

Polarization effects in multiphoton ionization of alkali atoms

G. A. Delone, N. L. Manakov, M. A. Preobrazhenskii, and L. P. Rapoport

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

(Submitted November 4, 1975)

Zh. Eksp. Teor. Fiz. 70, 1234–1242 (April 1976)

General formulas are obtained for the ionization probability of an atom in an elliptically polarized field. Results of numerical calculations are given for the cross sections for two-photon to five-photon ionization of alkali atoms by radiation with frequencies corresponding to the first and second harmonics of the radiation of ruby and neodymium lasers in fields of linear and circular polarization, together with experimentally measured ratios of the cross sections for multiphoton ionization in fields of linear and circular polarization for potassium and sodium atoms at the frequencies of a neodymium laser and its second harmonic. The results of theoretical and experimental investigations which have been carried out up to the present time are compared.

PACS numbers: 32.10.Qy

1. INTRODUCTION

The intensive study of multiphoton ionization of atoms has led to the possibility at the present time of clearly formulating and classifying the various phenomena arising in transition of an atomic electron from the ground state to a state of the continuum in a field of optical frequency. The great variety of possible situations is due to the effect on such a transition of both the spectral structure of a specific atom and the field characteristics—frequency, polarization, and intensity. In the present work we consider the question of the effect of polarization of an external field of optical frequency on the probability of the direct process (no intermediate resonances) of multiphoton ionization of alkali atoms under conditions where $\gamma = \omega\sqrt{2mI}/eE \gg 1$ (ω and E are the frequency and intensity of the electromagnetic field, m and e are the mass and charge of the electron, and I is the ionization potential of the atom). It is known^[1] that the probability of such processes is described by a functional dependence of the form $W \sim E^{2N}$ (N is the number of photons in the multiphoton ionization process) and that they occur in fields rather small in comparison with intra-atomic fields, and therefore there is reason to suppose that the ionization process should be satisfactorily described in terms of perturbation theory in the interaction of the atom with the electromagnetic field.

The specific selection rules in the matrix elements of perturbation theory for radiation of linear and circular polarization lead to various expressions for the probabilities, depending on the type of polarization.^[2–4] Numerical calculation of the ionization probability is complicated by the need of calculating compound matrix elements of N -th order perturbation theory which include summation over the discrete and continuous spectra of the atom. Thus, calculations of the probabilities of two- and three-photon ionization of alkali atoms by linearly polarized radiation carried out by Bebb^[5] are based on taking into account a finite number of terms in the infinite series of perturbation theory. Here the contribution of the continuous spectrum is taken into account. The method of “averaging” of oscillator

strengths in compound matrix elements, used by Morton,^[6] leads to an indeterminate accuracy of the calculations. Correct calculations have been carried out only for two-photon ionization of alkali metal atoms by Mizuno^[7] and Choudhury and Gupta.^[8] To avoid the difficulties associated with summation of the perturbation theory series, the calculations have utilized the method of Dalgarno^[9] which permits reducing the problem to numerical integration of an inhomogeneous differential equation of second order. However, the generalization of this method to the case $N > 2$ presents significant technical difficulties.

In the present work we give the results of numerical calculations of the quantities^[1] $\sigma_N = W_N/F^N$ (F is the photon flux density) for two- to five-photon ionization of alkali atoms by radiation with frequencies corresponding to the first and second harmonics of ruby and neodymium laser radiation, and the experimentally measured ratios of the cross sections for potassium and sodium atoms at the neodymium laser frequency and its second harmonic.

In Sec. 2 we obtain general formulas for the ionization probability of an atom in an elliptically polarized field. In Sec. 3 we give the results of numerical calculations of the cross sections for alkali atoms. In Secs. 4 and 5 we describe an experiment to measure the ratio of the ionization cross sections of sodium and potassium in fields of linear and circular polarization and present a comparison of theoretical and experimental results, including the results of other experimental studies^[10–12] carried out up to the present time.

