the fine-structure sublevels and for radiation of arbitrary polarization.

#### 4. CONCLUSION

To conclude this examination of the various physical effects which lead to a polarization of the electrons ejected from unpolarized atoms by photons, we review briefly the present state of the research. It is clear from the material presented in this review that the research is in different stages in different fields, ranging from preliminary qualitative results in some fields to an effort in others to optimize the experimental conditions for practical implementation. Under these circumstances it is important to define the key questions facing the theoreticians and the basic problems facing



the experimentalists.

The basic thrust of theoretical and experimental research in this area in the near future should obviously take the form of a systematic study of the electron polarization in the direct and resonance ionization of various atoms in experiments of various designs. There is still much theoretical work to be done here, since, at least in the case of resonance ionization, all the basic results which have been found refer to the simple scheme of  $S-P-D$  transitions in monovalent atoms. These results must obviously be extended to more general cases for a broad study of processes in various atoms. Of particular interest here are the processes involved in the photoionization of atoms with several valence electrons. It would also be worthwhile to analyze theoretically the possibilities of electron polarization in resonance ionization making use of emission-direction selection.

A natural problem for the experimentalists is the Fano effect induced by an intense external field; there is obvious interest in the observation and study of this effect. We also need a detailed study of the polarization properties of the electrons produced in various resonance-ionization schemes, primarily the observation of 100% polarization.

If we focus on the polarized electrons themselves and look at the physical effects described above as various methods for producing them, we see that only in the case of the Fano effect can we talk confidently about the quantitative characteristics of the method at this point. Such a discussion would be premature for all the other cases, especially in regard to the ratio of the number of polarized electrons, since this ratio is strongly affected by various technical characteristics of the apparatus used.

At the present state of research on electron collisions with atoms and molecules there is an urgent need for various experiments with polarized electrons, so that a field of application awaits developments in the effects discussed in this review.

