## RESONANCE IONIZATION OF ATOMS IN THE FIELD OF A STRONG ELECTROMAGNETIC WAVE

L. P. KOTOVA and M. V. TERENT'EV

Institute of General Inorganic Chemistry, Academy of Sciences, U.S.S.R.

Submitted to JETP editor September 30, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) 52, 732-741 (March, 1967)

In the case in which the atom has levels whose spacing is a multiple of the frequency of the light beam, there is a possible resonance mechanism of ionization in which there first occurs a real transition of the atom into an excited state with the absorption of N photons, and then there is ionization from the excited state. For the probability of ionization of the atom under these conditions we have derived the general resonance formula (24), which also takes into account the possibility that higher harmonics are present in the light beam. We also point out that it is possible for the probability to be considerably increased owing to resonance effects, and that the resonance is very wide.

• OWING to the development of laser techniques and the production of intense light beams it has become possible to observe the ionization of atoms by the field of an electromagnetic wave (see [1, 2]). The first theoretical study of the ionization of atoms by an alternating field was made by Keldysh.<sup>[3]</sup> Thereafter this problem was studied in greater detail in a series of papers (see [4-7]).

The probability of ionization by the action of an alternating field F(t) has been calculated for a system bound by short-range forces in the approximation  $F/F_0 \ll 1$ ,  $\omega/\omega_0 \ll 1$  (F<sub>0</sub> is the strength of the atomic electric field,  $\omega_0 = \kappa_0^2/2$  is the binding energy, and F,  $\omega$  are the intensity and frequency of the external field). Under the action of the field  $\mathbf{F}(t)$  there can be resonance transitions to excited states. If the characteristic times corresponding to these transitions are smaller than the time for ionization from the original level (the original level can be either the ground level or an excited level; the latter case is possible if we are considering the ionization of previously excited atoms), then there is first a transition to the excited state and ionization occurs only thereafter. Since for high levels the effective potential barrier is narrower, the probability for ionization should be much larger. In the present paper we use the method developed in <sup>[5]</sup> to derive the formula for the probability of ionization with resonance transitions taken into account. Resonance ionization owing to the admixture of higher harmonics in the light beam has also been investigated by Keldvsh.<sup>[3]</sup>

2. The integral equation for the quasistationary condition in an alternating electric field F(t) = F cos  $\omega t$  is of the form<sup>[5]</sup>

$$\psi(\mathbf{r},t) = -i \int_{t_0}^t dt' \int d\mathbf{r}' G(\mathbf{r},t;\mathbf{r}',t') V(r') \psi(\mathbf{r}',t'), \quad (1)$$

where it is stipulated that the field was turned on adiabatically at  $t = t_0$ . Here G(**r**, t; **r'**, t') is the Green's function for the electron moving in the uniform field **F**(t), and V(r) is the intraatomic potential. Let us expand the function  $\psi(\mathbf{r'}, \mathbf{t'})$  in the integrand in (1) in terms of the eigenfunctions of the Hamiltonian  $-\frac{1}{2}\nabla^2 + V(\mathbf{r})$ :

$$\psi(\mathbf{r}',t') = \sum_{j} a_{j}(t') \psi_{j}^{(0)}(\mathbf{r}') \exp(i\omega_{j}t')$$
(2)

 $(\omega_i \text{ is the energy of the jth level})$ . Then<sup>1)</sup>

$$V(\mathbf{r}')\psi(\mathbf{r}',t') = -\sum_{j} a_{j}(t')\left(\omega_{j} + \hat{p}^{2}/2\right)\psi_{j}^{(0)}(\mathbf{r}'),$$
$$\hat{p} = -i\nabla.$$
(3)

It is assumed that  $V(\mathbf{r}')$  is a short-range potential,<sup>2)</sup> and therefore the integration over  $\mathbf{r}'$  in (1) is effectively taken over the region inside the atom, where the field  $\mathbf{F}(t)$  can be regarded as a pertur-

<sup>&</sup>lt;sup>1)</sup>The atomic system of units with  $\hbar = e = m = 1$  is used.

<sup>&</sup>lt;sup>2</sup>)Owing to this condition all of the results that will follow are strictly applicable only to the ionization of negative ions. The main conclusions, however (at least for not too high frequencies of the external field), are also valid for the case of ionization of atoms, for which the potential V(r) has a Coulomb "tail" at infinity (see the discussion in Sec. 3).

bation. Then the coefficients  $a_j(t')$  can be expanded in powers of the external field.