2. PROBABILITY OF MULTIPHOTON IONIZATION IN AN ELLIPTICALLY POLARIZED FIELD

The amplitude of N -photon ionization of an atom with one valence electron in the first nonvanishing order of perturbation theory can be represented in the form (the atomic system of units is used)

$$U_{i \rightarrow E_f}^{(N)} = \left[\frac{2\pi^3}{k_e} \right]^{1/2} \sum_{LM} i^L \exp(i\eta_L) Y_{LM}(k_e) \times \langle E, LM | V(\mathbf{r}_N) G_{E_i + (N-1)\omega}(\mathbf{r}_N, \mathbf{r}_{N-1}) \dots G_{E_i + \omega}(\mathbf{r}_2, \mathbf{r}_1) V(\mathbf{r}_1) | n_i l_i m_i \rangle. \quad (1)$$

Here $|n_i l_i m_i\rangle$ and $|E_f L M\rangle$ are the wave functions of the initial and final states of the atom; k_e and η_L are the wave vector and phase of the incident electron; $G_E(\mathbf{r}, \mathbf{r}')$ is the Green's function of the optical electron; $V(\mathbf{r}) = -(1/2)E(\mathbf{e} \cdot \mathbf{r})$ is the Hamiltonian of the dipole interaction of the electron with the field $E(t) = E\mathbf{e} \cos \omega t$ (as a result of the large population numbers of the modes, the field can be considered classical).

For elliptically polarized radiation the polarization vector \mathbf{e} has the form $\mathbf{e} = \sin \psi \mathbf{e}_{-1} - \cos \psi \mathbf{e}_1$, where \mathbf{e}_1 and \mathbf{e}_{-1} are spherical unit vectors. It is assumed that the wave vector \mathbf{k} is directed along the z axis. The angle ψ varies in the range $0 \leq \psi \leq \pi/2$; here $\psi = \pi/4$ corresponds to linear polarization along the x axis and $\psi = 0$ ($\pi/2$) to right-hand (left-hand) circular polarization. We limit ourselves to the case of an initial S state ($l_i = 0$). Expanding G_E in Eq. (1) in spherical functions, we can obtain an expression for the probability W_N integrated over the emitted electron angles:

$$W_N = W_N^{(e)} + 2\pi \left(\omega \frac{F}{F_0} \right)^N \sum_{k=1}^{N(N/2)} A_N^{(k)} \sin^{2k} 2\psi,$$

$\langle N/2 \rangle$ is the integral part of $N/2$,

$$W_N^{(e)} = \int |U_{i \rightarrow E}^{(N,e)}|^2 \delta(E_f - E_i - N\omega) \frac{dk_z}{2\pi^3} = \frac{2\pi N!}{(2N+1)!!} \left(\omega \frac{F}{F_0} \right)^N |T_{N, N-1, \dots, 2}|^2 \quad (2)$$

is the probability of ionization for circularly polarized light. Here $F = cE^2/8\pi\hbar\omega$ is the density of the photon flux in $\text{cm}^{-2} \text{sec}^{-1}$, $F_0 = c/2\pi\alpha_0^2 = 3.22 \times 10^{34} \text{cm}^{-2} \text{sec}^{-1}$ is the atomic unit of flux density,

$T_{L, l_{N-1}, \dots, l_2}$

$$= \langle E_f L | r_N g_{N-1}(E_f + [N-1]\omega; r_N; r_{N-1}) r_{N-1} \dots g_1(E_f + \omega; r_2; r_1) r_1 | n_i \rangle$$

is the radial compound matrix element and g_l is the radial part of the Green's function. The expressions for $A_N^{(k)}$ in the general case are quite cumbersome and therefore below we give A for $N = 2-4$:

For $N = 2, 3$

$$2\pi \left(\omega \frac{F}{F_0} \right)^N A_N^{(1)} = W_N^{(1)} - W_N^{(e)}$$

For $N = 4$

$$A_4^{(1)} = \frac{3}{10} \left(\frac{2}{9} T_{012} + \frac{8}{45} T_{212} + \frac{6}{35} T_{232} \right)^2 - \frac{16}{735} T_{234}^2,$$

$$A_4^{(2)} = \frac{1}{81} \left(T_{010} + \frac{4}{5} T_{210} \right)^2 - \frac{8}{1575} T_{234}^2 - \frac{1}{3} A_4^{(1)}.$$

