

Negative ion in a strong optical field

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A review is given of theoretical descriptions of the nonlinear interaction of negative ions with a strong optical field. A brief account is provided of the main features of the structure of negative ions. A detailed analysis is made of the theoretical methods for describing nonlinear processes in negative ions on the basis of the one-electron approximation. The ranges of validity of the reviewed methods are given. Special attention is paid to the role of the electron–electron correlation interaction. The importance of the correlation interaction in weakly nonlinear processes is stressed. The review concludes with the formulation of a number of theoretical and experimental tasks for future investigations of nonlinear processes in negative ions.

1. INTRODUCTION

The rapid development of lasers has made possible practical investigations of the nonlinear interaction of atomic systems with electromagnetic radiation in a wide range of frequencies. Among the results of such interaction are nonlinear light scattering and multiphoton ionization.

Extensive theoretical and experimental investigations have been made of the nonlinear interaction of atoms with a strong optical field.^{1–3} The present review deals with nonlinear processes in negative ions which have been investigated much less thoroughly.

The energy of the affinity of an atom to an electron is always less than the first ionization potential of the atom. Consequently, in the case of laser radiation of definite frequency the process of nonlinear photodecay of a negative ion is always of lower order of nonlinearity than the process of ionization of an atom. Table I gives the orders of nonlinearity of the photodecay of various negative ions at the radiation frequencies of different lasers. The affinity energies listed in Table I are taken from Ref. 4.

We shall now consider briefly the characteristic features of the structure of negative ions. The fundamental difference between negative ions and atoms is the structure of the outer electron shell, which may contain an s, p, or d weakly bound electron. The differences between a negative ion and an atom become less as we go to deeper (inner) shells. Therefore, we shall consider both negative ions and atoms together, unless this will interfere with the integrity of our presentation. We shall always use the atomic system of units characterized by $m = e = \hbar = 1$.

The actual binding of an electron attached to a negative ion is primarily due to the electron–electron correlation interaction.⁵ This interaction, which governs that part of the electron–electron interaction which is ignored in the static Hartree–Fock approximation, is very important in the description of the structure of negative ions. It is convenient to mention here that all the atoms for which the energy of the affinity to an electron is positive have a partly filled outer shell (so that negative ions of rare gases do not form bound states). Consequently, an attached electron is always located in an open subshell of an atom. We shall not consider how the electron density in the outer subshell of an atom becomes redistributed in the process of formation of a negative ion. However, it is always true that the correlation interaction in negative ions is governed not only by the interaction of elec-

trons from different subshells, but also by the interaction of electrons from the same subshell. The latter is very important, because the overlap of the electron clouds formed by electrons from the same subshell is the strongest effect.

The correlation interaction which governs the nature of the attraction of an electron to an atom is of short-range nature and the potential of this interaction has the following asymptotic form in the limit $r \rightarrow \infty$ (Ref. 6):

$$U(r) \propto -\frac{\alpha}{r^4}, \quad (1)$$

The short-range nature of the interaction has the effect that practically all the states (with the possible exception of the first few excited states) form a continuous spectrum which in some cases is only slightly distorted by the interaction of an electron with an atom. The short-range nature of the interaction makes it possible to use the method of the zero-range potential (model δ potential), popular in atomic physics, in the description of the structure of negative ions with an outer s electron relying on the one-electron approximation.^{7,8} The wave function of a bound s state considered using the δ potential can be represented in the form

$$\psi(r) = \frac{B}{r} e^{-\chi r}, \quad (2)$$

where $E_0 = -\chi^2/2$ is the binding energy and B is the normalization coefficient. An allowance for the interaction with the δ potential is then equivalent to introduction of the following boundary condition:

TABLE I. Orders k of nonlinearity of photodecay of different negative ions at radiation frequencies of different lasers (S is the electron affinity energy taken from Ref. 4).

Negative ion	S , eV	k		
		ruby laser	neodymium laser	CO ₂ laser
Cs ⁻	0.47	1	1	4
Rb ⁻	0.49	1	1	5
K ⁻	0.50	1	1	5
Na ⁻	0.54	1	1	5
Li ⁻	0.61	1	1	6
C ⁻	1.26	1	2	11
Si ⁻	1.39	1	2	12
O ⁻	1.46	1	2	13
S ⁻	2.08	2	2	18
I ⁻	3.06	2	3	26
Br ⁻	3.36	2	3	28
F ⁻	3.45	2	3	29
Cl ⁻	3.61	3	4	31

$$\left. \frac{d \ln(r\psi(r))}{dr} \right|_{r=0} = -\kappa. \quad (3)$$

The short-range potential model is very convenient, as shown below, in studies of the general nature of the interaction of negative ions with an electromagnetic field and it makes it possible to describe this interaction analytically. However, a more thorough description of this interaction process must allow fully for the characteristic features of the structure of a specific negative ion. Moreover, as shown in Sec. 4, in a description of the interaction of an atomic system with an electromagnetic field the dynamic correlation interaction may also be important and this interaction cannot possibly be described by the one-particle model.

It follows from this discussion that the main difficulty in calculations of the binding energy and in the construction of the wave functions of negative-ion electrons is a rigorous allowance for the correlation interaction. In the case under discussion it is more important than in calculations of the various processes in atoms. For example, the static one-configuration Hartree-Fock approximation, based on the approximation of independent particles which can describe the main parameters of the structure of atoms, fails to provide even the correct sign of the energy of the affinity of an electron to an atom.⁵ One of the most successful one-electron approximations to an allowance for the electron-electron correlation interaction is the method of the functional of the local electron density. The importance of this method is that the energy of a system of N electrons exposed to an external field $v(\mathbf{r})$ is a single-valued functional of the electron density $\rho(\mathbf{r})$ (Ref. 9):

$$E_v[\rho(\mathbf{r})] = T_s[\rho(\mathbf{r})] + \int v(\mathbf{r})\rho(\mathbf{r})d\mathbf{r} + \frac{1}{2} \int \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}d\mathbf{r}' + E_{xc}[\rho(\mathbf{r})], \quad (4)$$

where the first term represents the kinetic energy of the non-interacting electron gas in its ground state and the last term allows for the exchange-correlation interaction. In the case of systems with a slowly varying density we can use the local density approximation:

$$E_{xc}[\rho] = \int \varepsilon_{xc}(\rho(\mathbf{r}))\rho(\mathbf{r})d\mathbf{r}, \quad (5)$$

where $\varepsilon_{xc}(\rho)$ is the local exchange-correlation energy per particle in a homogeneous electron gas of density $\rho(\mathbf{r})$.

The method of the functional of the local electron density is convenient in practical applications because of the local nature of $\varepsilon_{xc}(\rho)$ and, consequently, of the electron potential. The results of the application of the theory of the local electron density functional to the description of the various negative ions allowing for the correction representing the self-interaction can be found in Refs. 10 and 11.

A more self-consistent allowance for the correlations based on the many-electron approach, which includes the angular correlations, is ensured by the method of superposition of configurations⁵ and also by calculations based on the use of the hyperspherical basis of the wave functions.¹² However, calculations of the parameters of multiphoton processes in negative ions carried out using these methods are very difficult technically, are hardly ever used, and require further development.

We have summarized above the main methods for the description of the structure of negative ions. Each of these methods is or can be the basis for the development of a particular description of the processes of the interaction of a negative ion with an external electromagnetic field. We shall now consider these descriptions.

The main tasks of the present review include the following: determine the range of validity of the secular conversion of perturbation theory in calculations of the nonlinear characteristics of the interaction of negative ions with an electromagnetic field; identify the role of selection of the one-electron potential of the interaction of an outer electron with the core in describing the interaction of negative ions with the field considered in the one-electron approximation; identify other one-electron methods capable of providing a description of nonlinear processes in negative ions in the case of a strong field, and also in other situations in which the perturbation theory method is invalid; identify the role of the dynamic correlation interaction in the multiphoton processes in negative ions.

The present review is structured as follows. Section 2 is devoted to multiphoton processes and calculations of the characteristics of negative ions by perturbation theory methods. The validity of perturbation theory in the case of multiphoton processes in negative ions is considered and an analysis is made of calculations of multiphoton matrix elements carried out using Green's functions. The results are given of calculations of the dynamic polarizability and the cross sections representing multiphoton detachment of electrons from negative ions. The results of calculations of the multiphoton detachment cross sections are compared with experimental data. Section 3 deals with strong fields when perturbation theory methods can no longer be used. An analysis is made of the rigorous solution for the δ potential, of the semi-classical approximation, of the method of complex rotation of the coordinates, and of the direct computer modeling of the detachment of an electron from a one-dimensional short-range potential. The influence of the correlation effects on multiphoton processes in negative ions are considered in Sec. 4. A perturbation theory is formulated in terms of the external field and the correlation interaction, and the results of calculations allowing for the correlations based on perturbation theory are given. Generalization of the random phase approximation to the multiphoton case is presented. A polarization potential is introduced in terms of the quasienergy approach in the case of strong fields in which the perturbation theory method may no longer be applied to the external field. The polarization effects in a strong field are analyzed in different frequency ranges and for different field intensities. The Conclusions summarize the results of investigations of multiphoton processes in negative ions and consider future trends.

2. PERTURBATION THEORY IN THE INDEPENDENT PARTICLE APPROXIMATION

2.1. Main relationships in perturbation theory

A special feature of atomic systems compared with other many-electron systems is the action of an external central Coulomb field of the nucleus on electrons. The associated central symmetry is exact for one-electron systems and only approximate for many-electron systems. Therefore, in the

case of many-electron systems we can introduce the central field approximation in which the electrons are moving. Electrons are grouped in various shells and they experience different average fields. The field of an optical wave can be regarded as weak if it is less than the characteristic internal fields corresponding to a given shell. Clearly, a field which is strong for the outer electrons can be weak for the inner electrons.

We shall consider the action of a laser radiation field on such many-electron systems as negative ions. We shall be mainly interested in the processes of the interaction of radiation with electrons in the outer shell of an ion. The characteristic intensity of the internal field attracting an external electron to an atom is governed by the relationship $\mathcal{E}_0 = (2|E_0|)^{3/2}$, where E_0 is the energy of binding of an electron to an ion. If E_0 is a concrete energy (Table I), the field intensity \mathcal{E}_0 is of the order of 10^7 – 10^8 V/cm and it is important to stress that this is on the average two orders of magnitude less than the characteristic intensity of the intraatomic field. It should also be mentioned that such field intensities are readily realizable using commercially available serially produced laser systems.

In this section we shall consider the descriptions of the interaction of negative ions with an external field by the time-dependent perturbation theory method used in the central field approximation. This approximation is based on construction of a one-particle centrally symmetric potential in which the eigenvalues of the energy and the corresponding wave functions reproduce the binding energies and distributions of electron densities in the various electron shells. However, the fact that the one-electron approximation can be used to describe the structure of negative ions does not mean that this is a satisfactory approach to multiphoton processes. The one-electron approximation can be used correctly if at least the perturbation by an external field of all the other electrons (apart from the one being ionized) is weak. However, we shall show in Sec. 1 that there is always more than one electron in the outer subshell of a negative ion. Therefore, an external field influences equally an electron being ionized and the other electrons in the outer subshell. This gives rise to a dynamic correlation interaction. In the present section we shall consider the simplified problem and ignore at this stage the correlation effects; we shall return to these effects in Sec. 4.

The criterion of validity of perturbation theory in finding the probability of bound–free transitions is the inequality¹

$$d\vec{\mathcal{E}} \ll (E_n - E_0 \pm n\omega), \quad \omega, \quad (6)$$

where $\vec{\mathcal{E}}$ is the laser field intensity, d is the dipole moment of an electron, E_n is the energy of an excited state of this electron, ω is the laser radiation frequency, and n is the number of photons which ranges from unity to a value smaller than the number of photons necessary in the ionization process. Since in the case of negative ions the excited bound states are either absent or close to the continuous spectrum limit, they satisfy $E_n \gtrsim 0$. Therefore, in the case of negative ions in the absence of resonances only one inequality remains from Eq. (6):

$$d\vec{\mathcal{E}} \ll \omega. \quad (7)$$

This condition implies weakness of the interaction com-

pared with the energy of an electromagnetic radiation photon. It should be pointed out that in the description of the parameters of bound–bound transitions, such as the dynamic polarizability, the perturbation theory approach is valid provided just the first, weaker, of the inequalities in Eq. (6) is satisfied.

The condition (7) can be rewritten in terms of the binding energy of an electron E_0 and this can be done using an estimate for the dipole moment of an electron $d \sim 1/\kappa$. There, Eq. (7) is equivalent to

$$\gamma = \frac{\kappa\omega}{\mathcal{E}} \gg 1, \quad (8)$$

where γ is the familiar adiabatic parameter.¹³ The condition $\gamma \gg 1$ implies the multiphoton (and not tunnel) nature of the nonlinear interaction. The condition for validity of the perturbation theory of Eq. (8) leads directly to the condition of smallness of the intensity of the electromagnetic field compared with a certain critical value $\mathcal{E}_{cr} = \kappa\omega$. In the case of the one-photon interaction process the field \mathcal{E}_{cr} is equal to the intensity of the characteristic interaction field \mathcal{E}_0 . An increase in the order of nonlinearity reduces \mathcal{E}_{cr} . Therefore, the perturbation theory approach is valid only if $\mathcal{E} \ll \mathcal{E}_0$.

In the limiting tunnel case characterized by

$$\gamma \ll 1 \quad (9)$$

the interaction with the external field, expressed in terms of the energy, is much greater than the laser radiation photon. Hence, it is clear that the nature of the process should be qualitatively different: it should be semiclassical. If $\gamma \ll 1$, this case implies the possibility of a tunnel detachment of an electron. On the other hand, the condition (9) may be valid also if $\mathcal{E} \gtrsim \mathcal{E}_0$.

Up to now a theoretical description of the interaction of atoms and negative ions with the field in excess of the atomic value has not been developed much and we shall not discuss this case in detail. In the case of moderate external field intensities when the parameter satisfies $\gamma \ll 1$ and the perturbation theory approach is invalid, the process of nonlinear decay of a negative ion can be described as reported in the next section.

We shall also bear in mind that the perturbation theory approach is valid if the Stark shifts of the ground-state energy and of the threshold of the continuous spectrum are small compared with the frequency ω . This also imposes restrictions on the external field intensity. Thus, the limitations imposed on the Stark shift of the ground state require that the condition $\alpha\mathcal{E}^2/4 \ll \omega$, be satisfied, where α is the dynamic polarizability of a negative ion. If we use the estimate $\alpha \propto \kappa^{-3}$, we find the condition is $\mathcal{E} \ll (\omega\kappa^3)^{1/2}$. Allowing for realistic binding energies of electrons in negative ions, which are always less than the atomic energies, we can readily show that for any order of the nonlinearity of the interaction of a negative ion with radiation this criterion is less stringent than the condition (8).

The Stark shift of the limit of the continuous spectrum is $\mathcal{E}^2/4\omega^2$. The requirement of its smallness compared with ω leads to the condition

$$\eta = \frac{\mathcal{E}^2}{\omega^3} \ll 1. \quad (10)$$

In the case of stronger nonlinearities this condition imposes a more rigid restriction on the external field intensity and the

criterion of Eq. (8) sets fully the limits of validity of the perturbation theory approach only in the case of weak nonlinearities.

We shall point out one more circumstance associated with the above-threshold absorption of photons by a detached electron. If the field is sufficiently strong, this process is significant and the perturbation theory approach becomes invalid when describing the photodecay of a negative ion for this reason. The above-threshold absorption is discussed in detail in the next section. We should point out that, as shown in that section, the restriction imposed on the external field intensity due to the ability to neglect the above-threshold absorption and to use the perturbation theory approach in the description of the nonlinear photodecay of negative ions is identical with the restriction (10) associated with the smallness of the Stark shift of the limit of the continuous spectrum compared with ω .