In <sup>[5]</sup> the ionization of an atom from the ground level was considered, and the change of the wave function inside the atom owing to the electromagnetic field was not taken into account. Generally speaking this is justified, since the corrections depend on the parameter  $F/F_0 \ll 1$ . In this case  $\psi(\mathbf{r}', \mathbf{t}')$  in (1) is replaced by the wave function of the ground state of the free atom,  $\psi_0^{(0)}(\mathbf{r}') \exp(i\omega_0 \mathbf{t}')$ . If, however, there is a level with energy  $\omega_k$  whose distance from the ground state is a multiple of the frequency of the external field, i.e., if  $|\omega_0 - \omega_k|$  $\approx N\omega$  (N an integer), then the coefficient  $a_k(\mathbf{t}')$  in the expansion (2) which corresponds to this level is large and the perturbation theory does not apply.

Accordingly, in the immediate neighborhood of the resonance the states  $\psi_0^{(0)}$  and  $\psi_k^{(0)}$  are strongly intermixed, and  $\psi(\mathbf{r}', \mathbf{t}')$  is a superposition of the two functions, namely:

$$\psi(\mathbf{r}, t') = a_0^{(N)}(t') \psi_0^{(0)}(\mathbf{r}') \exp(i\omega_0 t') + a_k^{(N)}(t') \psi_k^{(0)}(\mathbf{r}') \exp(i\omega_k t').$$
(4)

The index N corresponds to the resonance transition with the absorption of N photons. For example, for resonance at the frequency  $\omega (\omega_0 - \omega_k) = \omega + \epsilon^{(1)}, \epsilon^{(1)} \rightarrow 0$ , with the initial condition at  $t = t_0$  the amplitudes  $a_0^{(1)}$  and  $a_k^{(1)}$  are given by <sup>[8]</sup>

$$a_{0}^{(1)}(t, t_{0}) = \frac{1}{2\Omega^{(1)}} e^{-i\omega_{0}t_{0}} \{x_{1}^{(1)} \exp\left[ix_{2}^{(1)}(t-t_{0})\right] - x_{2}^{(1)} \exp\left[ix_{1}^{(1)}(t-t_{0})\right]\},$$
(5)

$$a_{k}^{(1)}(t, t_{0}) = \frac{\frac{1}{2} (\mathbf{f}^{*} \mathbf{r})_{k,0}}{2\Omega^{(1)}} e^{-i\omega_{0}t_{0}} \left\{ \exp\left[-ix_{1}^{(1)} t_{0} - ix_{2}^{(1)} t\right] - \exp\left[-ix_{1}^{(1)} t - ix_{2}^{(1)} t\right] \right\},$$
(6)

where

$$x_{1;2}^{(l)} = -\frac{1}{2\epsilon^{(1)}} \pm \Omega^{(1)}, \quad \Omega^{(1)} = [(\frac{1}{2\epsilon^{(1)}})^2 + \frac{1}{2}(\mathbf{Fr})_{k,0}]^{2]_{\ell_2}}$$

The wave function (4) with the coefficients  $a_0^{(1)}$ and  $a_k^{(1)}$  given by (5) and (6) takes the effect of the external field on the motion of the electron in the atom into account correctly (to accuracy ~ F/F<sub>0</sub>) only for times t  $\ll$  t<sub>1</sub>, t<sub>r</sub> (t<sub>1</sub> is the characteristic time for ionization, and t<sub>r</sub> =  $1/\Gamma_r$ , where  $\Gamma_r$  is the radiative width of the level). For large times it is necessary to take into account the damping of  $\psi(\mathbf{r}', \mathbf{t}')$  inside the atom, associated with the radiative width and the probability of ionization. But t<sub>r</sub>, and even more so t<sub>i</sub>, is much larger than the characteristic atomic times (~  $1/\omega_0$ ) and the time for transitions between levels under the influence of the external field  $\mathbf{F}(t)$  (for more detailed estimates see Sec. 3). The subsequent passage to the limit  $t_0 \rightarrow -\infty$  is of course to be understood in a special sense. We require only that  $t - t_0$  be in the interval

$$\omega^{-1}, \omega_0^{-1}, \sqrt{\omega_0}/F \ll t - t_0 \ll t_r, t_1.$$

This is the region in which there is meaning to the concept of a time-independent probability of ionization per unit time. In first approximation the level widths owing to radiative decay and ionization can be taken into account phenomenologically in the final result.