The ionization probability for linearly polarized light has the form

$$W_N^{(l)} = 2\pi \left(\omega \frac{F}{F_0} \right)^N \sum_{l_1, l_{N-1}, \dots, l_2, l'_{N-1}, \dots, l'_2} \frac{1}{2L+1} C^L(l_2, \dots, l_{N-1}) \times C^L(l'_2, \dots, l'_{N-1}) T_{L, l_{N-1}, \dots, l_2} T_{L, l'_{N-1}, \dots, l'_2} \times \frac{C^L(l_2, \dots, l_{N-1})}{(1 \cdot \delta_{l_0} + 2\delta_{l_2}) (l_2 \delta_{l_2, l_{N-1}} + l_2 \delta_{l_2, l_{N-1}+1}) \dots (l_{N-1} \delta_{L, l_{N-1}-1} + L \delta_{L, l_{N-1}+1})} 1 \cdot 3 (2l_2 + 1) \dots (2l_{N-1} + 1), \quad (3)$$

δ is the Kronecker delta, and the summation in Eq. (3) is carried out over all l satisfying the condition $l_{i+1} = l_i \pm 1$.

The quantities $C^L(l_2, \dots, l_{N-1}) T_{L, l_{N-1}, \dots, l_2}$ in Eq. (3) can be considered as partial amplitudes corresponding to different combinations of orbital angular momenta of the electron in intermediate states permitted by the selection rules, and in this case $U_{i \rightarrow E_f}^{(N,l)}$ represents the sum of the probability amplitudes for transition of the electron to the continuum by various branches $0-1-l_2 - \dots - L$.

In the above we have not considered the statistics of the field—the radiation was assumed completely coherent. In the approximation of completely chaotic radiation we can obtain a general expression W_N for a partially polarized field. In particular, for $N = 2$ we have

$$W_2 = 2\pi \left(\omega \frac{F}{F_0} \right)^2 \left[\frac{1}{9} T_0^2 (1 - P^2 + 2\xi_3^2) + \frac{1}{5} T_2^2 (7 + 5P^2 - 4\xi_3^2) \right],$$

where P is the degree of partial polarization and ξ_3 is the Stokes parameter which determines the degree of linear polarization (the degree of circular polarization is $\xi_2 = \sqrt{P^2 - \xi_3^2}$).

As already noted in an earlier article,^[2] the characteristic feature of Eq. (2) is that in each interval between resonances $|E_n| < E_x < |E_{n+1}|$ (E_n and E_{n+1} are the energies of the atomic levels) there exists a frequency ω_0 for which $W_N^{(e)}(\omega_0) = 0$. Taking account of the nondipole nature of the interaction or of relativistic corrections leads to some shift of the point ω_0 . A non-zero probability in this case is obtained only when higher orders of perturbation theory are taken into account. This means that in the experimental measurement of the probability of ionization by radiation with frequency $\sim \omega_0$ the exponent of the power dependence of the probability on intensity can be greater than N for a sufficiently narrow spectral width of the radiation line.

The probability ratio $R = W_N^{(e)}/W_N^{(l)}$ is an interesting problem. Numerical calculations show that in most cases in the sum (3) the matrix element $T_{N, N-1, \dots, 2}$ is maximal. Dropping the remaining terms in Eq. (3), we have for R the following estimate^[4]:

$$R = (2N-1)!!/N! \quad (4)$$

It must be recalled that expression (4) gives an upper limit for R and in a number of cases may be violated. In the first place, the estimate (4) is not valid for $\omega \approx \omega_0$, where $W_N^{(e)} \approx 0$, but $W_N^{(l)} \neq 0$ if terms with $L < N$ are taken into account in the sum (3). In the second place, at certain frequencies $W_N^{(l)}$ can have intermediate N -photon resonances at levels with orbital angular momentum $l < n$ which do not appear in $W_N^{(e)}$. Here $W_N^{(l)} > W_N^{(e)}$. With increase of N the probability of such almost resonance situations increases (an exception is the hydrogen atom, where the resonances $W_N^{(l)}$ and $W_N^{(e)}$ coincide). In addition, for large N the number of matrix elements omitted in Eq. (3) rises rapidly and, as shown by numerical calculations^[13] for hydrogen, their combined contribution begins to exceed $T_{N, N-1, \dots, 2}$ for $N \geq 5$. Thus, the estimate (4) is qualitatively valid only for small N .

We note that in solution of the problem of multiphoton

TABLE 1.