The last factor which governs the validity of the perturbation theory approach is the ability to achieve saturation of the photodecay in the case of fairly long duration τ of the radiation pulses, i.e., the condition $W\tau \ll 1$ should be satisfied; here, W is the probability of photodecay per unit time.

It therefore follows that when the condition (10) is satisfied, we can use the perturbation theory approach in the one-electron approximation in the case of all atoms with the exception of hydrogen, and this provides a more or less reasonable description of the nonlinear processes that occur in atoms of alkali elements and negative ions. The interaction with the external field is described by

$$V(t) = V^{(+)}e^{-i\hat{\omega}t} + V^{(-)}e^{i\omega t},$$

$$V^{(+)} = V^{(-)*}, \quad (11)$$

where the operator $V^{(+)}$ represents the absorption and $V^{(-)}$ the emission of photons. The operators $V^{(+)}$ can be selected using various gauges: in the form of the momentum and in the form of the length. It is shown in Ref. 14 that when the full basis of the wave eigenfunctions of the static Hamiltonian is used, the results of calculations of multiphoton matrix elements carried out using perturbation theory are independent of the gauge. Therefore, in all cases except those mentioned specially, we shall use the dipole approximation for $V^{(\pm)}$ and in this approximation we have

$$V^{(+)} = -\frac{d\mathcal{E}}{2}. \quad (12)$$

The dipole approximation for the description of multiphoton processes is justified throughout the investigated optical frequency range,¹ where $c/\omega \gg r_a$, i.e., when the wavelength is greater than the dimensions of the system r_a . In the perturbation theory framework the probability of an n -photon detachment per unit time can be calculated using the Fermi golden rule:

$$W^{(n)} = \frac{\pi}{2^{2n-1}} |z_{fi}^{(n)}|^2 \mathcal{E}^{2n} \rho_f, \quad (13)$$

where $z_{fi}^{(n)}$ is the n -photon matrix element and ρ_f is the density of the final states. Therefore, the dependence of the probability of multiphoton detachment of an electron on the photon flux $F = \mathcal{E}^2/8\pi\alpha\omega$ (α is the fine structure constant) is described by the power law

$$W^{(n)} = \sigma^{(n)} F^n. \quad (14)$$

The coefficient of proportionality $\sigma^{(n)}$ acts as the multiphoton decay cross section and is frequently used to represent the decay.

2.2. Multiphoton matrix elements in the one-electron approximation

The wave functions of the initial $|i\rangle$ and final $|f\rangle$ states considered in the one-electron approximation are the eigenfunctions of the static Hamiltonian

$$H_0 = -\frac{1}{2}\nabla^2 + U(r), \quad (15)$$

where $U(r)$ is the central potential of the interaction of an electron with the core.

The matrix element $z_{fi}^{(n)}$ can be calculated using the one-electron Green's function:¹

$$z_{fi}^{(n)} = \langle f | z G_{E_1+(n-1)\omega} z \times \dots \times G_{E_1+\omega} z | i \rangle. \quad (16)$$

The Green's function satisfies the static Schrödinger equation with a δ -function inhomogeneity:

$$(H_0 - E) G_E(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'). \quad (17)$$

We can readily write down the matrix elements also for the nonlinear susceptibilities. For example, the dynamic polarizability of an isolated level is described by¹⁵

$$\alpha(\omega) = \langle i | z (G_{E_1+\omega} + G_{E_1-\omega}) z | i \rangle. \quad (18)$$

In general, the multiphoton matrix elements considered in the one-electron approximation can be expressed, after separation of the angular parts, in terms of the radial Green's functions $g_l(E, r, r')$ with the energy E and the orbital momentum l . The radial Green's functions are the solutions of the Schrödinger equation

$$\left(\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} - 2U(r) + 2E \right) g_l(E, r, r') = -\delta(r - r'). \quad (19)$$

The asymptotic form of the Green's function is selected so that decaying states are obtained for $E > 0$, i.e., the Green's function has the asymptotic behavior of a diverging wave.

In the case of the Coulomb potential and Coulomb-like model potentials the Green's function can be obtained analytically: it can be expressed in terms of the Whittaker functions.²

In the case of an electron bound weakly to a negative ion the Green's function can be constructed as follows:⁷

$$g_l(E, r, r') = \chi_{kl}^{(+)}(r_>) \chi_{kl}(r_<), \quad k = (2|E|)^{1/2}, \quad (20)$$

where $\chi_{kl}(r)$ is the regular and $\chi_{kl}^{(+)}(r)$ is the irregular solution of the radial Schrödinger equation, $r_>$ ($r_<$) is the larger (smaller) of the quantities r and r' . The solutions $\chi_{kl}^{(+)}$ and χ_{kl} are normalized by the following condition applying to the Wronskian:

$$\chi_{kl}^{(+)\prime}(r) \chi_{kl}(r) - \chi_{kl}^{(+)}(r) \chi_{kl}'(r) = 1. \quad (21)$$

These solutions can be found numerically. A widely used numerical method is the two-point Numerov technique of the fifth order of precision.¹⁶ The regular solution is found by using the behavior at zero:

$$\chi_{kl} \sim r^{l+1}, \quad (22)$$

whereas the irregular solution is obtained employing the semiclassical asymptote given by

$$\begin{aligned} \operatorname{Re} \chi_{kl}^{(+)}(r) &\sim \frac{1}{(\rho(r))^{1/2}} \cos \left(\int_{r_1}^r \rho(r) dr + \varphi \right), \quad E > 0, \\ \operatorname{Im} \chi_{kl}^{(+)}(r) = \chi_{kl}(r) &\sim \frac{1}{(\rho(r))^{1/2}} \sin \left(\int_{r_1}^r \rho(r) dr + \varphi \right), \quad E > 0, \end{aligned} \quad (23)$$

where

$$\rho(r) = \left[2 \left\{ E - U(r) - \frac{[l + (1/2)]^2}{2r^2} \right\} \right]^{1/2}$$

and

$$\chi_{kl}^{(+)}(r) \sim e^{-kr}, \quad E < 0. \quad (24)$$

The solution of the homogeneous equation $\chi(r)$ ($E > 0$) which is regular at zero allows us to find also the scattering phase δ_l .

The perturbation theory methods for the interaction with the laser radiation field have been used successfully in describing nonlinear processes in alkali metals with a clearly distinguished single optical electron. Extensive use has also been made of the construction of the Green's functions and calculations of the matrix elements, as well as of the model potential and the quantum defect methods. These methods and the results obtained are discussed in detail in Refs. 2 and 17.

2.3. Dynamic polarizability of negative ions

We shall consider the dynamic polarizability of the simple negative ion H^- with the electron binding energy $E_0 = -0.75$ eV.

The wave function of a weakly bound electron can be described by

$$\begin{aligned} \psi_0 &= \frac{B}{r} \left(\frac{\kappa}{2\pi} \right)^{1/2} (e^{-\kappa r} - e^{-\beta r}), \\ B &= \frac{[\beta(\beta + \kappa)]^{1/2}}{\beta - \kappa}, \\ \beta &= 0.742, \quad \kappa = 0.234, \end{aligned} \quad (25)$$

which is an approximation of the result obtained in the variational method for the H^- ion in Ref. 18. The role of the potential of the interaction of an electron with an atom can be determined by comparing the results of calculations using different potentials:¹⁹

$$\begin{aligned} U_1(r) &= 0, \\ U_2(r) &= -\frac{\alpha'}{2(r+r_c)^4}, \\ U_3(r) &= -\left(1 + \frac{1}{r}\right) e^{-2r} \\ &- \frac{\alpha'}{2r^4} \left\{ 1 - \left[1 + 2r + \frac{(2r)^2}{2} + \frac{(2r)^3}{6} + \frac{(2r)^4}{36} \right] e^{-2r} \right\}. \end{aligned} \quad (26)$$

The potential U_1 represents the complete neglect of the interaction of an electron with an atom in the states of the continuous spectrum of H^- . The potential U_2 corresponds to a phenomenological allowance for the long-range polarization interaction of an electron with an atom: $\alpha' = 9/2$ is the static polarizability of the hydrogen atom and $r_c = 0.74$ is the cutoff radius. The potential U_3 is obtained by an ana-

lytic approximation of the potential of the interaction of an electron with a hydrogen atom obtained using the second order of perturbation theory.

The results of numerical calculations of the polarizability $\alpha(\omega)$ are presented in Figs. 1 and 2. Curves 1–3 correspond to the potentials U_1 , U_2 , and U_3 . Figure 1 gives also the results of Ref. 20 (curve 4) with an "error corridor" represented by dashed curves where the polarizability of H^- for $\omega < |E_0|$ is calculated applying the variational method to the correction to the energy in the second order of perturbation theory.

The results of these calculations demonstrate that the dynamic polarizability of a negative hydrogen ion is not very sensitive to the actual form of the potential of the interaction of an electron with an atom and a fully satisfactory result is obtained if we ignore this potential and use primarily the wave function of the negative H^- ion.

It therefore follows that in the case of the H^- ion the dynamic polarizability can be calculated using the one-electron Green's function derived using the plane-wave approximation. However, this potential must be allowed for in the case of negative ions which have p electrons in the outer electron shell ($l = 1$). In accordance with the dipole selection rules these electrons can go over virtually to the d and s states. The latter states are distorted greatly at distances of the order of the atomic dimensions, which make the dominant contribution to the matrix element. Negative ions with partly filled shells have a nonzero total angular momentum and, consequently, their ground state has a multiplet structure. Nonresonant perturbation of an atomic multiplet by an elliptically polarized laser field can be described by the effective Hamiltonian \tilde{H} allowing for the multiplet splitting and for the "principal" part of the interaction of the multiplet levels with different momenta and projections M with the radiation field.² In the first order of perturbation theory in respect of the spin-orbit interaction and in the second order in respect of the interaction with the wave field the effective Hamiltonian is given by

$$\begin{aligned} \tilde{H} &= \Delta E_J |NJM\rangle \langle NJM| \\ &- \frac{1}{4} \sum_{\mu, \nu, J', M'} e_{\mu}^* e_{\nu} |NJM\rangle \chi_{JM', M'}^{\mu\nu} \langle NJ'M'\rangle, \end{aligned} \quad (27)$$

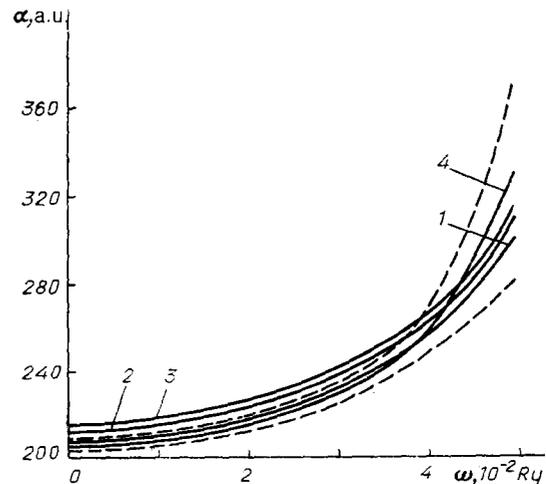


FIG. 1. Polarizability of H^- at field frequencies ω lower than the photodetachment potential.²¹

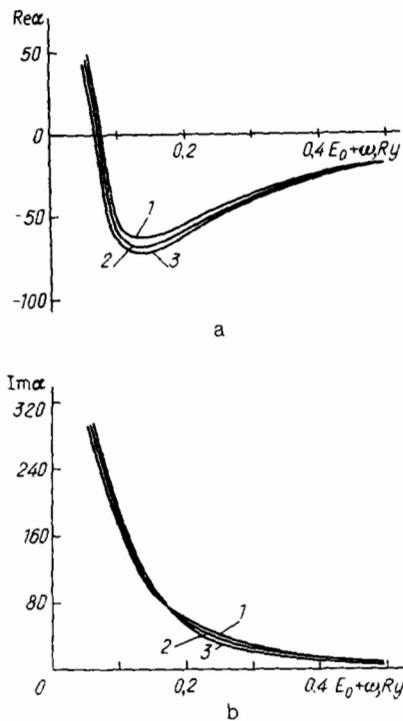


FIG. 2. Real (a) and imaginary (b) parts of the polarizability of H^- at field frequencies ω higher than the photodetachment potential.²¹

where ΔE_j is the multiplet splitting in the absence of the radiation field, $\chi^{\mu\nu}$ is the light scattering tensor, e_μ are the spherical components of the polarization vector, and I is the light intensity.

Diagonalization of the symmetric matrix of the effective Hamiltonian \tilde{H} makes it possible to derive quasistationary states of the system as a linear superposition of the states in the absence of the field and with weightings equal to the components of the eigenvectors of the matrix

$$\Phi^{(r)} = \sum_{J,M} f_{JM}^{(r)} |NJM\rangle. \quad (28)$$

These eigenvalues give the change in the level energies $\Delta E^{(r)}$, where r are the quantum numbers representing a new system of states. If $\omega > |E_{NJ}|$, then the matrix $\tilde{H}_{JM'J'M'}$ is non-Hermitian and the functions $\Phi^{(r)}$ are nonorthogonal, because the states with different values of J and M have common photodecay channels. The real part of $\Delta E^{(r)}$ determines the shift of the level and the imaginary one governs its width. In view of the symmetry of the matrix \tilde{H} the system of $f^{(r)}$ vectors forms a complete orthogonal set:

$$f^{(r)} f^{(r')} = \delta_{rr'}. \quad (29)$$

Therefore, any solution of the Schrödinger equation can be expanded in terms of a new system of functions $\Phi^{(r)}$ and the effective Hamiltonian can be rewritten in the form

$$\tilde{H} = \sum_r |\Phi^{(r)}\rangle \Delta E^{(r)} \langle \Phi^{(r)}|. \quad (30)$$

If the spin-orbit interaction is much stronger or much weaker than the field splitting of the multiplet levels, these two interactions can be considered separately and only the effect of the field can be separated. Then, the shift of the level

is proportional to the radiation intensity:

$$\Delta E^{(r)} = -\frac{\alpha^{(r)}}{4} I, \quad (31)$$

where the dynamic polarizabilities $\alpha^{(r)}$ are the eigenvalues of the matrix

$$\hat{Q} = \sum_{\mu,\nu} e_\mu^* e_\nu \chi_{JM'J'M}^{\mu\nu}.$$

Figure 3 gives the results of calculations of the dynamic polarizability α of the O^- (2P) ion at the ruby laser frequency $\omega = 14\,400\text{ cm}^{-1}$ (Ref. 21) for light with complete elliptic polarization and a unit polarization vector $\mathbf{e} = \mathbf{e}_+ \cos \theta - \mathbf{e}_- \sin \theta$. The parameter θ is related to the degree of polarization by $I = \sin 2\theta$. The symmetry of the problem ensures that if $\theta = 0$ or $\pi/2$, the projection of the momentum along the direction of propagation of an electromagnetic wave has a specific value. If $\theta = \pi/4$, the projection of the momentum along the field polarization is fixed and, in accordance with the Kramers theorem, the levels are doubly degenerate in respect of the sign of the projection. The coefficients in the expansion of the wave function in terms of unperturbed states of a negative ion are identical with elements of the finite rotation matrix $D^J(0, \pi/2, 0)$ governing the change in the wave function due to rotation that makes the z axis of the coordinate system coincide with the field polarization direction. These properties make it possible to find the quantum numbers of the projection of the momentum along the relevant directions. These quantum numbers are plotted in Fig. 3.