Using Eqs. (1), (4)-(6) and the expression for the Green's function  $G(\mathbf{r}, t; \mathbf{r}', t')$  [see Eq. (17) of <sup>[5]</sup>], we get the expression for the current vector for  $t_0 \rightarrow -\infty$ :

$$\mathbf{j} = \frac{1}{8\pi^3} \sum_{n_1, n_2 = -\infty}^{\infty} \sum_{\substack{j_1 = 0, k \\ j_2 = 0, k}} \int d\mathbf{p}_1 d\mathbf{p}_2(\pi_1(t) + \pi_2(t)) F_{n_1^{j_1}}(\mathbf{p}_1) F_{n_2^{\bullet j_2}}(\mathbf{p}_2) I_{j_1^{n_1}}(p_1) I_{j_2^{\bullet n_2}}^{\star n_4}(p_2) \times \exp \{i\{(\mathbf{p}_1 - \mathbf{p}_2) (\mathbf{r} - \xi(t)) + \frac{i}{2}(p_2^2 - p_1^2)t\}\}.$$
(7)

Here  $\pi(t) = \mathbf{p} - \mathbf{A}(t)$  is the generalized momentum, and

$$\boldsymbol{\xi}(t) = -\int_{-\infty}^{t} \mathbf{A}(\tau) d\tau,$$

 $A(\tau)$  being the vector potential of the external electromagnetic field;

$$F_{n^{j}}(\mathbf{p}) = \frac{1}{2T} \int_{-T}^{T} \chi_{j}$$

$$\times (\pi(\tau)) \exp\left\{i\left[n\omega\tau - \frac{\mathbf{p}F}{\omega^{2}}\cos\omega\tau - \frac{F^{2}}{8\omega^{3}}\sin 2\omega\tau\right]\right\} d\tau, (8)$$

where

1

$$T = 2\pi/\omega, \qquad \chi_j(\boldsymbol{\pi}) = (\pi^2/2 + \omega_j) \psi_j^{(0)}(\boldsymbol{\pi}),$$

and  $\psi_j^{(0)}(\pi)$  is the Fourier transform of the function  $\psi_j^{(0)}(\mathbf{r})$ ; and

$$\tilde{a}_{j}^{(1)}(p) = \int_{-\infty}^{t} \tilde{a}_{j}^{(1)}(t') \exp\left\{i\left(\frac{p^{2}}{2} + \omega_{j} + \frac{F^{2}}{4\omega^{2}} - n\omega\right)t'\right\}dt',$$
(9)  
$$\tilde{a}_{j}^{(1)} = \begin{cases} -\frac{x_{2}^{(1)} \exp\left[ix_{1}^{(1)}t\right]}{2\Omega^{(1)} - i\delta} & \text{for } j = 0\\ \frac{1/2(\mathbf{Fr})_{k,0} \exp\left[-ix_{2}^{(1)}t\right]}{2\Omega^{(1)} - i\delta} & \text{for } j = k \end{cases}$$
(10)

The ionization probability W is calculated as the flux at infinity through a plane perpendicular to the direction of the field F(t):

$$W = \frac{4\pi}{|(\mathbf{Fr})_{k,0}|^2 + (\epsilon^{(1)})^2} \sum_{n=-\infty}^{n=\infty} \int d\mathbf{p}$$
  
×  $\delta\left(\frac{p^2}{2} + \omega_0 + \frac{F^2}{4\omega^2} + \Omega^{(1)} - \frac{\epsilon^{(1)}}{2} - n\omega\right)$  (11)  
×  $\left|\left(\Omega^{(1)} + \frac{\epsilon^{(1)}}{2}\right) F_n^{(0)}(\mathbf{p}) + \frac{1}{2} (\mathbf{Fr})_{k,0} F_{n-1}^k(\mathbf{p})\right|^2$ .

The integrals  $F_n^{(0)}(\mathbf{p})$  and  $F_{n-1}^k(\mathbf{p})$  come in only for

$$n\omega = \frac{p^2}{2} + \omega_0 + \frac{F^2}{4\omega^2} + \frac{1}{2} \varepsilon^{(1)} - \Omega^{(1)}.$$
 (12)

Using this condition, we get from (8)

$$F_{n^{0}}(\mathbf{p}) = \frac{i^{n}}{2\pi} \int_{-\pi}^{\pi} \chi_{0}(\pi(\beta))$$

$$\times \exp\left\{-i \frac{\omega_{0}}{\omega} \left[\int_{0}^{\beta} \left(1 + \frac{\pi^{2}(\alpha)}{\kappa_{0}^{2}}\right) d\alpha + \frac{1/2\varepsilon^{(1)} - \Omega^{(1)}}{\omega_{0}}\beta\right]\right\} d\beta,$$
(13)

$$F_{n-1}^{h}(\mathbf{p}) = \frac{i^{n-1}}{2\pi} \int_{-\pi}^{\pi} \chi_{h}(\pi(\beta))$$

$$\times \exp\left\{-i \frac{\omega_{0}}{\omega} \left[ \int_{0}^{\beta} \left(1 + \frac{\pi^{2}(\alpha)}{\varkappa_{0}^{2}}\right) d\alpha + \left(\frac{1/2\epsilon^{(1)} - \Omega^{(1)}}{\omega_{0}} - \frac{\omega}{\omega_{0}}\right) \beta \right] \right\} d\beta, \qquad (14)$$

where  $\pi(\beta) = \mathbf{p} + \mathbf{F}\omega^{-1} \cos \beta$ .