Atom	N	ω	$\sigma_N^{(l)}$	$\sigma_N^{(c)}$	a_N	b_N	c_N
Cs	2	$2\omega_N$	$7.41 \cdot 10^{-49}$	$1.01 \cdot 10^{-48}$	$2.89 \cdot 10^{-49}$	$5.70 \cdot 10^{-49}$	$6.03 \cdot 10^{-49}$
Cs	2	$2\omega_R$	$2.52 \cdot 10^{-49}$	$2.74 \cdot 10^{-49}$	$1.08 \cdot 10^{-49}$	$1.96 \cdot 10^{-49}$	$1.47 \cdot 10^{-49}$
Cs	3	ω_R	$9.57 \cdot 10^{-76}$	$1.36 \cdot 10^{-75}$	$1.90 \cdot 10^{-75}$	$7.38 \cdot 10^{-75}$	$2.70 \cdot 10^{-75}$
Cs	4	ω_N	$1.09 \cdot 10^{-107}$	$1.83 \cdot 10^{-107}$	—	—	—
Rb	2	$2\omega_N$	$1.80 \cdot 10^{-49}$	$2.31 \cdot 10^{-49}$	$7.74 \cdot 10^{-50}$	$1.48 \cdot 10^{-49}$	$1.38 \cdot 10^{-49}$
Rb	2	$2\omega_R$	$2.53 \cdot 10^{-50}$	$3.62 \cdot 10^{-50}$	$1.11 \cdot 10^{-50}$	$2.44 \cdot 10^{-50}$	$2.16 \cdot 10^{-50}$
Rb	3	ω_R	$1.14 \cdot 10^{-78}$	$2.68 \cdot 10^{-78}$	$3.55 \cdot 10^{-78}$	$1.48 \cdot 10^{-77}$	$5.33 \cdot 10^{-78}$
Rb	4	ω_N	$1.32 \cdot 10^{-107}$	$2.67 \cdot 10^{-107}$	—	—	—
Li	2	$2\omega_N$	$2.42 \cdot 10^{-49}$	$3.44 \cdot 10^{-49}$	$6.09 \cdot 10^{-49}$	$1.42 \cdot 10^{-49}$	$2.05 \cdot 10^{-49}$
Li	3	$2\omega_N$	$7.14 \cdot 10^{-82}$	$3.21 \cdot 10^{-83}$	$3.44 \cdot 10^{-82}$	$1.23 \cdot 10^{-82}$	$6.39 \cdot 10^{-83}$
Li	4	ω_R	$3.52 \cdot 10^{-106}$	$7.64 \cdot 10^{-106}$	—	—	—
Li	5	ω_N	$1.37 \cdot 10^{-137}$	$5.08 \cdot 10^{-137}$	—	—	—
K	2	$2\omega_N$	$1.61 \cdot 10^{-49}$	$1.87 \cdot 10^{-49}$	$6.86 \cdot 10^{-50}$	$1.39 \cdot 10^{-49}$	$1.12 \cdot 10^{-49}$
K	2	$2\omega_R$	$3.08 \cdot 10^{-50}$	$3.32 \cdot 10^{-50}$	$1.35 \cdot 10^{-50}$	$2.41 \cdot 10^{-50}$	$1.98 \cdot 10^{-50}$
K	3	ω_R	$1.87 \cdot 10^{-79}$	$4.60 \cdot 10^{-79}$	$6.41 \cdot 10^{-79}$	$2.29 \cdot 10^{-79}$	$9.15 \cdot 10^{-79}$
K	4	ω_N	$4.38 \cdot 10^{-107}$	$1.76 \cdot 10^{-106}$	—	—	—
Na	2	$2\omega_R$	$6.26 \cdot 10^{-52}$	$5.56 \cdot 10^{-52}$	$3.08 \cdot 10^{-52}$	$3.67 \cdot 10^{-52}$	$3.32 \cdot 10^{-52}$
Na	3	ω_R	$9.33 \cdot 10^{-78}$	$2.33 \cdot 10^{-77}$	$1.88 \cdot 10^{-77}$	$1.09 \cdot 10^{-76}$	$4.63 \cdot 10^{-77}$
Na	3	$2\omega_N$	$2.32 \cdot 10^{-78}$	$7.78 \cdot 10^{-79}$	$2.61 \cdot 10^{-78}$	$5.34 \cdot 10^{-78}$	$1.55 \cdot 10^{-78}$
Na	5	ω_N	$3.96 \cdot 10^{-138}$	$1.52 \cdot 10^{-139}$	—	—	—