In this subsection we have thus considered one of the main characteristics of the interaction of an atomic system with an external field, which is the dynamic polarizability. Negative ions with a low binding energy of an outer electron, such as the ion and negative ions of alkali atoms, are polarized more strongly. For example, in the static limit the value of $\alpha(0)$ for these ions is of the order of 10^2 a.u. The high polarizability of the negative ions with a small binding energy can be explained quite simply. The lower the binding energy of an electron, the further it is located from the nucleus of an atom; the average distance in the distribution of the electron density is then $\langle r \rangle \sim |E_0|^{-1/2}$. On the other hand, the polarizability obeys $\alpha \propto r^3$, so that the polarization is manifested more strongly when the distance between the nucleus and the electron is greater. In the range of radiation frequencies where the polarizability reaches its maximum value, the polarizability of negative ions of the alkali atoms and also of H^- is of the order of 10^3 a.u.

Polarization of an atomic system in an external field implies a change in the initial state of electrons under the influence of an electromagnetic wave. For example, the energy of the initial state changes by $\Delta E = -\alpha \mathcal{E}^2/4$. If the field intensity \mathcal{E} is sufficiently high, the value of ΔE may be quite considerable. Therefore, in the case of strong fields it may be necessary to allow for the change in the initial state in calculation of the various nonlinear characteristics in a strong electromagnetic field.

There is an interesting possibility associated with an opportunity for investigating the shifts of the levels and their widths on the basis of resonances in the scattering of electrons by atoms in the presence of an electric field.^{22,23}

The resonant scattering of an electron by an atom in the presence of a field involves radiation trapping of an electron

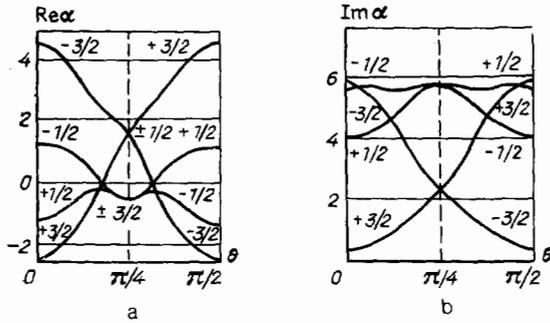


FIG. 3. Real (a) and imaginary (b) parts of the polarizability of $O^- (^2P_{3/2})$ at the ruby laser frequency.²¹

by an atom followed by dissociation of the negative ion. In the case of a resonance accompanied by the emission of m and absorption of n photons the differential resonant scattering cross section is

$$\frac{d\sigma^{(n-m)}}{d\Omega_{p'}} = \frac{1}{4\pi^2} \left| \sum_r \frac{A_{ip',r}^{(n)} A_{r,ip}^{(m)}}{E_{NJ} + \Delta E^{(r)} + n\omega - (1/2)p^2} \right|^2, \quad (32)$$

where $A_{r,ip}^{(m)}$ and $A_{ip',r}^{(n)}$ are, respectively, the amplitudes of the m -photon capture and n -photon detachment of electrons.

The results of calculations of the dynamic polarizabilities²¹ demonstrate that the shifts and widths of resonances are comparable. Consequently, in the case of a weak multiplet splitting they should overlap strongly. If $n = 0$, the process becomes of one-stage nature and it describes the multiphoton capture of an electron by an atom.²³ Its cross section can be found from the principle of detailed balance using the cross sections of multiphoton detachment of electrons.

The dynamic polarizability affects also the bremsstrahlung emitted as a result of the scattering of electrons by negative ions.²⁴ This effect is observed when an external field induces an alternating dipole moment in a negative ion and the scattered electron interacts with this moment. Therefore, in addition to the usual static bremsstrahlung, we can expect additional and comparable polarization bremsstrahlung.²⁴

2.4. Multiphoton detachment of electrons from negative ions. Perturbation theory calculations

As pointed out already, an electron attached to a negative ion in a continuous spectrum interacts relatively weakly with an atom. Therefore, at least in the case of negative atomic ions of hydrogen and possibly also in the case of negative ions of alkali metals the Green's function can be described by the plane-wave approximation.²¹ It was used in Ref. 25 to obtain the two-photon detachment cross section. If we select the interaction of an electron with the laser radiation field in the form of the velocity

$$V^{(+)} = -\frac{1}{2} \frac{\hat{p} \vec{\mathcal{E}}(t)}{\omega}, \quad (33)$$

$$\hat{p} = -i\nabla,$$

we can complete the calculation of the n -photon matrix element. In fact, in view of the orthogonality of the plane waves,

the matrix elements of free-free transitions exhibit δ functions. This makes it possible to calculate the sums over the intermediate states. In the case of the initial s states the probability of n -photon detachment per unit time is obtained as a result of integration over the emission angles:

$$W = \frac{8\pi^2 |\langle k_f | 0 \rangle|^2 (k_f \mathcal{E} / \omega)^{2n}}{[(E_0 + (n-1)\omega - E_f) \times \dots \times (E_0 + \omega - E_f)]^2 (2n+1)}, \quad (34)$$

where $\langle k_f | 0 \rangle$ is the wave function of a weakly bound electron considered in the momentum representation; k_f is the momentum in the final state. In the case of H^- when the wave function is selected in the form of Eq. (25), we find that in momentum space

$$\langle k_f | 0 \rangle = B \frac{\kappa^{1/2}}{\pi} \left(\frac{1}{\beta^2 + k_f^2} - \frac{1}{\kappa^2 + k_f^2} \right). \quad (35)$$

In the case of negative ions with p electrons the plane-wave approximation is not satisfactory. This is demonstrated, in particular, by a comparison of the results of calculations of the cross section for the two-photon detachment of an electron from the negative I^- ion. Figure 4 gives the results of calculations of the cross section obtained in the zero-potential approximation (dashed curve) and also allowing for the interaction of an electron with an atom²⁶ (continuous curve) considered as a function of the energy E_k of the detached photoelectron. This figure gives also the results of a determination of the cross section of two-photon detachment of an electron by ruby laser radiation: $\sigma^{(2)} = (3.5 \pm 1.5) \cdot 10^{-49} \text{ cm}^4 \cdot \text{s}$ (Ref. 27). We can see that the theory agrees well with the experimental results only if we allow for the interaction of an electron with an atom.

The central field approximation was also used in Ref. 28 to calculate the cross section for the two-photon decay of Cl^- and F^- ions and also of Na^- . The results calculated for Cl^- and F^- , are plotted in Fig. 5. The central potential is the analytic approximation of the self-consistent Hartree-Fock potential considered in the local approximation:²⁹

$$U(r) = -\frac{1}{r} \left\{ Z - N_a \left[1 - \frac{1}{H(e^{r/d} - 1) + 1} \right] \right\}, \quad (36)$$

where Z is the charge of a nucleus, N_a is the number of electrons in the core, and the parameters H and d are selected to ensure that the energy of a bound electron in the potential of Eq. (36) is identical with the exact value of the affinity

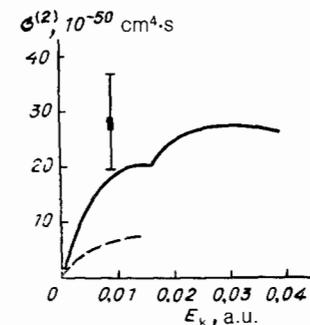


FIG. 4. Dependence of the cross section of two-photon decay of the I^- ion on the kinetic energy E_k of the detached electron. The dashed curve represents the results of calculations carried out in the plane wave approximation,²⁵ whereas the continuous curve represents calculations made allowing for the plane wave scattering.²⁶ The experimental point was taken from Ref. 27.

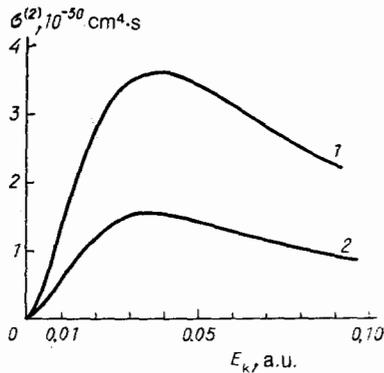


FIG. 5. Dependence of the cross section of two-photon decay of Cl^- (curve 1) and F^- (curve 2) ions on the kinetic energy E_k of the detached electron, obtained in the central field approximation of Eq. (36) (Ref. 28).

of an atom to an electron, whereas the asymptotic behavior of the wave function of the bound state corresponds at large distances to the more rigorous calculations carried out by the Hartree-Fock method. Calculations with the potential in the form of a sum of the atomic potential of the type described by Eq. (36) and of the polarization potential given in Ref. 29 yield results very close to those plotted in Fig. 5 (we shall not give them here). This is due to the numerical similarity of the two approximations for the potential.

It was established in Ref. 28 that an allowance for the potential of the interaction of an electron with an atom may alter the multiphoton decay cross section by a factor of 2–3. Moreover, the maximum of the dispersion curve of the detachment cross section shifts toward lower frequencies. This is due to the fact that the attractive potential “pushes” the wave functions of the continuous spectrum toward the nucleus of an atom.

In this section we ignored any perturbations of the initial state by the field, mentioned at the end of Sec. 2.3. As shown in Sec. 2.1, this approximation is justifiable if $\gamma \gg 1$.

2.5. Summary

The range of validity of perturbation theory in the description of nonlinear processes in negative ions is thus governed by the condition $\gamma \gg 1$, which leads directly to the condition that the electromagnetic field should be weak: $\mathcal{E} \ll \mathcal{E}_0$. The numerical value of \mathcal{E}_0 for negative ions is of the order of 10^7 V/cm for negative ions of alkali atoms and can be up to 10^8 V/cm for negative halogen ions.

An external field alters the initial state of an electron and this is manifested by the polarization of a negative ion. Moreover, the limit of the continuous spectrum is shifted. At high field intensities these changes may be significant and they must be allowed for in calculations of the nonlinear characteristics of negative ions. The initial state can be regarded as fixed only in moderately strong fields. Therefore, there is one further restriction on the external field intensity. In the case of processes involving few photons it can be expressed numerically in the form $\mathcal{E} \ll \mathcal{E}_0$. When the number of photons is higher, the restriction on the field intensity becomes more stringent. It should also be pointed out that a description of the processes characterized by a high degree of nonlinearity by perturbation theory methods allowing for

the interaction of the escaping electron with an atom is a technically difficult task. In practice, the calculations have been limited only to two-photon processes in negative ions.

In calculations of the matrix elements the correct allowance for the interaction of a detached electron with an atom is generally important and the effect is strongest when the composite matrix element includes the s states from the continuous spectrum. This is due to the fact that the s waves are scattered most strongly by the central potential.

We shall show later (see Sec. 4) that an allowance for the influence of a perturbation of atomic electrons by an external field on the process of nonlinear decay of negative ions is also important.

We shall now consider the methods which make it possible to describe the process of the interaction of negative ions with an external field in the case when the perturbation theory is invalid.

3. DESCRIPTION OF NONLINEAR DECAY OF NEGATIVE IONS IN THE ONE-ELECTRON APPROXIMATION GOING BEYOND THE PERTURBATION THEORY FRAMEWORK

3.1. Quasienergy approach used in the δ -potential model

As demonstrated above, perturbation theory can be used if $\kappa\omega/\mathcal{E} \gg 1$. However, this condition may not be satisfied in the case of higher field intensities or processes involving more photons. We shall now consider different methods which allow us to describe the process of interaction also in the case when $\gamma \ll 1$. For example, in Refs. 13 and 30–33 the problem of photodecay of a negative ion reduces to the problem of the photoionization of an electron in the presence of a short-range potential. The main advantage of this approach is the ability to obtain the final result analytically. An allowance is made for the possibility of the detachment of an electron directly along all the open channels (involving absorption of different numbers of photons needed to transfer a bound electron to the continuous spectrum). The validity of this approach is based on the short-range nature of the attractive potential which is exerted by a negative ion on the outer electron.

In the one-electron approximation the short-range potential may be replaced by the three-dimensional model δ potential:³⁰

$$U(r) = -\frac{2\pi}{\kappa} \delta(r) \frac{\partial}{\partial r} r. \quad (37)$$

There is only one bound state in the potential of Eq. (37) and its wave function has the following form in the limit $r \rightarrow 0$:

$$\psi(r, t) \approx \left(\frac{1}{r} - \kappa\right) e^{-iEt}. \quad (38)$$

Under the influence of an external field the state of an electron can be regarded as quasistationary because for any parameters of the field there is always a finite probability of an electron transition to the continuous spectrum.

It therefore follows that an alternating field $V(r, t)$ with a period T converts a level E_0 to a quasienergy state with a complex quasienergy E . The wave function of such a state can be represented in the form

$$\psi_E(r, t) = u_E(r, t) e^{-iEt}. \quad (39)$$

The function $u_E(r, t)$ is periodic in time with a period T and it satisfies the Schrödinger equation:

$$-\left(i \frac{\partial}{\partial t} - H_0(\mathbf{r}) - V(\mathbf{r}, t)\right) u_E(\mathbf{r}, t) = E u_E(\mathbf{r}, t), \quad (40)$$

where $H_0(\mathbf{r})$ is the Hamiltonian of the unperturbed system. The quantity E is defined to within the term $n\omega$. This circumstance reflects the fact that an electron in an alternating field has a whole spectrum of equidistant energies and not just one specific energy. The quasienergy is selected so that it reduces to the conventional energy in the limit $V(\mathbf{r}, t) \rightarrow 0$. The real part of the complex quasienergy E governs the position of the level under the influence of an alternating field, whereas the imaginary part is the photoionization probability.

The quasienergy approach was used in Refs. 30 and 31 in an analysis of the ionization of a particle with a short-range potential under the action of a circularly polarized electromagnetic wave

$$\mathcal{E}_x = \mathcal{E} \cos \omega \left(t - \frac{z}{c}\right), \quad \mathcal{E}_y = -\mathcal{E} \sin \omega \left(t - \frac{z}{c}\right), \quad \mathcal{E}_z = 0,$$

where \mathcal{E} is the electric field intensity. In this case we can construct analytically the Green's function for a particle in the field of a wave allowing for the boundary conditions in the limit $r \rightarrow \infty$ (Ref. 30). This function is an infinite sum of outgoing waves with frequencies which are a multiple of ω , which reflects the presence of harmonics of the quasienergy state of a particle. It should also be mentioned that if $E_0 + n\omega > 0$, we indeed have outgoing waves, whereas for $E_0 + n\omega < 0$, we have an exponentially damped wave.

This approach is used in Ref. 30 to solve Eq. (40) allowing for Eq. (37) in the case of a short-range potential by reducing it to the solution of the integral equation

$$u_E(\mathbf{r}, \tau) = -\frac{2\pi}{\kappa} \int \mathrm{d}\mathbf{r}' \int_0^{2\pi} \mathrm{d}\tau' G_{E'}(\mathbf{r}, \mathbf{r}', \tau, \tau') \delta(\mathbf{r}') \frac{\partial}{\partial r'} (r' u_E(\mathbf{r}', \tau')), \quad (41)$$

where $G_{E'}(\mathbf{r}, \mathbf{r}', \tau, \tau')$ is the Green's function, $E' = E - \mathcal{E}^2 / (2\omega^2)$, and $\tau = \omega t$. The presence of the δ function in the integrand and the representation of the Green's function by an algebraic series makes it possible to solve Eq. (41) analytically. As a result we obtain the following transcendental equation for the determination of the quasienergy:

$$F(\varepsilon) = 1 + \frac{1}{2} i \sum_{n=-\infty}^{\infty} (\varepsilon + n\omega)^{1/2} \times \int_0^{2\pi} \sin \theta J_n^2(\rho (\varepsilon + n\omega)^{1/2} \sin \theta) \mathrm{d}\theta = 0, \quad (42)$$

where $J_n(x)$ is the Bessel function and $\varepsilon = E' / |E_0|$, $\omega = \omega / |E_0|$, $\rho = 2^{1/2} v / \omega^2$, $v = \varepsilon / |E_0|^{3/2}$. The solution of Eq. (42) can be represented in the form

$$E_r = E_0 + \Delta - \frac{1}{2} i\Gamma, \quad (43)$$

where Δ is the shift of the level and Γ is its width. It is clear from the expression in Eq. (42) that all the open channels ($\varepsilon + n\omega > 0$) contribute to the shift and width of the level, whereas the closed channels ($\varepsilon + n\omega < 0$) contribute only to the level shift.