- <sup>1</sup>N. F. Mott and H. S. W. Massey, *Theory of Atomic Collisions*, Oxford Univ. Press, London, 1965 (Russ. transl. Mir, Moscow, 1969).
- <sup>2</sup>J. Kessler, *Rev. Mod. Phys.* **41**, 3 (1969).
- <sup>3</sup>D. W. Walker, *Adv. Phys.* **20**, 257 (1971).
- <sup>4</sup>P. S. Farago, *Rept. Progr. Phys.* **34**, 1055 (1971).
- <sup>5</sup>J. Kessler, *Polarized Electrons*, Springer-Verlag, Berlin-Heidelberg-New York, 1976.
- <sup>6</sup>G. F. Drukarev, in: *Voprosy atomnykh stolknoveniy* (Questions of Atomic Collisions), Izd. Leningr. Univ., Leningrad, 1975, No. 1, p. 10.
- <sup>7</sup>G. F. Drukarev, *Stolknoveniya elektronov s atomami i molekulami* (Electron Collisions with Atoms and Molecules), Nauka, Moscow, 1978.
- <sup>8</sup>S. M. Bilen'kiĭ, L. I. Lapidus, and R. M. Ryndin, *Usp. Fiz. Nauk* **84**, 243 (1964) [*Sov. Phys. Usp.* **7**, 721 (1965)].
- <sup>9</sup>A. V. Vaganov, *Usp. Fiz. Nauk* **119**, 257 (1976) [*Sov. Phys. Usp.* **19**, 481 (1976)].
- <sup>10</sup>V. W. Hughes, R. L. Long, M. S. Lubell, M. Posner, and W. Raith, *Phys. Rev. A* **5**, 195 (1972).
- <sup>11</sup>M. J. Alguard *et al.*, *Bull. Am. Phys.* **21**, 35 (1976).
- <sup>12</sup>V. D. Ob'edkov, *Pis'ma Zh. Eksp. Teor. Fiz.* **21**, 220 (1975) [*JETP Lett.* **21**, 98 (1975)].
- <sup>13</sup>P. J. Keliber, R. E. Gleason, and G. K. Walters, *Phys. Rev. A* **11**, 1279 (1975).
- <sup>14</sup>D. T. Pierce and F. Meier, *Phys. Rev. B* **13**, 5484 (1976).
- <sup>15</sup>N. Müller, W. Eckstein, W. Helland, and W. Zinn, *Phys. Rev. Lett.* **29**, 1651 (1972).
- <sup>16</sup>E. Kisker, G. Baum, A. H. Mahan, W. Raith, and K. Schröder, *Phys. Rev. Lett.* **36**, 982 (1976).
- <sup>17</sup>W. von Drachenfels, U. T. Koch, R. D. Lepper, T. M. Müller, and W. Paul, *Z. Phys.* **269**, 387 (1974).
- <sup>18</sup>E. H. A. Granneman, M. Klewer, K. J. Nygaard, and M. J. Van der Weil, *J. Phys. Ser. B* **9**, No. 5, L87 (1976).
- <sup>19</sup>U. Fano, *Phys. Rev.* **178**, 131 (1969); **184**, 250 (1969).
- <sup>20</sup>A. I. Andryushin and M. V. Fedorov, *Zh. Eksp. Teor. Fiz.* **75**, 2037 (1978) [*Sov. Phys. JETP* **48**, 1025 (1978)].
- <sup>21</sup>N. B. Delone and V. P. Kraĭnov, *Atom v sil'nom svetovom pole*. (An Atom in a Strong Optical Field), Atomizdat, Moscow, 1978, Ch. 9.
- <sup>22</sup>N. B. Delone, B. A. Zon, and M. V. Fedorov, *Pis'ma Zh. Tekh. Fiz.* **4**, 229 (1978) [*Sov. Tech. Phys. Lett.* **4**, 94 (1978)]; *Zh. Eksp. Teor. Fiz.* **76**, 505 (1979) [*Sov. Phys. JETP* **49**, 155 (1978)]; M. V. Fedorov, *Opt. Commun.* **26**, 183 (1978).
- <sup>23</sup>A. I. Akhiezer and V. B. Berestetskiĭ, *Kvantovaya elektrodinamika* (Quantum Electrodynamics), Nauka, Moscow, 1969.
- <sup>24</sup>L. D. Landau and E. M. Lifshitz, *Kvantovaya mekhanika*, Nauka, Moscow, 1974, § 29 (Quantum Mechanics—Nonrelativistic Theory, Pergamon Press, New York, 1977).
- <sup>25</sup>G. Marr, *Photoionization Processes in Gases*, Academic Press, New York-London, 1967.
- <sup>26</sup>I. I. Sovel'man, *Vvedenie v teoriyu atomnykh spektrov* (Introduction to the Theory of Atomic Spectra), Nauka, Moscow, 1977, § 34.
- <sup>27</sup>J. Samson, *Phys. Rev.* **28**, 303 (1976).
- <sup>28</sup>N. A. Cherepkov, *Phys. Lett. A* **40**, 119 (1972).
- <sup>29</sup>M. J. Seaton, *Proc. R. Soc. A* **208**, 408, 418 (1951).
- <sup>30</sup>A. Burgess and M. Seaton, *Rev. Mod. Phys.* **30**, 992 (1958).
- <sup>31</sup>J. W. Cooper, *Phys. Rev.* **128**, 681 (1962).
- <sup>32</sup>J. Cooper and U. Fano, in: *Proceedings of the Second International Conference on the Physics of Electron and Atomic Collisions*, New York, 1960.
- <sup>33</sup>U. Fano and J. Cooper, *Spectral Distributions of Oscillator Strengths in Atoms* (Russ. transl., Nauka, Moscow, 1972).
- <sup>34</sup>U. Heizmann, J. Kessler, and J. Lorenz, *Z. Phys.* **240**, 42 (1970).
- <sup>35</sup>G. Baum, M. S. Lubell, and W. Raith, *Phys. Rev. A* **5**, 1073 (1972).
- <sup>36</sup>E. Granneman, *Thèse*, Amsterdam, 1976.
- <sup>37</sup>W. Drachenfels, U. Koch, T. Müller, and H. Schaefer, *Phys. Rev. A* **51**, 445 (1975).
- <sup>38</sup>J. Van Klinken, *Nucl. Phys.* **75**, 161 (1966).
- <sup>39</sup>G. Holzwarth and H. Meister, *Table on Asymmetry for Mott Scattering of Electrons by Screened Gold Nuclei*, Univ. München, 1964.
- <sup>40</sup>L. Armstrong Jr., B. L. Beers, and S. Feneuille, *Phys. Rev. A* **12**, 1903 (1975).
- <sup>41</sup>Yu. I. Geller and A. B. Popov, *Kvantovaya Elektron. (Moscow)* **3**, 1129 (1976) [*Sov. J. Quantum Electron.* **6**, 606 (1976)].
- <sup>42</sup>H. Zeman, in: *Proceedings of the International Symposium on Electron and Photon Interactions with Atoms*, New York, 1976, p. 581.
- <sup>43</sup>E. H. A. Granneman, M. Klewer, G. Nienhuis, and M. J. Van der Weil, *J. Phys. Ser. B* **10**, 1625 (1977).
- <sup>44</sup>P. S. Farago and D. W. Walker, *J. Phys. Ser. B* **6**, L280 (1973).
- <sup>45</sup>P. Lambropoulos, *J. Phys. Ser. B* **7**, L33 (1974).
- <sup>46</sup>P. Lambropoulos and M. Lambropoulos, cited in Ref. 42, p. 525.
- <sup>47</sup>N. B. Delone, B. A. Zon, V. P. Kraĭnov, and V. A. Kho-

- dovoi, Usp. Fiz. Nauk **120**, 3 (1976) [Sov. Phys. Usp. **19**, 711 (1976)].
- <sup>48</sup>M. V. Fedorov, Izv. Akad. Nauk SSSR, Ser. Fiz. **41**, 2569 (1977).
- <sup>49</sup>M. V. Fedorov, Zh. Eksp. Teor. Fiz. **74**, 1342 (1978) [Sov. Phys. JETP **47**, 702 (1978)].
- <sup>50</sup>P. Lambropoulos, Phys. Rev. Lett. **30**, 413 (1973).
- <sup>51</sup>E. H. A. Granneman, M. Klewer, and M. J. Van der Wiel, J. Phys. Ser. B **9**, 2819 (1976).
- <sup>52</sup>M. R. Teague, P. Lambropoulos, D. Goodmanson, and D. W. Norcross, Phys. Rev. **A14**, 1057 (1976).
- <sup>53</sup>N. A. Cherepkov, Zh. Eksp. Teor. Fiz. **65**, 933 (1973) [Sov. Phys. JETP **38**, 463 (1974)].
- <sup>54</sup>N. A. Cherepkov, Zh. Eksp. Teor. Fiz. **75**, 827 (1978) [Sov. Phys. JETP **48**, 418 (1978)]; Phys. Lett. **A66**, 204 (1978).

Translated by Dave Parsons