ionization in the quasiclassical approximation^[14] ($N \gg 1$) the ratio R systematically decreases with increase of N . This is due to the fact that in the quasiclassical regime the structure of the spectrum of bound states is not taken into account and R is determined only by the ratio of probabilities of photoelectron emission in states with different orbital angular momentum l for a fixed energy. In this case from classical considerations the probability of large values of l ($l=N$ in a circular field) is strongly suppressed.

3. NUMERICAL RESULTS FOR ALKALI ATOMS

Values of the cross sections for multiphoton ionization of the alkali atoms $\sigma_N^{(l)} = W_N^{(l)}/F^N$ and $\sigma_N^{(c)} = W_N^{(c)}/F^N$ have been calculated for all degrees of multiphoton ionization N possible for the frequencies of ruby and neodymium lasers and their second harmonics: $N=2-5$. In calculation of the matrix elements T we used the wave functions and the radial Green's function $g_l(E_x; r, r')$ in the approximation of the quantum defect method (QDM). Details of the calculations with g_l have been described in Refs. 2 and 15. Here we note only that for energies $E_x - E_n = -Z^2/2(n_r + l + 1)^2$ corresponding to the hydrogen spectrum, calculation of g_l by means of the formulas given in Ref. 15 is impossible. This is due to the fact that g_l has the form of the difference of two terms, each of which diverges for $X = (E_x - E_n) \rightarrow 0$. To obtain a finite result in this case it is necessary to expand both terms of g_l in the small parameter X , to carry out a subtraction, and to set $X=0$. For example, for $n_r = l = 0$ we have

$$g_{l=0}(E_x; r, r') = 4Ze^{-2(r+r')} \left\{ 2 \ln Zr_{<} - \pi \operatorname{ctg} \pi \mu_0(E_x) - \frac{1}{2Zr_{<}} + \psi(r) + \sum_{k=1}^{\infty} \frac{(2Zr_{<})^k}{k(k+1)!} \right\}.$$

Here $\psi(a)$ is the logarithmic derivative of the Γ function, $\mu_l(E)$ is the quantum defect determined from the experimental spectrum of the atom,^[15] and $r_{<}$ is the smaller of the quantities r and r' .

Generally speaking, in actual calculations we can use as the initial- and final-state wave functions in T , functions obtained by other methods, for example, from Hartree-Fock calculations. Although in this case the method ceases to be self-consistent, in a number of

cases this substitution can be physically justified. This is valid, in particular, for multiphoton transitions from S states to a state with higher orbital angular momentum l . In this case the states with $l \geq 1$ are satisfactorily described in the QDM and the S wave can be improved from other calculations.

Numerical values of $\sigma_N^{(l)}$ and $\sigma_N^{(c)}$ are given in Table 1. Also shown are the angular distribution coefficients for photoelectrons for $N=2$ and 3:

$$dW_N^{(l)}/d\Omega = F^N (a^{(N)} + b^{(N)} \cos^2 \beta + c^{(N)} \cos^4 \beta),$$

where β is the angle between the wave vector of the photoelectron and the polarization vector. For circular polarization

$$\frac{dW_N^{(c)}}{d\Omega} = \frac{(2N-1)!!}{6\pi} W_N^{(c)} (\sin \theta)^{2N},$$

θ is the angle between the electron and photon wave vectors. For $N=2$ our results agree satisfactorily with those of Mizuno^[7] except for σ_2 for potassium at the frequency ω_{2R} , where the difference amounts to ~ 2 . This is due to the existence of a deep valley in the frequency dependence of σ_2 at $\omega \sim \omega_{2R}$ and to the difference in the numerical values $\omega_{2R} = 28796 \text{ cm}^{-1}$ from Ref. 7 and $\omega_{2R} = 28804 \text{ cm}^{-1}$ used in Table 1. The experimental measurement of $W_2 = \sigma_2 F^2$ would be extremely interesting, since in this case the exponent of the power dependence of W_2 on intensity may be different from 2 for a sufficiently narrow spectral width of the radiation line.