It should be mentioned that the quasienergy approach applies provided the conditions $|\Delta|, \Gamma \ll |E_0|$ are obeyed. These conditions are less stringent than the criterion given by Eq. (8).

Equation (42), which implicitly defines E_r , can be solved only numerically. Figure 6 shows the results of such a numerical calculation³¹ of the width Γ governing the total probability of the ionization of a bound state per unit time for two values of the frequency ω . In weak fields the width Γ exhibits a power-law dependence on the field intensity, in agreement with perturbation theory. In strong fields an increase in the field intensity reduces the probability smoothly, compared with the predictions of perturbation theory and differs from these predictions significantly only for $\gamma = 1$. Using the asymptotic behavior of the wave function $u_E(r, t)$ in the limit $r \rightarrow \infty$, we can also find the probability of an electron transition to the continuous spectrum as a result of the absorption of a certain number of photons n (on condition that $E_0 + n\omega > 0$):³⁰

$$W^{(n)} = C (\varepsilon_r + n\omega)^{1/2} \int_0^\pi \sin \theta J_n^2(\rho (\varepsilon_r + n\omega)^{1/2} \sin \theta) \mathrm{d}\theta. \quad (44)$$

The factor C is found from the condition that the sum of all the probabilities $W^{(n)}$ should be equal to the total ionization probability given by Γ .

We can see from Eq. (44) that, apart from the channel representing the absorption of the minimum number of photons n_0 needed for ionization, we can also have channels with $n > n_0$ (above-threshold absorption). In the case of linearly polarized radiation the dominant channel is that with $n = n_0$. This is not true of elliptically polarized radiation, since bending of the below-barrier path gives rise to a non-zero component of the electron momentum for escape from under the barrier at right-angles to the direction of detachment. The presence of an additional component of the momentum increases the most probable number of the absorbed photons.

It therefore follows that the solution of the model problem of the ionization of an electron from the δ potential makes it possible to describe generally the process of photodecay of negative ions in the one-particle approximation for a wide range of frequencies and intensities of an external field. Another possible approach to the process of photodecay, which allows for the real form of the one-particle poten-

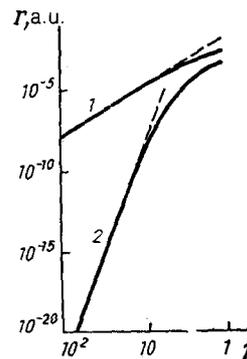


FIG. 6. Dependence of the width of a level Γ on the adiabatic parameter γ plotted for two values of the external field frequency: 1) $\omega = 0.6$; 2) $\omega = 0.27$. It is assumed that the energy of the level is $E_0 = -1$.

tial of the interaction of a weakly bound electron to the core, is the time-dependent semiclassical approach.

3.2. Semiclassical approach

Different variants of the semiclassical description of the ionization of an electron experiencing a short-range potential are given in Refs. 13, 32, and 33. The essence of the semiclassical approximation is that in the presence of an external field the initial electron state ψ_0 is assumed to be independent of time. Thus, if we write down the probability of the ionization amplitude in the form

$$A_i = -i \int_0^t \langle \psi_p^{(0)} | U(r) | \psi \rangle dt', \quad (45)$$

where $\psi(r, t)$ is the exact wave function of a weakly bound electron in an external field and $\psi_p^{(0)}$ is the wave function of a free electron in an electromagnetic field, we find that the semiclassical approximation corresponds to the replacement of ψ with ψ_0 (Ref. 32). The validity of the semiclassical approximation is based on the fact that in the short-range potential the wave function of an electron is localized near the origin of the coordinate (reference) system where the potential of the external field is weak. Therefore, outside the range of action of the atomic forces the interaction with the external field can be allowed for rigorously, whereas in the region defined by this short range this interaction can be ignored. The range of validity of the quasiclassical approximation is governed by the conditions $\omega \ll |E_0|$ and $\mathcal{E} \ll \mathcal{E}_0$, where \mathcal{E}_0 is the internal field intensity.

The expressions for the probability of photodecay of a system obtained in Refs. 13, 32, and 33 using the semiclassical approximation are the same, apart from the preexponential factor:

$$W = A(\omega, \mathcal{E}) \exp\left(-\frac{2}{3} \frac{\mathcal{E}_0}{\mathcal{E}} g(\gamma)\right), \quad (46)$$

where

$$g(\gamma) = \frac{3}{2\gamma} \left[\left(1 + \frac{1}{2\gamma^2}\right) \text{Arch } \gamma - \frac{(1 + \gamma^2)^{1/2}}{2\gamma} \right], \quad \gamma = \frac{\kappa\omega}{\mathcal{E}}.$$

The preexponential factor $A(\omega, \mathcal{E})$ in Eq. (46) represents a sum of the terms each of which corresponds to a channel of decay of the system accompanied by the absorption of a certain number of photons $n \geq n_0$. Since Eq. (46) is derived in the semiclassical approximation, the preexponential factor $A(\omega, \mathcal{E})$ cannot be determined sufficiently accurately.

A characteristic parameter which reveals the main features of the photoionization process is the adiabatic ratio γ . Physically it represents the ratio of the tunneling time of a particle across a potential barrier to the field period. In accordance with the value of γ there are two limiting cases of decay of the system in an alternating field. If $\gamma \ll 1$, the decay probability is

$$W \sim \exp\left(-\frac{2}{3} \frac{\mathcal{E}_0}{\mathcal{E}}\right),$$

i.e., the effect of an alternating field is the same as that of a constant field of the same intensity and the decay is of the tunnel nature. If $\gamma \gg 1$, the probability W depends in accordance with the power law on the external field intensity \mathcal{E} . In this case the decay is of multiphoton nature and consists of the continuous absorption of n field photons by an electron.

It is still not clear how accurate is the solution of the

problem of the ionization of an electron in a short-range potential obtained by the semiclassical approximation. According to Refs. 34 and 35, an allowance for the time-dependent evolution of the wave function $\psi(r, t)$ of the state of an electron in an external field may result in a radical change of the result for the photodecay probability. A shortcoming of this theory is also the absence of gauge invariance of the interaction with the external field. Nevertheless, the semiclassical approximation can provide a qualitative description of a number of experimental results on the above-threshold ionization of atoms^{36,37} and is at present one of the very few analytic methods which make it possible to obtain specific results on the problem of the interaction with a strong field.

In the case of negative ions the semiclassical approach had been used in Ref. 38 to obtain concrete results by calculation of the probability of two-photon decay of negative ions of alkali atoms. The short-range potential was selected in the form

$$U(r) = -\frac{\alpha_a}{2(r+r_c)^4}, \quad (47)$$

which allows for the polarization nature of the attraction of an electron to an atom over large distances. The following notation is used in Eq. (47): α_a is the static polarizability of an atom and r_c is the cutoff radius comparable with the size of an atom.

The frequency dependence of the probability of two-photon decay obtained in Ref. 38 (curve 1) is compared in Fig. 7 with the results of the numerical calculations given in Ref. 21 (curve 2). We can see that the results agree well both in the vicinity of the two-photon decay threshold and near the probability maximum. Therefore, the semiclassical solution of the problem of detachment of an electron from a short-range potential can describe generally the process of nonlinear decay of negative ions.

3.3. "Field-dressed" ion. Determination of the complex rotation of coordinates by the quasienergy method

The semiclassical approximation described above is valid if $\omega \ll |E_0|$, i.e., when the photodecay process involves many photons. At low degrees of nonlinearity an effective method is provided by discussing a system of an "ion dressed by the field." This method may be useful also in such strong fields that the perturbation theory approach is no longer valid. The method in question was used in Ref. 39 to describe nonlinear decay of negative halogen ions.

The Hamiltonian of the "ion + field" system is

$$H = H_I + H_F + H_{IF}, \quad (48)$$

where the term

$$H_I = \sum_i \left(-\frac{1}{2} \Delta_i - \frac{Z}{r_i} \right) + \sum_{i,j} \frac{1}{r_{ij}} \quad (49)$$

describes the behavior of the ion in the absence of the field; $H_F = \omega \hat{a}^+ \hat{a}$ is the Hamiltonian of the field. The last term in Eq. (48) describes the interaction of the ion with the field:

$$H_{IF} = \sum_i \mathbf{r}_i \cdot \mathbf{e} (\hat{a} + \hat{a}^+), \quad (50)$$

where \mathbf{e} is the vector representing the linear polarization of the field; \hat{a}^+ and \hat{a} are the creation and annihilation operators of a photon of frequency ω .

The state of the whole system is expanded in the basis of the states of the field $|N\rangle$ with different numbers of photons N and in the basis of the states of the ion. The ground state of a negative ion is calculated in Ref. 39 by the Hartree–Fock method.⁴⁰ Its excited states are described in the frozen core approximation. This approximation involves an analysis of the excited states simply by considering one outer electron, whereas the state of the core remains unchanged. The Hamiltonian H_0 describing the behavior of an optical electron is constructed by integration of the Hamiltonian (49) over the states of the Z electrons in the core.

The characteristics of the interaction of an ion with electromagnetic radiation calculated in Ref. 39 using the method of complex rotation of the coordinates (complex dilatation), which has been applied earlier successfully, for example, to the problem of the ionization of the hydrogen atom.⁴¹ The method involves transformation of the coordinates $r' = kre^{i\varphi}$ in the Hamiltonian, which does not alter the eigenvalues corresponding to the quasistationary states. In this transformation the radial wave functions of an electron transform the space of the quadratically integrable functions, which can be expanded in the Slater-type basis:

$$\psi_{lnk} = (kre^{i\varphi})^{l+n} \exp\left(-\frac{1}{2}kre^{i\varphi}\right), \quad (51)$$

where l is the orbital angular momentum, $n \geq 1$ is an integer, and $0 \leq \varphi < \pi/2$. The Hamiltonian matrix constructed on the basis of the states of the “field-dressed ion” system, i.e., allowing for the change in the wave functions of an atom in an external field is non-Hermitian (because $\varphi \neq 0$) and its eigenvalues are complex. The imaginary part of the eigenvalue governs the probability of decay of the system, whereas its real part determines the energy of the system allowing for the field shift, i.e., for the Stark effect. The results obtained by this method correspond to calculations involving perturbation theory of all the q , if we consider only the states of the field $|M\rangle$ characterized by the number of photons $N - q < M < N$, where N is the original number of photons.

This method is very similar to the quasienergy approach described above, except that now the external field is

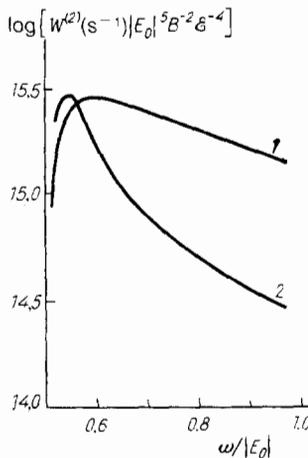


FIG. 7. Frequency dependence of the probability $W^{(21)}$ of two-photon decay of a negative alkali ion, multiplied by the product $|E_0|^5 B^{-2} G^{-4}$: 1) results taken from Ref. 38; 2) results taken from Ref. 21. Here, B is a numerical coefficient in the asymptotic expansion of the wave function of a weakly bound electron.¹² The probability $W^{(21)}$ is expressed in reciprocal seconds (s^{-1}) and the other quantities are given in the atomic system of units.

described using the second-quantization language. We now know exactly the energy of the state of the whole “ion + field” system, whereas the energy of an electron is known to within the term $n\omega$. A detailed analysis of this approach can be found in Ref. 41.

In Ref. 39 the “field-dressed ion” system is considered and the cross sections of multiphoton decay of negative halogen ions are calculated for degrees of nonlinearity ranging from $n = 2$ to $n = 5$. Figure 8 gives the results of calculations of the dependence of the two-photon decay cross sections of the F^- and Cl^- ions on the kinetic energy of a detached photoelectron. The crosses correspond to the approximately frozen core in the description of an ion. The continuous curves are obtained ignoring the interaction of a detached electron with an atom. The results differ from one another by no more than a factor of two. This degree of accuracy was assumed to be acceptable in Ref. 39 and the photodecay cross sections of negative F^- , Cl^- , Br^- , and I^- ions were determined for the degree of nonlinearity $n = 2-5$ using the plane-wave approximation for the states in the continuous spectrum. Figure 9 gives the results of calculations of the dependence of the cross sections of five-photon decay of these ions on the energy of the detached electron.

It should be pointed out that in these calculations the number of the reemitted photons was restricted, i.e., restrictions were placed on the possibility of, for example, virtual emission of s photons and absorption of $n + s$ photons in the process with the minimum number of the emitted photons equal to n , so that there is an implicit limitation on the optical flux. It should also be pointed out that the calculation procedure described above is valid until the effect of an external field alters somewhat the initial bound state.

3.4. Numerical modeling of nonlinear photodetachment of electrons from negative ions

As pointed out already, the most serious restriction in the application of perturbation theory in respect of the external field to nonlinear processes in negative ions is the requirement of the smallness of the shift of the limit of the continuous spectrum compared with the photon energy, i.e., the restriction is expressed in the form of the condition $\eta = \mathcal{E}^2/\omega^3 \ll 1$.

If the above condition is disobeyed, then transitions between the states in the continuous spectrum become important in the description of nonlinear photodecay of a negative ion. In an analysis of their role we must consider a matrix element of a transition between the states in the continuum. In multiphoton decay of a negative ion with one-photon above-threshold absorption the result based on perturbation theory in the model of the three-dimensional δ potential is obtained in Ref. 42. It is shown that the ratio of the amplitudes of transitions to the continuous spectrum, allowing for and ignoring the absorption of an additional photon, is of the same order as the parameter η . Semiclassical estimates of the matrix elements of free–free transitions near the threshold in the case of an arbitrary short-range potential give the same result.⁴³ It should also be pointed out that the criterion of validity of perturbation theory $\eta \ll 1$ had been found earlier from the solution in a linearly polarized field using the model of the δ potential⁴⁴ and analyzing the mathematical structure of the resultant solutions with-

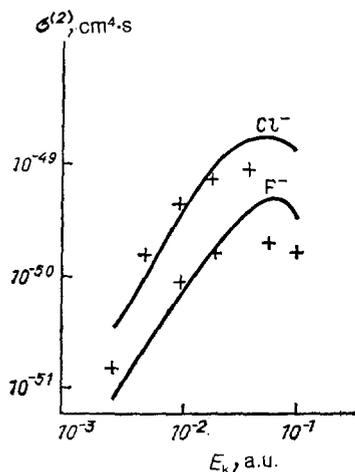


FIG. 8. Cross section of two-photon decay of the F^- and Cl^- ions plotted as a function of the kinetic energy E_k of the detached electron.³⁹ The calculations were carried out by the method of complex rotational coordinates (complex dilatation) allowing for (symbols) and ignoring (continuous curves) the interaction of the detached electron with the core.

out appeal to clear physical mechanisms. The physical meaning of the parameter η given above allows us to draw the conclusion that an increase in this parameter makes the perturbation theory approach invalid and this is accompanied by a number of new phenomena. They include above-threshold absorption of photons by electrons in the course of the detachment process, an increase in the number of photons in the process because of closure of multiphoton detachment channels in the case of the Stark shift of the limit of the continuous spectrum, and possible partial stabilization exhibited for this reason by negative ions on increase in the intensity of the external electromagnetic field.