For calculation of σ_4 of potassium and rubidium atoms we used the Hartree-Fock wave functions of the ground states, improved by the pseudopotential method.^[16] Comparison with the calculations in the QDM:

$$\begin{aligned} \text{QDM: } K \rightarrow \sigma_i^{(l)} &= 4.38 \cdot 10^{-107}; & \text{Rb} \rightarrow \sigma_i^{(l)} &= 1.32 \cdot 10^{-107}; \\ \text{H-F: } K \rightarrow \sigma_i^{(l)} &= 1.38 \cdot 10^{-108}; & \text{Rb} \rightarrow \sigma_i^{(l)} &= 8.86 \cdot 10^{-108}, \end{aligned}$$

shows that the value of the cross section for multiphoton ionization of potassium is sensitive to the choice of the initial-state wave function. For the single-photon ionization cross section a similar sensitivity has been noted by Seaton.^[17]

4. EXPERIMENTAL INVESTIGATION OF POLARIZATION EFFECT IN MULTIPHOTON IONIZATION OF POTASSIUM AND SODIUM ATOMS

In the experiment we measured the ratios of the cross sections of two- and four-photon ionization of the potassium atom and of three- and five-photon ionization of the sodium atom in linearly and circularly polarized fields with frequencies corresponding to the first and second harmonics of neodymium laser radiation.^[18]

A beam of neutral atoms of sodium or potassium ($n \sim 10^9 \text{ cm}^{-3}$) was intersected by the light beam. We measured the ratio of amplitudes of the ion signals obtained as the result of multiphoton ionization of potassium and sodium atoms in linearly and circularly polarized fields for identical radiation energies. For space-time characteristics of the radiation identical in the two cases,

this ratio is equal to the ratio of the cross sections for multiphoton ionization.

The neodymium laser operated in the axial modes. To obtain linear polarization we placed in the beam beyond the laser an Archard-Taylor prism. The second harmonic was obtained by means of a KDP crystal (the transformation $oe-e$). The polarization of the laser radiation was varied by means of a quartz quarter-wave plate of special orientation with which the linear polarization could be converted to circular. The quartz plate was placed in the radiation beam directly in front of the chamber in which interaction of the radiation with the potassium or sodium atoms occurred.

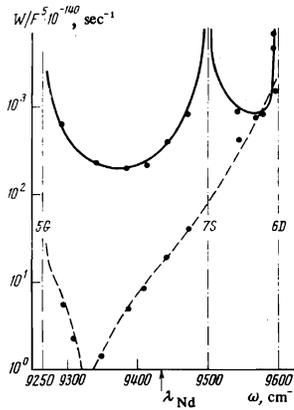
The characteristics of the laser radiation and the numbers of ionized atoms were measured by means of the standard apparatus usually used in experiments on multiphoton ionization of atoms (see, for example, Ref. 19). The laser mode was carefully stabilized. In the experiment we simultaneously measured the amplitudes of the ion signal, the energy and space-time distribution of the radiation in the laser pulse, and the frequency and nature of the field polarization, which enabled us to avoid cases of appreciable change of mode.

To reduce the effect of instability of the space-time distribution of the radiation during the experiment we carried out alternately a measurement of the ion signals for two orientations of the quartz quarter-wave plate corresponding to positions at which the radiation polarization was either linear or circular.

The results of the experiment are given in Table 2. The error arising in calculation of the absolute cross sections does not enter into the cross-section ratio in our experiment, and this enables us to measure the latter with high accuracy, determined in the present case only by the repeatability of the space-time distribution of the radiation from pulse to pulse. The errors in the cross-section ratio are the mean square errors of the series of measurements, multiplied, because of the small number of measurements (~ 10), by a confidence factor of ~ 3 .

5. COMPARISON OF THEORETICAL AND EXPERIMENTAL RESULTS

Comparison of the theoretical calculations of the ratio of the cross sections of alkali metal atoms (see Table 1) with the experimental results will be made on the basis



The cross sections $\sigma_5 = W_5 / F^5$ for five-photon ionization of the sodium atom as a function of the radiation frequency in a linearly polarized field (solid line) and a circularly polarized field (dashed line). The arrow shows the frequency corresponding to the neodymium laser frequency.

of the data of the experiment just described and the results of other studies published up to the present time. [10-12,20]

We note that for the case $\gamma \gg 1$ in comparison of the theoretical and experimental values of the cross-section ratios it is of no significance that the theoretical calculation was carried out for monochromatic radiation while the experiment in a number of cases used a multi-mode laser.