The last of the above effects (partial stabilization) in one-photon decay was investigated in Refs. 45 and 46. In these two cases an allowance was made not only for the time dependence of the amplitude of the probability of a transition of an electron to the continuous spectrum, associated with variation of the amplitude of the electric field intensity

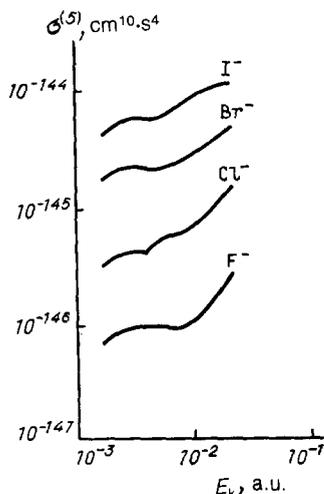


FIG. 9. Dependence of the cross section of five-photon decay of negative halogen ions on the kinetic energy E_k of the detached electron.³⁹ The calculations were made by the method of complex rotation of coordinates (complex dilatation) without allowance for the interaction of the detached electron with the core.

with time, but also for the dynamic Stark effect. It was established in particular that the law deviates from the linear dependence $W \propto \tau$, which nevertheless remains valid if the Stark effect is weak and the frequency of the incident radiation exceeds the threshold considerably. In the case of negative ions with one bound electron of energy E_0 when the frequency is $\omega \approx |E_0|$ and the polarizability obeys $\alpha(\omega) > 0$, it is found that in the range of high polarizabilities $\alpha \gg 1/\tau$, Δ_0 ($\Delta_0 = \omega + E_0$) and a large or small, compared with the width of a laser pulse $1/\tau$, detuning of the frequency from the threshold ($|\Delta_0| \lesssim 1/\tau$) when the decay probability reaches saturation the probability no longer depends on the field intensity. This is regarded as partial stabilization of a negative ion in an electromagnetic field. The physical reason for the stabilization is that an increase in the field intensity shifts the energy level downward because of the Stark effect, so that the frequency falls below the threshold value. This strongly reduces the probability of decay and alters the time dependence. At high frequencies, when $\Delta_0 \gg \alpha$, the decay is generally linear. The whole role of the Stark effect then reduces to deformation of the dispersion curve $\sigma(\omega)$. This was observed experimentally in Ref. 47.

Analytic descriptions of the above effects in multiphoton decay have not yet been published. Numerical modeling of nonlinear photodecay based on direct computer solution of a one-dimensional time-dependent (secular) Schrödinger equation is currently very popular. Different actual expressions for the short-range potential are used in this approach.⁴⁸⁻⁵⁰ Moreover, the potential employed in Ref. 50 admits the existence of several bound states and, therefore, it allows for the possibility of the appearance of resonances in the processes under discussion. Specific numerical methods for the solution of the one-dimensional Schrödinger equation can vary considerably if they include the following: construction of the evolution operator for short time intervals using operator exponential functions,⁵⁰ expansion in terms of a plane-wave basis,⁴⁸ use of direct network methods, as employed in the calculations of the dynamics of atoms.⁵¹ These investigations have revealed the following main features of the process of nonlinear photodecay of negative ions.

1. An increase in the intensity of an external electromagnetic field makes the rise of the probability of decay of a negative ion a nonmonotonic function.^{49,50} Parts of the probability rise change to regions of its fall and the positions of these falling parts are governed by the condition of closure of an electron detachment channel with a given number of ab-

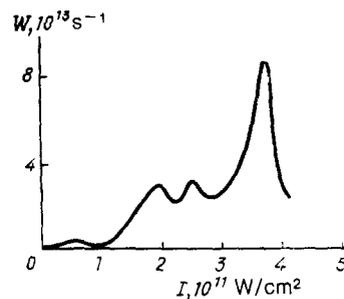


FIG. 10. Dependence of the probability of photodetachment of an electron from a one-dimensional short-range potential on the intensity of electromagnetic radiation.⁴⁹

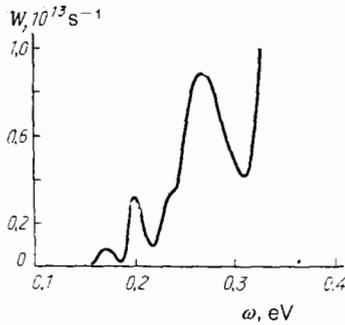


FIG. 11. Dependence of the probability of photodetachment of an electron from a one-dimensional short-range potential on the frequency of the external field.⁴⁹

sorbed photons because of the Stark shift of the limit to the continuous spectrum. The relevant dependence taken from Ref. 49 is plotted in Fig. 10.

Moreover, it is reported⁴⁹ that the frequency dependence is also nonmonotonic: it is plotted in Fig. 11 for a constant external field intensity. The drops in the frequency dependence are attributed to a reduction in the photodecay probability for the minimum possible number of the absorbed photons on increase in the frequency. On the other hand, an increase in the frequency results in opening of the photodecay channels accompanied by the absorption of a small number of photons, associated directly with an increase in the frequency and with a reduction in the Stark shift of the continuum. This is reflected by a corresponding rise of the probability on further increase in the frequency. Since the degree of nonlinearity of the photodecay process decreases on increase in the frequency, the subsequent maxima in the frequency dependence are large. It should be pointed out that nonmonotonic rise of the probability of nonlinear photodetachment of electrons from negative ions was predicted first theoretically by Ostrovskii and Tel'nov⁸⁵ using the adiabatic approximation.¹⁾

A recent experiment⁵² on the breakup of a beam of relativistic H^- ions by CO_2 laser radiation supported the importance of these effects. The results confirmed the nonmonotonic dependence of the probability H^- of photodecay on the radiation frequency. The experimentally observed dips and the frequency shift of the whole dispersion dependence can be explained only if we allow for the Stark shift of the continuum, which in this experiment exceeds the binding energy of an electron in an ion.

2. The spectrum of detached electrons found by numerical modeling has a structure consisting of isolated clear peaks⁵⁰ separated from one another by an amount ω on the energy scale, as shown in Fig. 12. The spectrum of above-threshold electrons also exhibits a shift and the lower peaks in the spectrum become suppressed because of the Stark shift of the limit of the continuous spectrum.

We can thus expect the models of the Keldysh approximation type to be capable of describing above-threshold absorption. In this case after the absorption of the minimum number of photons necessary for the detachment an electron becomes free and the relative intensities of the lines in the energy spectrum of electrons are described by the following relationship:⁵³

$$W_s = \left| \sum_{k=-\infty}^{\infty} J_k \left(\frac{\eta}{8} \right) J_{s+2k} (2s\eta)^{1/2} \right|^2, \quad (52)$$

where J_k is a Bessel function and s is the number of the observed above-threshold peak, counted from the threshold after allowing for the Stark shift. In fact, the numerical calculations (at least in the case of the Coulomb-like long-range potentials) give results⁵³ which differ considerably from Eq. (52). The reason is probably the need to allow more fully for the potential, including that of the final state, because this is ignored completely in the approximation described by Eq. (52). On the other hand, the replacement of a plane wave e^{ipx} in the final state with the exact wave function in the continuum⁵⁴ makes it possible to improve the agreement with the numerical calculations. The problem of the importance of the interaction with an atom in the final state in the case of above-threshold detachment of electrons from negative ions has not yet been solved. We shall simply mention that numerical experiments involving a one-dimensional Coulomb-like potential predict, in agreement with Eq. (52), scaling of the average number of the above-threshold absorbed photons with the parameter η . In all probability, this means that the main characteristics of the process of above-threshold absorption depend most strongly on the field parameters and much less on the parameters of the potential.

3.5. Summary

The calculation methods described in the present section make it possible to find the probability of nonlinear photodecay of a negative ion if the fields are sufficiently strong so that perturbation theory is invalid. A comparison demonstrates that the deviation of the exact solution from the perturbation theory results can be very considerable even for $\gamma \approx 1$.

The quasiclassical approximation method and the exact solution of the model problem of the detachment of an electron from the δ potential are very useful in the case of weak fields. They allow us to find the probability of photodecay when the degree of nonlinearity is high ($n \gg 1$) so that it is technically difficult to apply perturbation theory, and also to establish the general nature of the dependences of the probability on such parameters as \mathcal{E} and ω .

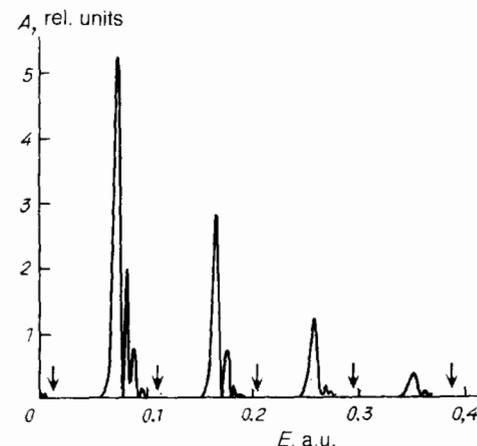


FIG. 12. Energy spectrum of electrons in the case of above-threshold detachment from a one-dimensional short-range potential.⁵⁰ The arrows identify the positions of the peaks obtained ignoring the Stark shift of the limit of the continuous spectrum.

It must be stressed once again that the precision of the semiclassical approximation is not known, whereas the exactly soluble δ -potential model suffers from the shortcoming that it is approximate. The method of complex rotation of the coordinates (complex dilatation) makes it possible to allow in principle for the real many-electron structure of negative ions, but it is limited by the external field and corresponds to the quasisteady formulation of the problem.

The direct numerical modeling of the one-dimensional Schrödinger equation makes it possible to consider strong fields in which the Stark shifts are large and the above-threshold absorption of photons is important. This reveals such new effects as, for example, closure of the photodecay channels and the Stark shift of the peaks in the electron spectrum. Further developments of computer modeling allowing for the three-dimensional nature of the problem and of the electron-electron interaction could give the most complete picture of the nonlinear photodecay.

We have considered so far the methods which ignore the role of the electron-electron correlation interaction in nonlinear processes. This role is discussed in the next section.

4. CORRELATION INTERACTION IN NONLINEAR PROCESSES

4.1. Perturbation theory in terms of the external field and the correlation interaction

We have pointed out already that the correlation interaction is important in the description of the structure of negative ions and that it can affect significantly the process of the interaction of negative ions with an electromagnetic field. A clear idea of the dynamics created by the correlation interaction can be obtained on the basis of an analogy with classical electrodynamics of continuous media.^{55,56}

It follows from the classical model that electrons in an atom oscillate harmonically relative to the nucleus, i.e., that an atom can be regarded as a set of harmonic oscillators. Since a complex atomic system consists of many atomic shells with different resonance frequencies, it follows that different shells dominate the contribution to the permittivity at different frequencies. Moreover, both amplification and weakening of the external field are possible in different frequency ranges.

A theoretical description of multiphoton detachment is possible if we use the field-theoretic many-particles theory.

If the laser field intensity is low, we can use perturbation theory in terms of the external field.⁵⁷ An atomic system with charge Z is then described by the Hamiltonian of Eq. (49). The problem can be simplified if $\Sigma(1/r_{ij})$ is approximated by a sum of the ΣV_i type, where V_i is a one-particle potential which allows for the interaction of the i th electron with the remaining $N - 1$ electrons. The Hamiltonian then becomes

$$H_1 = H_0 + H_c, \quad (53)$$

where

$$H_0 = \sum_{i=1}^N \left(-\frac{\nabla_i^2}{2} - \frac{Z}{r_i} + V_i \right), \quad (54)$$

$$H_c = \sum_{\substack{i,j=1 \\ i < j}}^N \frac{1}{r_{ij}} - \sum_{i=1}^N V_i. \quad (55)$$

Moreover, if the atomic system under investigation is exposed to the electromagnetic field of laser radiation, the interaction can be described in the dipole approximation by

$$V(t) = \vec{\mathcal{E}} \cos \omega t \sum_{i=1}^N \mathbf{r}_i, \quad (56)$$

where \mathbf{r}_i is the dipole moment of the i th electron.

If we allow both for the interaction with the field and the correlation interaction in perturbation theory, we find that the terms of the perturbation theory series can be obtained from the expression for the \hat{S} matrix in the form of the T exponential function:⁵⁸

$$\hat{S} = T \exp \left(-i \int_{-\infty}^{\infty} \hat{H}_{\text{int}} d\tau \right), \quad (57)$$

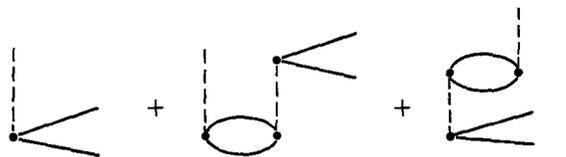
where

$$\hat{H}_{\text{int}} = \hat{H}_c + \hat{V}(t). \quad (58)$$

The operators H_c and V are considered here in the second-quantization representation.⁵⁹

The \hat{S} matrix can be expanded as a series and the T product can be expanded using the Wick theorem.⁵⁸ This makes it possible to formulate the rules for the diagram technique. Since we are considering excitations of a many-electron system, the vacuum can be defined by replacing it with an unexcited state of a negative ion. This redefinition is meaningful in the case of weak perturbations of a Fermi system.⁶⁰ The formation of a vacancy in a negative ion then corresponds to the appearance of a hole. The direction of propagation of the hole is opposite to the direction of propagation of an electron. The time axis is horizontal from left to right. A continuous line between two nodes (vertices) ($\cdot - \cdot$) corresponds to the Green's function of an electron. The free ends indicate the final or initial state. The dashed line ($--$) represents the absorption or emission of a photon. A node denotes the interaction. The electron-electron interaction is represented by a dashed line joining two nodes ($\cdot --- \cdot$) and is vertical.

This procedure can be used to calculate the one-photon decay cross sections of negative ions of alkali elements and of hydrogen. In the first order of perturbation theory in terms of the correlation interaction, we then allow for the Feynman diagrams of the type



$$(59)$$

In selecting the wave function of an electron weakly bound to a negative ion in the form of Eq. (25), we find that the photodetachment cross section is described by the relationship⁶¹

$$\sigma(\omega) = \frac{4\pi}{3c} B^2 \frac{\gamma p^3}{\omega^2} \left[1 - \left(\frac{\gamma^2 + p^2}{\beta^2 + p^2} \right)^2 - \frac{\alpha(\omega)(\gamma^2 + p^2)}{4p^3} (f(p, \gamma) - f(p, \beta)) \right]^2, \quad (60)$$

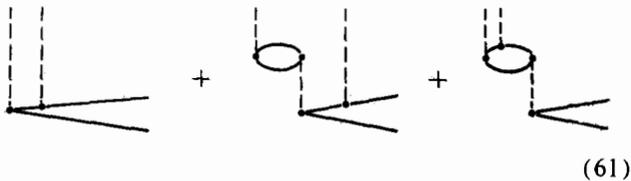
where

$$f(p, \gamma) = (\gamma^2 + p^2) \operatorname{arctg} \frac{p}{\gamma} - \gamma,$$

$\alpha(\omega)$ is the dynamic polarizability of an atom, and p is the momentum of an electron emitted on absorption of a photon of frequency ω . The correlation interaction is then allowed for in Eq. (60) by the term containing the dynamic polarizability. Equation (60) is derived on the assumption that the centrally-symmetric part of the interaction of an electron with an atom in the continuous spectrum can be ignored, so that the wave functions are plane waves. The results of the calculations reported in Ref. 61 for the Li^- ion ($\gamma = 0.21$, $\beta = 0.46$) are represented by curve 1 in Fig. 13. They agree well with the experimental results reported in Ref. 62 (curve 2) and with the theoretical calculations carried out by the method of strong coupling between the channels (curve 3) reported in Ref. 63.