In Table 2 we have given the ratios of the cross sections for multiphoton ionization of atoms in linearly and circularly polarized fields, measured experimentally and calculated for these cases in the present work. As can be seen from the table, good agreement of the experimental and theoretical results is observed for the cross-section ratios within the experimental error except for the case five-photon ionization of the sodium atom by neodymium laser radiation. In this case the discrepancy may result from the fact that in a circularly polarized field near the neodymium laser frequency the multiphoton ionization cross section goes to zero (see the figure). The accuracy of the theoretical calculation of the cross sections near such frequencies is reduced. As a result the reliability of the theoretical value of the cross-section ratio in linearly and circularly polarized fields is also reduced. For a final conclusion regarding the cause of the discrepancy between the theoretical and experimental results in this case, of course, additional measurements at several frequencies in the region indicated above are necessary. Nevertheless, the good agreement of the theoretical numbers with the experimental values in all other cases listed in Table 2 permits us to conclude that nonstationary perturbation theory satisfactorily describes the polarization dependence of the direct multiphoton ionization of an atom, at least for field strengths $E \leq 5 \times 10^6$ V/cm.

The authors are grateful to N. B. Delone for interest in this work and for valuable remarks.

TABLE 2.

Atom	N	$n\omega$, eV	$\sigma^{(l)}/\sigma^{(c)}$	
			Experiment	Theory *
Na	3	2.36	$2.4 \pm 20\%$ *	3
	3	1.78	$0.43 \pm 4\%$ [10]	0.4
	5	1.18	$2 \pm 20\%$ *	14
K	2	2.36	$0.83 \pm 30\%$ *	0.86
	3	1.78	$0.38 \pm 4\%$ [10]	0.4
	4	1.18	$0.6 \pm 30\%$ *	0.25
Cs	2	3.56	$0.78 \pm 16\%$ [11]	0.92
	3	1.78	$0.45 \pm 6\%$ [10]	0.7
	3	1.78	$0.47 \pm 18\%$ [11]	
Rb	4	1.18	0.25 [20]	0.36
	3	1.78	$0.46 \pm 6\%$ [10]	0.43

*Results of the present work.

¹The symbol σ_n is used to designate the cross section for multiphoton ionization of an atom.

- ¹L. V. Keldysh, Zh. Éksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys. JETP 20, 1307 (1965)].
- ²B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Éksp. Teor. Fiz. 61, 968 (1971) [Sov. Phys. JETP 34, 515 (1972)].
- ³P. Lambropoulos, Phys. Rev. Lett. 28, 585 (1972); 29, 453 (1972).
- ⁴S. Klarsfeld and A. Maquet, Phys. Rev. Lett. 29, 79 (1972).
- ⁵H. B. Bebb, Phys. Rev. 149, 25 (1966); 153, 23 (1967).
- ⁶V. Morton, Proc. Phys. Soc. 92, 301 (1967).
- ⁷J. Mizuno, J. Phys. B6, 314 (1973).
- ⁸B. J. Choudhury and R. P. Gupta, Phys. Lett. 50A, 377 (1974).
- ⁹A. Dalgarno and J. T. Lewis, Proc. Roy. Soc. A223, 70 (1955).
- ¹⁰M. R. Cervenán and N. R. Isenor, Opt. Comm. 10, 280 (1974). N. R. Isenor, II Conference on the Interactions of Electrons with Strong Electromagnetic Fields (1975), p. 2-37, Budapest.