The correlation interaction can be allowed for similarly in the case of two-photon processes of decay of negative ions.

The virtual excitation of a core (or an atom) can be allowed for in the first order of perturbation theory in respect of the correlation interaction and in the second order in respect of the interaction with the field by considering diagrams of the type



as well as the exchange diagrams resulting from transposition of the order in which the interactions are allowed along the time axis.

The results of a calculation²¹ of the probability per unit time of two-photon detachment of electrons from negative ions of alkali elements in the range $\text{Li}^- - \text{Cs}^-$ by circularly polarized light are presented in Fig. 14. The electric field of an optical wave is assumed formally to be equal to the atomic field intensity

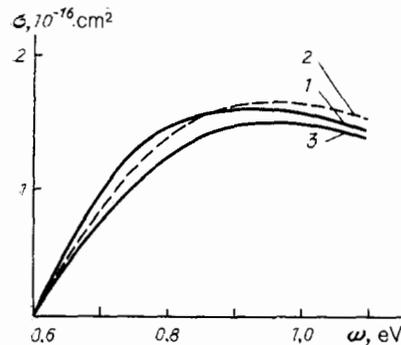


FIG. 13. Dependence of the photodecay cross section of Li^- on the radiation frequency.⁶¹

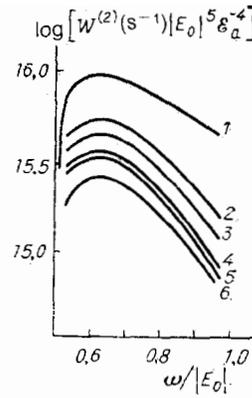


FIG. 14. Frequency dependence of the probability of two-photon decay of negative alkali ions interacting with circularly polarized light.²¹ Curve 1 represents the results of calculations made ignoring the correlation effects. Curves 2-6 are calculations carried out for the Li^- , Na^- , K^- , Rb^- , and Cs^- ions allowing for the virtual excitation of atoms.

$$\mathcal{E}_a = 5,14 \cdot 10^9 \text{ V/cm.}$$

In the case of realistic intensities \mathcal{E} we have to multiply the probability by $(\mathcal{E}/\mathcal{E}_a)^4$. In calculations dealing with a weakly bound electron we can use the wave function in the δ -potential model⁷ and the wave functions in the continuous spectrum can be assumed (as in the one-photon case) to be plane. A comparison of the curves demonstrates that the correlation effects for circular polarization of an external field reduce the probability of decay of an ion. The physical meaning of this result is demonstrated in the next section. It should be pointed out that the use of the δ -potential model for the two-photon detachment of the s electrons from negative ions by the field of linear polarization obviously give rise to large errors, because the absorption of two photons by an electron may result in its transfer to an s state in the continuous spectrum.

In this case the matrix elements of the correlation interaction will be greatly overestimated because then the singular nature of the wave function of the bound s state affects the results most strongly.

In spite of the usefulness of the perturbation theory approach demonstrated above, its direct use in the description of the processes of higher orders meets with technical and fundamental difficulties.

The technical problem is that the number of Feynman diagrams rises steeply on increase in the number of photons participating in the process and also in attempts to allow more fully for the correlation interaction. It is not then clear which part of the correlation interaction is allowed for. The latter difficulty is of fundamental nature.

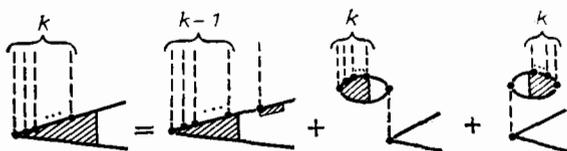
4.2. Random phase approximation applied to multiphoton processes

The full resolution of the problems mentioned in Sec. 4.1 has not yet been achieved. However, there is a way for complete summation of a certain class of diagrams which makes it possible to allow for the influence of virtual excitation of an atom on photoprocesses, including multi-photon processes. This method is the random phase approximation and, if exchange is allowed for, the approximation of random phases with exchange. A reasonably thorough discussion of these methods, applied to the description of the photoionization of atoms, can be found in the monograph of Amus'ya.⁶⁴

It follows from the theory of finite Fermi systems that inclusion of the residual interaction of electrons results in renormalization of the one-photon vertex.⁶⁴ This can be expressed most clearly in diagram language.⁶⁰

The approximation of random phases has been used successfully in calculations dealing with complex atomic systems,⁶⁵ including negative ions.^{66,67}

The diagram technique provides a simple and clear means for generalizing the random phase approximation to multiphoton processes and for obtaining the integral equation for the renormalization of the k -photon vertex. The graphical equation is then



Consequently, we can write down the integral equation for the renormalization of a k -photon vertex

$$D_{fi}^{(k)}(\omega) = D_{fi}^{(k)} - \sum_n \frac{2\omega_{ni} W_{ifni} D_{ni}^{(k)}(\omega)}{\omega_{ni}^2 - (k\omega)^2}, \quad (62)$$

where

$$W_{ifni} = \langle if | |r - r'|^{-1} | ni \rangle,$$

$D_{ni}^{(k)}(\omega)$ is the matrix element of the k -photon process allowing for renormalization, and $D_{fi}^{(k)}$ is a matrix element in the course of calculating which the $(k-1)$ -photon matrix element, is renormalized, i.e.,

$$D_{fi}^{(k)} = \sum_n \frac{D_{fi}^{(1)} D_{ni}^{(k-1)}(\omega)}{\omega_{ni} - (k-1)\omega}. \quad (63)$$

This random phase approximation had been used in calculation of the cross sections of two-photon ionization of He, and Xe atoms.^{68,69} Only the one-photon vertex $D^{(1)}$ was in fact renormalized. Figure 15 shows the results of a calculation of the cross section of two-photon ionization of the xenon atom.⁶⁹ The continuous curve gives the results of the calculations carried out in the random phase approximation using the Hartree-Fock Hamiltonian. The dashed curve represents the results of calculations of the cross sections in the random phase approximation using the Hamiltonian ob-

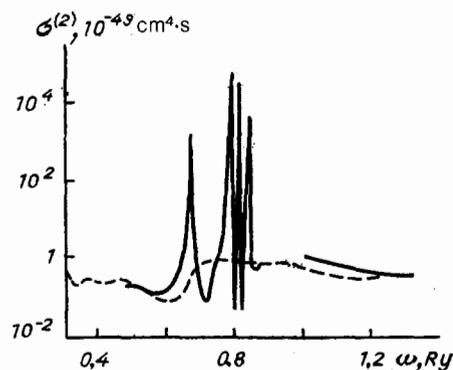


FIG. 15. Frequency dependence of the cross section of two-photon ionization of the Xe atom.⁶⁹ The calculations were made in the random phase approximation: the continuous curve represents the results based on the Hartree-Fock Hamiltonian and the dashed curve is based on the Hamiltonian obtained in the approximation of the local electron density functional.

tained in the approximation of the local electron density functional. We can see that calculations based on this functional give the average pattern of variation of the cross section with the frequency and are unsatisfactory only in the vicinity of the resonances. We can therefore expect the random phase approximation based on the local electron density functional to give good results in calculations of the cross sections of multiphoton detachment of electrons from negative ions at frequencies up to the first excitation potential of the relevant atom. At higher frequencies the dispersion curve is unavoidably affected seriously by the closely spaced resonances with the autoionizing states of a negative ion.

Unfortunately, in spite of the promising nature of the approximation represented by the local electron density functional used in calculations of multiphoton cross sections of decay of negative ions, practical calculations have not yet been carried out.

The random phase approximation based on the Hartree-Fock Hamiltonian had been employed⁷⁰ to calculate the two-photon decay cross sections $\sigma^{(2)}(\omega)$ of the iodine ion (I^-) and also in Ref. 71 to calculate $\sigma^{(2)}(\omega)$ for the chlorine (Cl^-) ion. The results of calculations for I^- are presented in Fig. 16, where curve 1 represents the random phase approximation, whereas curve 2 gives the results of calculations carried out using the Hartree-Fock approximation ignoring the correlations.⁷⁰ This figure includes also the results taken from Ref. 26 (dashed curve) and from Ref. 39 (chain curve) where the correlation effects were also ignored. The results demonstrate the appearance of the screening of the external field by electrons in an atom, since the values of the cross sections obtained allowing for the correlations are in a wide range of radiation frequencies much smaller than the cross sections calculated ignoring the correlation interaction. On the other hand, the cross sections calculated allowing for the correlations are somewhat smaller than the experimental result²⁷ obtained at the frequency of a ruby laser, but subject to a large experimental error (Fig. 16). Therefore, a more reli-

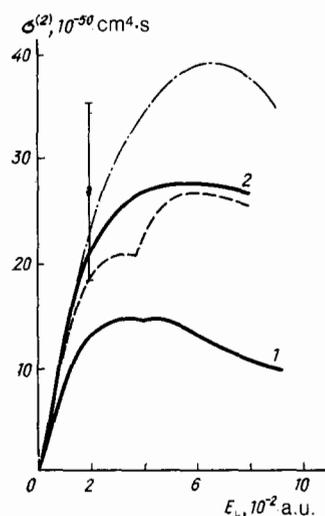


FIG. 16. Dependence of the cross section of two-photon decay of the I^- ion on the kinetic energy E_k of the detached electron. Curve 1 gives the results of calculations carried out using the random phase approximation;⁷⁰ the other calculations were carried out ignoring the correlation effects (curve 2 is based on Ref. 70, the dashed curve is based on Ref. 26, and the chain curve is based on Ref. 39).

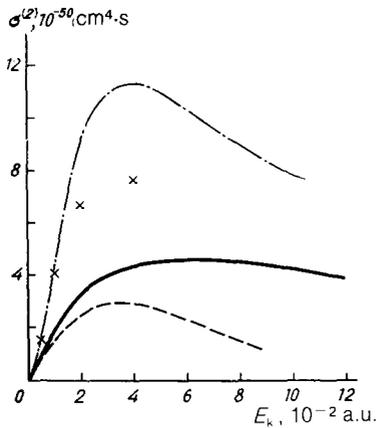


FIG. 17. Dependence of the cross section of two-photon decay of the Cl^- ion on the kinetic energy E_k of the detached electron. The continuous curve is calculated in the random phase approximation,⁷¹ whereas the dashed curve is calculated allowing for the correlation effects and using the polarization approximation;²⁸ the other calculations are made ignoring the correlation effects (the chain curve represents the results of Ref. 26 and the symbols are taken from Ref. 39).

able comparison of the theory and experiment would require an improvement in the precision of both experimental data and of the theoretical calculations.

A similar situation is observed also in the case of two-photon decay of Cl^- considered in Ref. 71, where once again the screening of the external field is manifested and the cross section for two-photon detachment decreases due to the correlation effects. The results of calculations of $\sigma^{(2)}(\omega)$ of the Cl^- ion carried out in the random phase approximation using the Hartree–Fock Hamiltonian⁷¹ are represented by the continuous curve in Fig. 17. This figure includes also the results from Ref. 26 (chain curve) and Ref. 39 (symbols), where the correlation interaction was ignored.

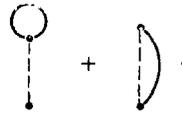
It was reported in Ref. 72 that the cross section of two-photon decay of Cl^- was determined experimentally to within 30% when the kinetic energy of a photoelectron was $E_k = 0.005$ a.u. and this cross section was $1.3 \times 10^{-50} \text{ cm}^4 \cdot \text{s}$. Calculations given in Ref. 71 gave $\sigma^{(2)} \approx 1.1 \times 10^{-50} \text{ cm}^4 \cdot \text{s}$, which agreed quite well with the experimental result. Unfortunately, experimental studies of nonlinear interaction of negative ions with an electromagnetic field are not very advanced and it is not possible to compare fully the theoretical results with the experimental data. The dashed curve in Fig. 17 represents the calculations reported in Ref. 28, where the interaction was allowed for in the polarization approximation. This approximation will be discussed in detail in the next subsection.

4.3. Polarization approximation

In calculations of multiphoton processes that occur in complex atomic systems subjected to strong optical fields we have to allow not only for the renormalization of the interaction of the field because of the residual electron–electron interaction represented by H_c , but also to consider the possibility of reemission of photons and generally allow for electron states which are “dressed” by the field. This can be done partly by introduction into the numerator of Eq. (62) of a matrix element due to an additional term Σ_{in} , which allows for the Stark effect in an external field.⁶⁸

In our opinion the quasienergy approach is clearer. Moreover, there is no need to use perturbation theory in terms of the external field when investigating multiphoton

processes. Using the quasienergy basis, we allow for the correlation interaction exactly as in the absence of a strong field and use perturbation theory in terms of this interaction. If we sum the same class of the diagrams



as in the Hartree–Fock approximation, the result is the effective potential. The form of this potential is governed by the effect on an arbitrary one-particle wave function ψ :

$$V_{\text{eff}}\psi(\mathbf{r}, t) = [V_0(\mathbf{r}) + V(\mathbf{r}, t)]\psi(\mathbf{r}, t) + \int \frac{\delta\rho(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}'\psi(\mathbf{r}, t) - \sum_{\mu \leq \mu_F} \psi_\mu(\mathbf{r}, t) \int \frac{\psi_\mu^*(\mathbf{r}', t)\psi(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}', \quad (64)$$

where $\delta\rho$ is the change in the electron density caused by an external time-dependent field. It is described by

$$\delta\rho = \sum_{\mu \leq \mu_F} (|\psi_\mu|^2 - |\psi_\mu^0|^2), \quad (65)$$

where ψ_μ^0 and ψ_μ are, respectively, the unperturbed states and the quasienergy states in the laser radiation field. The summation in Eqs. (64) and (65) is carried out over all the filled states and μ_F is the state corresponding to the Fermi limit. The first term in Eq. (64) describes the interaction of an electron with the core $V_0(\mathbf{r})$ and with the external field $V(\mathbf{r}, t)$ without allowance for the correlation effects. The second term adds the polarization interaction of an electron with oscillations of the electron density $\delta\rho$, whereas the third term represents the exchange effects and is nonlocal. The relationship (64) can also be derived by a different method. In this method it is sufficient to represent the wave function of the whole system by an antisymmetrized product of one-particle quasienergy states. The Schrodinger equation with the Hamiltonian of Eq. (49) then splits into a system of N coupled equations for one-particle quasienergy states with a potential V_{eff} given by Eq. (64).

The effective potential can be derived similarly using the method of the local electron density functional.⁹ In this approach the last term of Eq. (64) is replaced with variation of the local approximation for the exchange–correlation potential

$$\delta V_{\text{xc}} = V_{\text{xc}}[\rho_0 + \delta\rho] - V_{\text{xc}}[\rho_0], \quad (66)$$

where ρ_0 allows for the exchange–correlation interaction in the form which can be used in the case of a homogeneous electron gas. However, great caution should be exercised in the use of Eq. (66). This is because the actual form of the function is taken to be the same as in the case of a homogeneous electron gas under steady-state conditions. In the case of an atomic system we not only are dealing with a strong inhomogeneity, because the product of the characteristic momentum and the size of an atom is $pa \sim 1$, but also with a deviation from steady-state conditions. Clearly, we must at least allow for the first term of the expansion of V_{xc} as a series in $\delta\rho$. In this case the relative contribution of the harmonics is retained at the same level as in the Hartree–Fock approximation. Otherwise, the results of the calculations be-

come less accurate, as demonstrated by the calculated polarizabilities reported in Ref. 73.

The various methods of constructing the effective potential make it possible to develop convenient numerical algorithms for the calculation of the parameters of multiphoton transitions. The local potentials have major advantages in the numerical solutions of the problems. This is the reason for the recent extensive use of the local electron density functional.⁹

If a system of N equations for particles subject to an effective potential of Eq. (64) is solved simultaneously, we can determine not only the self-consistent potential V_{eff} , but also the change in the electron density

$$\delta\rho(\mathbf{r}, t) = \sum_{n=-\infty}^{\infty} e^{in\omega t} \delta\rho(\mathbf{r}, n\omega) \quad (67)$$

and find then the nonlinear response of the system. The polarization vector is then described by

$$\mathbf{P}(t) = \int \mathbf{r} \delta\rho(\mathbf{r}, t) d\mathbf{r} = \sum_{n=-\infty}^{\infty} \mathbf{P}_n e^{in\omega t}, \quad (68)$$

where

$$\mathbf{P}_n = \int \mathbf{r} \delta\rho(\mathbf{r}, n\omega) d\mathbf{r} \quad (69)$$

is the polarization vector at a frequency $n\omega$. If the field is weak compared with the atomic field and perturbation theory can be used in calculations, we have

$$\mathbf{P}_n = \hat{\chi}^{(n)} \vec{\mathcal{E}}^n, \quad (70)$$

where $\hat{\chi}$ is the nonlinear susceptibility of the system which can be calculated as described above.

This approach was used in the static limit $\omega \rightarrow 0$ in Ref. 73 to calculate the polarizabilities and hyperpolarizabilities of atomic systems with closed electron shells, such as those of the F^- and Cl^- ions. Table II lists the results of calculations of the polarizabilities α made in the Hartree-Fock approximation allowing for the correlation interaction (HF-C) and ignoring this interaction (HF), and also in the approximation of the local electron density functional corrected to exclude the self-interaction of electrons (SI-C). Clearly, inclusion of the correlation interaction in the framework of the local electron density functional changes considerably the value of α . However, we must point out that the values of the static polarizabilities obtained in Ref. 73 are greatly overestimated compared with the experimental values⁴ because of the overestimate of the exchange-correlation effects mentioned above.

The polarization approximation introduced by us on the basis of the effective potential provides a means for describing the dynamics of electrons in a complex atomic sys-

TABLE II. Dipole polarizability α (10^{-24} cm³) of negative F^- and Cl^- ions⁷³ in units of 10^{-24} cm³.

	F^-	Cl^-
HF	1.58	4.66
HF-C	2.48	5.49
SI-C	1.80	5.47

tem subjected to the action of a strong optical field and such a description can be provided for a wide range of the optical field parameters.

4.3.1. Influence of inner electrons on multiphoton detachment of outer electrons in the absence of resonances

In the case of spatial separation of the shells and subject to an allowance for their mutual influence, we can ignore the exchange term in the expression for the effective potential given by Eq. (64). If, moreover, in the Coulomb interaction $|\mathbf{r} - \mathbf{r}'|^{-1}$ we consider only the dipole term, $|\mathbf{r} - \mathbf{r}'|^{-1} \sim r^{-1} + (\mathbf{r}\mathbf{r}'/r^3)$, we find that the time-dependent part of the effective potential \tilde{V}_{eff} of Eq. (64) can be represented in the form

$$\tilde{V}_{\text{eff}}(\mathbf{r}, t) = V(\mathbf{r}, t) + V_{\text{pol}}(\mathbf{r}, t), \quad (71)$$

where $V_{\text{pol}}(\mathbf{r}, t) = \mathbf{r}\mathbf{P}(t)/r^3$ and $\mathbf{P}(t)$ is the polarization vector of the core of an atom.

The potential $\tilde{V}_{\text{eff}}(\mathbf{r}, t)$ describes the effective interaction of an electron with an external field. This interaction was allowed for in Ref. 28 using perturbation theory and calculations of the polarizabilities and two-photon decay cross sections of negative ions, halogens, and alkali atoms. The second term in Eq. (71), corresponding to the interaction of an electron with a dipole moment of an atom induced by an external field, is approximated analytically:

$$V_{\text{pol}}(\mathbf{r}, t) = \frac{\alpha_a \vec{\mathcal{E}} \mathbf{r}}{r^3} [1 - e^{-(r/r_0)^3}] \cos \omega t, \quad (72)$$

where α_a is the polarizability of an atom, r_0 is the radius of the outer atomic shell, and the factor in the square brackets is introduced to suppress the divergence limit $r \rightarrow 0$ found in numerical calculations. A similar polarization potential V_{pol} was introduced phenomenologically in Ref. 74 to allow for the polarization effects active in nonlinear processes in atoms.

The results of calculations²⁸ of the cross section for two-photon decay of a Cl^- ion allowing for the polarization potential of Eq. (72) are represented by a dashed curve in Fig. 17. It had been established that inclusion of the correlation interaction in the polarization approximation reduces by a factor of 1.5–2 the two-photon cross sections of decay of negative ions. This result is in full agreement with those given in the papers cited earlier.^{21,70,71}

Physically, the appearance of nonlinear characteristics of an atomic system because of the correlation interaction can be explained as follows. Under the influence of an external field the electron density of the core is redistributed. The electric field induced in this situation compensates partly for the influence of an external field within an atom. Consequently, the net effect experienced by a photoelectron is weaker.

Following Ref. 75, we shall demonstrate the role of the polarization effects in multiphoton detachment of, specifically, an s valence electron from a negative ion. As already pointed out, in the case of these negative ions a weakly bound electron is located at distances considerably greater than the size of an atom. In fact, if E_a is the binding energy of an electron to an atom and E_0 is the potential of the detachment of an electron from a negative ion, the ratio of these distances can be estimated to be $(E_a/E_0)^{1/2} \gg 1$. Therefore, harmonics of the polarization potential corresponding to the dipole mo-

ments induced in an atom by an external field have the asymptotic form

$$V_{\text{pol}}^{(k)} = \frac{\mathcal{E} \mathbf{r}}{2r^3} \mathcal{E}^{k-1} \chi^{(k)} (e^{-ik\omega t} + e^{ik\omega t}), \quad (73)$$

where $\chi^{(k)}$ is the nonlinear susceptibility of a neutral atom at the relevant harmonic. There are no terms with even values of k because the parity selection rules ensure that the corresponding susceptibilities vanish. The negative-frequency part of Eq. (73) corresponds to the absorption by an atom of photons of frequency $k\omega$ ($k = 1, 3, 5, \dots$). The positive-frequency part describes the emission of such photons. Since we are considering only the photodetachment process, then in the lowest order of perturbation theory we can ignore the positive-frequency perturbations and thus neglect the processes of photon reemission. Since originally an atom is in the ground state, it follows that $\chi^{(k)}$ are scalar quantities.

The subsequent solution of the problem is based on a procedure proposed in Ref. 32. An integral equation describing decay of a quasistationary state is

$$\psi(\mathbf{r}, t) = -i \int_{-\infty}^t dt' \int dr' G(\mathbf{r}, t, \mathbf{r}', t') U(\mathbf{r}') + V_{\text{pol}}(\mathbf{r}', t) \psi(\mathbf{r}', t'), \quad (74)$$

where $G(\mathbf{r}, t, \mathbf{r}', t')$ is the Green's function of an electron moving in a field $\mathcal{E}(t) = \mathcal{E}_0 \cos \omega t$ (Ref. 32) and $U(\mathbf{r})$ is the potential of the interaction of the electron with an atom.

Since U and V_{pol} fall in the limit $r \rightarrow \infty$ faster than does $1/r$, it follows that the integral equation (74) can be simplified by replacing the wave function $\psi(\mathbf{r}', t')$ on the right-hand side with an unperturbed wave function of a weakly bound electron:

$$\psi(\mathbf{r}', t') = \varphi_0(\mathbf{r}') e^{-iE_0 t'}. \quad (75)$$

The probability W of detachment of an electron per unit time by a field linearly polarized along the z axis can be determined in terms of the time-average total flux of electrons along the direction of polarization of the external field.³²

We shall now estimate the contribution made to the multiphoton detachment cross section by higher-order nonlinear susceptibilities. Under conditions when the characteristic parameter of the problem obeys $\gamma = \kappa\omega/\mathcal{E} \gg 1$, the amplitude of the probability of absorption of one photon is proportional to $\gamma/2$. We can then use the description of the interaction by Eq. (73) to estimate the ratio of the terms in the expression for the total detachment amplitude involving k and $k+2$ harmonics of the polarization interaction. An estimate of the ratio of the susceptibilities gives

$$\xi^{(k)} \gg \frac{(\bar{E} - E_a)^2}{4(d_z^2)_{00} \omega_0^3}, \quad (76)$$

where $(d_z^2)_{00}$ is the average square of the dipole moment of an atom and \bar{E} is the characteristic energy of the excitation of an atom. Numerical estimates for alkali atoms give $\xi \gtrsim 10$. Therefore, we can ignore the contribution made by the higher-order ($k > 1$) nonlinear susceptibilities to the photodetachment probability.

If we consider only the polarizability at the frequency ω and calculate the integral in Eq. (74) by the steepest-descent method bearing in mind that $\omega/|E_0| \ll 1$, we find that the

probability of n -photon detachment is given by

$$W_n = \tilde{W}_n \left(1 - \omega^2 \frac{2^{2/3} \chi^{(1)}}{B |E_0|^{1/4}} \Phi(\chi) \right)^2, \quad (77)$$

where $\Phi(\chi)$ is the Fourier transform of the product of the polarization interaction and the wave function of a weakly bound state $\varphi_0(r)$, B is a coefficient in the asymptotic expression

$$\varphi_0(r) \sim \frac{B}{r} \left(\frac{|E_0|}{2\pi^2} \right)^{1/4} e^{-\kappa r}, \quad r \rightarrow \infty, \quad (78)$$

and \tilde{W}_n is the probability of photodetachment deduced ignoring the polarization effects.³²

The presence of two terms in the parentheses in Eq. (77) reflects interference of the direct multiphoton ionization with the process involving the excitation of an atom. The factor allowing for the polarization effects is universal in the sense that it is independent of the number of photons taking part in a given process and of the field intensity. The expression describing this factor has a minimum (where it vanishes) at $\omega > |E_0|$, so that the polarization effects appear most strikingly in the one-photon detachment process and an increase in the number of photons needed for detachment weakens such effects as the field frequency decreases.

Enhancement of the polarization effects on increase in the frequency has a clear physical meaning. An increase in the energy absorbed by an atom increases its effective dimensions and enhances the overlap of the wave functions of the weakly bound and atomic electrons. There is a corresponding increase in the correlation interaction between these electrons.

Figures 18 and 19 give the results of calculations of the probability of two- and five-photon detachment of electrons from negative ions of alkali metals exposed to laser radiation with a field intensity $\mathcal{E} = 10^6$ V/cm. The values of the polarizabilities and parameters of the wave functions of the weakly bound electrons are taken from Ref. 4.

An allowance for the polarization of an atom by an external field alters the dispersion dependence of the probability of multiphoton detachment of electrons. In particular, the absolute probability decreases, as expected, on the basis of our analysis of the physical nature of the phenomenon.

4.3.2. Influence of the polarization effects on multiphoton detachment of electrons in the presence of a resonance

On approach to a resonance the relative weight of the corresponding quasienergy harmonic at a frequency $k\omega$ rises steeply. Consequently, there should be a strong enhancement of the polarization effects. Resonant polarization may appear in multiphoton processes involving negative ions on excitation and detachment of the inner electrons. In the case of atoms such a resonant enhancement is suggested and considered qualitatively in Ref. 76 using perturbation theory.

We shall consider specifically a one-photon resonance between the states $|n\rangle$ and $|m\rangle$ of an electron in an outer shell. If we ignore the exchange effects and consider shells separated sufficiently in space, we find that the resonant polarization potential is

$$V_{\text{pot}}(\mathbf{r}, t) = \int \frac{\delta\rho(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}', \quad (79)$$

where $\delta\rho(\mathbf{r}, t)$ is determined by the time evolution of a resonant two-level system. By analogy with the calculation of the

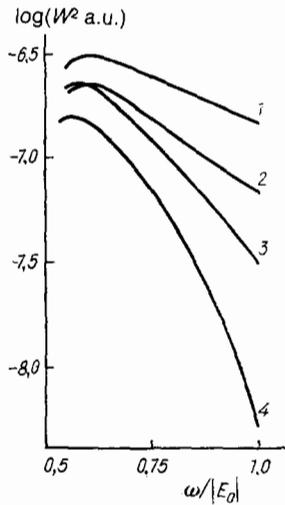


FIG. 18. Frequency dependence of the probability of two-photon decay of negative alkali ions calculated allowing for the polarization effects (curves 2, 3, and 4 represent Na^- , K^- , and Rb^- , respectively), and also ignoring such effects (curve 1 represents Na^-).⁷⁵

average dipole moment,¹ in this resonant enhancement approximation we find that

$$V_{\text{pol}}(\mathbf{r}, t) = ae^{i\omega t} \langle n | |\mathbf{r} - \mathbf{r}'|^{-1} | m \rangle + \text{c.c.}, \quad (80)$$

where

$$a = - \frac{2[\omega_{mn} - \omega - (i\gamma_m/2)] \mathbf{r}_{mn} \vec{\mathcal{E}}}{4(\omega_{mn} - \omega)^2 + 2|\mathbf{r}_{mn} \vec{\mathcal{E}}|^2 + \gamma_m^2}, \quad (81)$$

ω_{mn} is the frequency of the resonant transition, γ_m is the width of the resonating level, and \mathbf{r}_{mn} is the dipole matrix element.

In discussing the interaction between spatially separated shells we need consider only the dipole term of the expansion and use the classical mechanics relationships valid in the case of the matrix elements; this gives⁷⁷

$$r_{12}^{-1} \approx \frac{\mathbf{r}_1 \mathbf{r}_2}{r_2^3} = \frac{\omega_{mn}^2}{Z^*} (\mathbf{r}_1 \mathbf{r}_2). \quad (82)$$

The subscript 2 applies to an electron in a resonant state inside an outer shell and Z^* is the effective charge of the core. Substituting Eqs. (82) and (81) into Eq. (80), we obtain

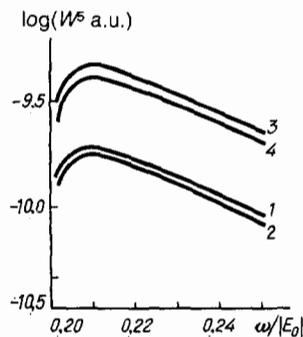


FIG. 19. Frequency dependence of the probability of five-photon decay of negative alkali ions.⁷⁵ The notation is the same as in Fig. 18.

$$V_{\text{pol}} = -2 \frac{(\mathbf{r}_{mn}) (\vec{\mathcal{E}} \mathbf{r}_{mn}) \omega_{mn}^2 [\omega_{mn} - \omega - (i\gamma_m/2)] e^{i\omega t}}{Z^* [4(\omega_{mn} - \omega)^2 + 2(\mathbf{r}_{mn} \vec{\mathcal{E}})^2 + \gamma_m^2]} + \text{c.c.} \quad (83)$$

Under the exact resonance conditions when $\omega_{mn} - \omega = 0$, assuming that $r_{mn} \sim 1$ and $Z^* \sim 1$, we obtain the following estimate for the polarization potential:

$$V_{\text{pol}} \propto \frac{i\gamma_m \mathbf{r}_{mn} \vec{\mathcal{E}} e^{i\omega t}}{2\mathcal{E}^2 + \gamma_m^2} + \text{c.c.} \quad (84)$$

At low field intensities the important parameter is the width of the resonating level and we then have

$$V_{\text{pol}}(\mathbf{r}, t) \sim \frac{i\mathbf{r}_{mn} \vec{\mathcal{E}}}{\gamma_m} e^{i\omega t} + \text{c.c.} \quad (85)$$

It should be pointed out that if the natural level width is $\gamma_m \sim 10^{-7}$ a.u., then in real situations it can increase by a large factor because of the Doppler ionization broadening and because of the finite width of the laser radiation line. However, in real situations we frequently have $\gamma_m \ll 1$, which means there is always an increase in the effective intensity of the field acting on an electron compared with the intensity of the external field.