- ¹¹R. M. Kogan, F. A. Fox, G. T. Burnham, and E. J. Robinson, Bull. Am. Phys. Soc. 16, 1411 (1971).
- ¹²R. A. Fox, R. M. Kogan, and E. J. Robinson, Phys. Rev. Lett. 26, 1416 (1971).
- ¹³Y. Gontier and M. Trahin, Phys. Rev. 7A, 2069 (1973).
- ¹⁴A. M. Perelomov, V. S. Popov, and M. V. Tenen'ev, Zh. Éksp. Teor. Phys. 50, 1393 (1966) [Sov. Phys. JETP 23, 924 (1966)].
- ¹⁵V. A. Davydkin, B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Éksp. Teor. Fiz. 60, 124 (1971) [Sov. Phys. JETP 33, 70 (1971)].
- ¹⁶L. Szasz and C. McGinn, J. Chem. Phys. 47, 3495 (1967).
- ¹⁷A. Burgess and M. J. Seaton, Mont. Not. Roy. Astr. Soc. 120, 121 (1960).
- ¹⁸G. A. Delone, in *Kratkie soobshcheniya po fizike* (Brief Communications in Physics) 8, FIAN, 1975, p. 28.
- ¹⁹G. A. Delone, N. B. Delone, and G. K. Piskova, Zh. Éksp. Teor. Fiz. 62, 1272 (1972) [Sov. Phys. JETP 35, 672 (1972)].
- ²⁰T. U. Arslanbekov, V. A. Grinchuk, G. A. Delone, and K. B. Petrosyan, in *Kratkie soobshcheniya po fizike* (Brief Communications in Physics) 10, FIAN, 1975, p. 33.

Translated by Clark S. Robinson

Excitation mechanisms in the systems $\text{Na}^+ + \text{Ne}$ and $\text{O}^+ + \text{Ne}$

E. E. Nikitin, M. Ya. Ovchinnikova, and A. I. Shushin

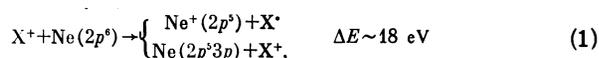
Institute of Chemical Physics, USSR Academy of Sciences
(Submitted November 5, 1975)
Zh. Eksp. Teor. Fiz. 70, 1243-1252 (April 1976)

On the assumption of definite molecular excitation mechanisms, a calculation has been made of the relative intensities and polarizations of radiation from the multiplets $\text{Ne}^*(2p^5 3p)$ and $\text{X}^*(3p)$ for $\text{X} = \text{Na}$ and O , excited in collisions of X^+ with neon at high incident-ion energies ($E \gtrsim 10$ keV). The mechanism assumed excitation to the lowest Σ term (with the least spin) of the X^+Ne^+ ionic cores with possible σ or π molecular states of the excited electron. The calculation enables us to understand a number of correlations of experimental data on polarization. Comparison of the theoretical intensity distribution with experimental values shows that excitation of the $3p$ states of X and Ne occurs both through σ and through π terms of the outer electron, but the oscillatory structure of the total cross sections is due to interference of π states.

PACS numbers: 34.50.Hc, 31.50.+w

INTRODUCTION

Measurements made by various workers^[1-6] of the excitation function of optical lines in the systems Na^+Ne and others and recent systematic studies of the excitation processes



for $\text{X}^+ = \text{Na}^+$, O^+ , Mg^+ , and N^+ have shown^[7] a regular oscillating structure of the cross sections due to interference of energetically close channels of direct excitation and charge exchange,^[3,8] and also a strong polarization of the excited lines. In addition to the phase of the oscillations, which characterizes the relative arrangement of the molecular terms taking part in the interference,^[3,8] measurements have also been made of the distribution of intensities of the various components of the $3p-3s$ transitions of $\text{Ne}^*(2p^5 3p)$ and $\text{X}^*(3p)$ and

their polarization. On the basis of these data, possible molecular excitation mechanisms have been discussed.

In the present work, in order to clarify the excitation mechanism at higher collision energies ($E \gtrsim 10$ keV), we have calculated the relative intensities and polarizations of the components of $3p-3s$ transitions of Ne^* and X^* in reaction (1) for $\text{X}^+ = \text{Na}^+(^1S)$ and $\text{O}^+(^4S)$. Only at such high energies, when only the main components of the molecular interactions can have a substantial influence on the evolution of the excited states arising, is it possible to calculate with relative simplicity the probabilities of population of individual components of the multiplets in such multilevel systems as Ne^+Na^+ and Ne^+O^+ . Calculations carried out for two possible mechanisms (σ and π) permit an understanding of a number of correlations of the experimental data and enable us to conclude what is the role of each of the excitation mechanisms at high energies.