The absorption of a resonant photon and the transfer of its energy to another nonresonant electron results in breakdown of the resonance conditions and the resonance does not appear in the subsequent absorption of photons, because in this case an outer electron is already in the field of the excited core. This effect occurs also in the case of a multiphoton resonance. All the expressions obtained above remain valid provided we replace the one-photon matrix element $(1/2)\vec{\mathcal{E}} \mathbf{r}_{mn}$ with a multiphoton element.

4.3.3. Detachment of inner electrons in a superatomic field

We shall now consider the case when an external field is high compared with the field in which a weakly bound electron is moving. We shall assume that this electron becomes free on application of this field and that it moves along a classical path in the external field $\vec{\mathcal{E}} = \vec{\mathcal{E}}_0 \sin \omega t$. The outer electrons are still described quantum-mechanically. A more self-consistent quantum approach is proposed in Ref. 78.

If we ignore the atomic potential and the initial momentum p , we find that a weakly bound electron exposed to such a field begins to oscillate in accordance with the law

$$\mathbf{r}' = \mathbf{r} - \frac{\vec{\mathcal{E}}_0 \sin \omega t}{\omega^2}. \quad (86)$$

The neglect of the initial momenta is justified if $\mathcal{E}_0/\omega \gg p \sim 1/a$.

We shall ignore the exchange effects in view of the spatial separation of the inner and outer electrons. Then, the polarization interaction of an outer electron with an inner one is given by the expression

$$V_{\text{pol}}(\mathbf{R}, t) = \int \frac{\rho(\mathbf{r}) d\mathbf{r}}{|\mathbf{R} - \mathbf{r} + [\vec{\mathcal{E}}_0 (\sin \omega t)/\omega^2]|}, \quad (87)$$

where averaging is carried out over the initial density $\rho(\mathbf{r})$ of the distribution of a weakly bound electron.

We shall estimate the probability of ionization of inner electrons W under the influence of V_{pol} by finding the coefficients in front of the time harmonics. Adopting the momentum representation for V_{pol} and ρ , we obtain a matrix element for the process of n -photon detachment from a state

$|0\rangle$ with an energy E_0 to a state $|\mathbf{p}\rangle$ with a momentum \mathbf{p} :

$$M_{\mathbf{p}0}^{(n)} = 4\pi \int \frac{\rho(\mathbf{q})}{q^2} J_n \left(-\frac{\mathbf{q}\vec{\mathcal{E}}}{\omega^2} \right) \langle \mathbf{p} | e^{-i\mathbf{q}\mathbf{R}} | 0 \rangle d\mathbf{q}. \quad (88)$$

The matrix elements described by Eq. (88) vanish for even values of n because of the parity selection rule, similar to that applicable to the polarization potential of Eq. (71). The matrix element $(4\pi/q^2) \langle \mathbf{p} | e^{-i\mathbf{q}\mathbf{R}} | 0 \rangle$ is identical with the amplitude of the ionization of an atom by electron impact considered in the first Born approximation.

Since typical momenta of electrons from an inner shell are considerably greater than the momenta of electrons in an outer shell, we can expand the exponential function as a series accurate to within the dipole term: $e^{-i\mathbf{q}\mathbf{R}} \approx 1 - i\mathbf{q}\mathbf{R}$.

Integration over the directions of the vector variable \mathbf{q} gives

$$M_{\mathbf{p}0}^{(n)} = -i8\pi^2 \langle \mathbf{p} | R_z | 0 \rangle P_n \left(\frac{\mathcal{E}_0}{\omega^2} \right), \quad (89)$$

where

$$P_n \left(\frac{\mathcal{E}_0}{\omega^2} \right) = \int_0^\infty \int_{-1}^1 \rho(q) J_n \left(-\frac{\mathcal{E}_0 q u}{\omega^2} \right) q u dq du.$$

The probability \mathcal{W} of the detachment of an electron (per unit time) is

$$\mathcal{W} = 32\pi^3 \frac{c}{\omega'} P_n^2 \left(\frac{\mathcal{E}_0}{\omega^2} \right) \sigma(\omega'), \quad (90)$$

where c is the velocity of light and $\sigma(\omega')$ is the cross section for one-photon detachment of an electron from an inner shell by a photon of frequency $\omega' = n\omega$. Estimates show⁷⁹ that if $\mathcal{E}_0/\omega^2 \ll n$ and $n \gg 1$, the direct multiphoton detachment process predominates. However, in the case of negative ions with outer p and d electrons the situation may not change fundamentally because of a strong binding of electrons in various shells.

4.4. Summary

An analysis of the dynamic correlation interaction given in this section demonstrates that its role is very important in the photoprocesses experienced by negative ions and by atoms. An allowance for the correlations may alter severalfold the nonlinear characteristics of a negative ion and can change the dispersion dependences of these characteristics.

The role of the low-frequency correlation effects for both atoms and ions can be described as a reduction in the values of the nonlinear characteristics. This is attributed to a screening effect on the core electrons. Under the influence of an external field the electron density of the atomic system becomes redistributed in such a way that the induced electric field compensates partly the influence of the external field within the atomic system.

It follows from the polarization approximation that the strongest correlation effects appear in the processes characterized by a low degree of nonlinearity and their role falls on reduction in the frequency. This observation has a simple physical interpretation. The higher the radiation frequency the higher the energies of the states which can be excited in an atom. An increase in the energy absorbed by an atom increases its dimensions. There is a corresponding increase in the overlap of the wave functions of an attached electron and of the atomic electrons, which enhances the correlation interaction. It would be interesting to obtain the same result

on the basis of more detailed numerical calculations. Such calculations can be carried out, for example, on the basis of a quasienergy approach based mainly on the assumptions given at the beginning of Sec. 4.3. A more detailed account of this approach can be found in Ref. 28, although concrete calculations have not yet been carried out.

It would also be interesting to investigate in greater detail the process of resonant absorption of the energy from the field by one electron and its transfer to another electron. In the present section it is shown that the effective intensity of the field can increase resonantly. Consequently, such a process can be very important both in ions and atoms.

Our analysis deals with the influence of the correlation effects on multiphoton one-electron detachment of electrons and it demonstrates the importance of an allowance for these effects. The influence of these effects on many-electron multiphoton processes is equally important. This is evident if only from the fact that without the correlation interaction it would have been impossible to excite autoionizing states. However, the problem in hand goes well beyond the scope of our review dealing with multiphoton properties of negative ions.

5. CONCLUSIONS

We shall briefly summarize the main conclusions reached above. If the field is sufficiently weak ($\mathcal{E} \gg 10^7 - 10^8$ V/cm) and the degree of nonlinearity is not very high, the description of multiphoton processes in negative ions can be provided by perturbation theory. An important feature is then the correct allowance for the interaction of a detached electron with the core in constructing the states in the continuous spectrum. An allowance for this interaction can alter the photodecay cross section severalfold.

Qualitative information on the influence of a strong field on a negative ion can be obtained from the exact solution of the model problem of detachment of an electron from a δ -like potential. It is shown that deviation of the perturbation theory results from the exact solution can be very large already for $\gamma \approx 1$. A more consistent approach to the description of the interaction of a negative ion with a strong field, which allows for the real structure of this ion, is the method of complex rotation of coordinates (complex dilatation).

When the degree of nonlinearity of the interaction process is high, we can describe the photodecay of a negative ion qualitatively on the basis of the exact solution of the problem for the δ -potential model; we can also obtain more accurate quantitative results by the method of complex rotation of coordinates. The main features of the nonlinear photodecay are frequently described using also the semiclassical approximation, but the question of the precision of this approach has not yet been answered. An effective method for the study of the nonlinear photodetachment of an electron from a negative ion is direct computer modeling based on numerical solution of the one-dimensional Schrödinger equation. This makes it possible to lift restrictions on the field intensity and to allow for the real time structure of the radiation pulses. However, it is not clear how these results can change if we allow for the three-dimensional nature of the potential describing the interaction of an electron with an atom.

Computer modeling of the one-dimensional Schrödinger equation demonstrates the effectiveness of the above-threshold absorption of photons in the course of photodecay

of negative ions in strong optical fields. It also manifests the Stark shift of the continuum, which closes some of the above-threshold decay channels and shifts the energy spectrum of the above-threshold electrons. The first experimental confirmation of these effects was reported in Ref. 52.

Inclusion of the correlation effects in the theory of nonlinear processes experienced by negative ions is of fundamental importance. They can be included outside the perturbation theory framework if use is made of the random phase approximation or of the polarization potential approximation; such inclusion reduces the probability of destruction of a negative ion by the field. Physically this implies a screening of the external field by the electrons of an atom perturbed by this field. The polarization approximation shows that the role of the correlation effects increases on increase in the radiation frequency, i.e., on reduction in the degree of nonlinearity of the interaction process. It would be interesting to obtain the same result on the basis of detailed numerical calculations.

In conclusion, one should mention also that since there is no criterion of weakness of the electron–electron interaction, the criterion of validity or precision of any given method can strictly speaking be formulated only in the one-electron approximation and even then not always. In the methods described above such criteria are exhibited only by two methods: that employing the perturbation theory to deal with the effects of the external field and the method of complex quasienergies. All the other methods, both one-electron and many-electron (semiclassical approximation, complex rotation of coordinates or complex dilatation, random phase approximation, and polarization approximation) do not have internal criteria of validity. Their precision can be judged by comparing the results of calculations with one another and with the experimental data. One can mention separately numerical modeling involving direct solution of the Schrödinger equation. In this case the precision of the solution itself is governed solely by the selected numerical method and by the discretization scale (step).

Our review does not deal with the multichannel quantum effect or direct calculations based on the transient Hartree–Fock method.⁸⁰ These methods have not yet been applied to negative ions, although they have been used successfully in atomic problems. Our review ignores also the possibility of using an approach involving a hyperspherical basis¹² to describe multiphoton processes. This applies particularly to negative ions of hydrogen and alkali metals, which have two *s* valence electrons. Studies of the potentialities of these very promising methods in the description of nonlinear processes in negative ions are highly desirable.

We have considered the continuous spectrum of a negative ion, using only the central field approximation in describing the interaction of an electron with an atom from which the ion is formed. Negative ions do have a complex structure of autoionizing states,⁸¹ which may be manifested in different ways in nonlinear processes. In particular, they may give rise to resonant frequency dependences of the nonlinear susceptibilities and cross sections of multiphoton detachment of electrons from negative ions. It is interesting to note the possibility of manifestation of the vibrational–rotational structure of autoionizing states of negative ions with the two valence electrons.⁸²

The present review does not deal with the time profile of

laser pulses and its influence on nonlinear processes in negative ions. During a laser pulse the Stark shift changes from zero to some maximum value. This means that the closing of channels because of the Stark effect mentioned above is a dynamic process and it depends on the time structure, duration, and amplitude of a pulse. The influence of the time structure of a pulse on one-photon decay of a negative ion is considered theoretically in Refs. 45 and 46. It would be of interest to carry out further experimental and theoretical studies of nonlinear photodecay of negative ions allowing for the pulse profile. It would be particularly interesting to consider the processes stimulated by short femtosecond laser pulses.

At first sight it might seem that there is nothing in common between the processes of nonlinear ionization of an atom and multiphoton decay of a negative ion. For example, in one case the electron being ionized experiences the Coulomb attraction to the core, whereas in the other the attractive potential is of short-range nature. However, when we adopt the many-electron approach this difference is minimized. The methods considered in the present review have been used successfully to describe the interaction of atoms with an electromagnetic field. It is then found that the photoprocesses in atoms and negative ions have much in common. In particular, there is the same suppression of the nonlinear characteristics of the atoms because of the dynamic correlation interaction. The physical interpretation of this effect is analogous: the core (positive ion) electrons screen the external field.

It is difficult to say which is easier to describe theoretically: photoprocesses in atoms or in negative ions. At first sight it might appear that negative ions are easier to tackle, because they do not have any or have only a few bound excited levels. Consequently, the problem of describing resonances of negative ions in the one-electron approximation is much less acute. Moreover, there is not infinite sum of bound intermediate states, which would have to be allowed for in calculating the characteristics of the interaction of an atom with an external field. However, as shown in Sec. 4, in describing photoprocesses it is very important to allow correctly for the dynamics of the electron–electron interaction. We must stress once again that negative ions have the same complex many-electron structure as atoms. For example, in the case of negative ions there is a whole spectrum of autoionizing states which must be allowed for in the description of the interaction with the field. A correct allowance for the electron–electron interaction is a fairly difficult task and at this moment it has not yet been done for atoms and for negative ions.

Nonlinear processes in negative ions have been investigated experimentally much less than theoretically and, as pointed out already, it is not yet possible to compare fully the theoretical predictions with experiments. At present we only have the absolute cross sections of two- and three-photon decay of the I^- ion^{27,83} and of two-photon decay of the Cl^- ion,⁷² measured at a specific frequency (ruby laser or neodymium laser frequency and second harmonics).²⁾ From the point of view of experimental investigations of nonlinear processes in negative ions it would be of interest to determine the cross sections of multiphoton detachment by radiation from a tunable laser. Such an experimental investigation can give only the dependence $\sigma^{(k)}(\omega)$, but can also reveal ex-

perimentally the influence of autoionizing resonances and shape resonances on multiphoton processes. The problem of many-electron multiphoton ionization remains open from the theoretical and experimental points of view and this applies to the formation of atoms and positive ions in excited states from negative ions, and a comparison of the efficiency of this process with the corresponding process in the case of atoms. The available data on two-electron one-photon detachment⁸⁴ allow us to assume that such a process can be observed also in the multiphoton case, as is true of atoms of alkaline earth elements.⁴³

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¹¹ The method is based on the Perelomov–Popov–Terent'ev integral equation³² and the wave function of a bound state refined in the adiabatic approximation. The results of specific calculations are very close to those presented in Figs. 10 and 11. The adiabatic approximation together with the quasienergy method can give an analytic three-dimensional solution of the problem of the detachment of an electron by a strong field from a short-range potential. Details of this elegant method can be found in Ref. 85.

²¹ A recent experimental study⁸⁶ involving the use of neodymium laser radiation of wavelength $\lambda = 1064$ nm and its harmonics yielded the cross sections of one-, two-, and three-photon detachment of electrons from negative F^- ions. The values obtained were $\sigma^{(1)} = 2.8(5) \cdot 10^{-18}$ cm², $\sigma^{(2)} = 2.0(7) \cdot 10^{-50}$ cm⁴·s, and $\sigma^{(3)} = 7.9 \cdot 10^{-83}$ cm⁶·s².

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