The presence of rapidly oscillating exponentials in (14) and (13) allows us to apply the method of steepest descents. In both cases the position of the saddle point is given approximately by the condition  $\pi^2 = -\kappa_0^2$ . At this same point  $\psi_0^{(0)}(\pi)$ , which occurs in (13), has a pole. Accordingly, the value of the integral  $F_n^{(0)}(\mathbf{p})$  is determined by the behavior of  $\psi_0^{(0)}$  in the neighborhood of the pole. This means that for the explicit calculation of  $F_n^{(0)}(\mathbf{p})$ we need to know only the asymptotic behavior of the function  $\psi_0^{(0)}(\mathbf{r})$ , which is known. The result of the integration is

$$F_{n^{0}}(\mathbf{p}) = F_{n}(\mathbf{p}) \exp\left\{\frac{\Omega^{(1)} - \varepsilon^{(1)}/2}{\omega} \operatorname{arsh} \gamma\right\}, \quad (15)$$

where  $\gamma = \kappa_0 \omega/F$ , and the expression for  $F_n(p)$  is given in <sup>[5]</sup> [Eq. (53)].

The essential feature of the integral  $F_{n-1}^{k}(p)$ , Eq. (15), is that in general the saddle point  $\pi^{2} = -\kappa_{0}^{2}$  and the pole  $(\pi^{2} = -2\omega_{k})$  of the function  $\psi_{k}^{(0)}(\pi)$  do not coincide. The variation of  $\psi_{k}^{(0)}(\pi)$  occurs over ranges  $\sim \omega_{k} = \kappa_{k}^{2}/2$ , and the difference between the positions of the saddle point and the pole is of the order of  $\omega_{k}(\omega_{0} - \omega_{k})/\omega_{k} \ll \omega_{k}$  (sic). Therefore in the approximation considered the integral (14) is determined by the behavior of  $\psi_{\rm k}^{(0)}(\pi)$  in the neighborhood of the pole and is given by

$$F_{n-1}^{\mathbf{k}}(\mathbf{p}) = F_{n'}(\mathbf{p}) \exp\left\{\left[1 + \left(\Omega^{(1)} - \frac{1}{2}\varepsilon^{(1)}\right)\frac{1}{\omega}\right]\operatorname{arsh}\gamma\right\}.$$
(16)

The integral  $F'_{n}(\mathbf{p})$  is the same as  $F_{n}(\mathbf{p})$ , if we replace the factor  $C_{\kappa_{0} l_{0}}$  [the coefficient in the asymptotic formula for  $\psi_{0}^{(0)}(\mathbf{r})$ ] by the factor  $C_{\kappa_{k} l_{k}}$  [the coefficient in the asymptotic formula for  $\psi_{k}^{(0)}(\mathbf{r})$ ] and replace the orbital angular momentum  $l_{0}$  of the electron in the level  $\omega_{0}$  by the angular momentum  $l_{k}$  corresponding to the level  $\omega_{k}$ . (We take the magnetic quantum number m to be zero for the levels  $\omega_{0}$  and  $\omega_{k}$ , since for unpolarized atoms the ionization occurs mainly from levels with m = 0 (cf. <sup>[5]</sup>).

Substituting the values of the integrals  $F_n^{(0)}(\mathbf{p})$ and  $F_{n-1}^k(\mathbf{p})$  into (11) and integrating over the momenta, we get

$$W = \left| \left( \frac{\varepsilon^{(1)}}{2} + \Omega^{(1)} \right) + \frac{(\mathbf{Fr})_{k,0}}{2} \frac{C_{\times_k l_k}}{C_{\times_0 l_0}} \sqrt{\frac{2l_k + 1}{2l_0 + 1}} \right. \\ \left. \times \left( \gamma + \sqrt{1 + \gamma^2} \right) \right|^2 \frac{W_0}{(\varepsilon^{(1)})^2 + |(\mathbf{Fr})_{k,0}|^2}, \tag{17}$$

where  $W_0$  is the probability of ionization from the ground level, as calculated, for example, in <sup>[5]</sup>.

In the region far from resonance, where  $\epsilon^{(1)} = \omega_0 - \omega_k - \omega \sim \omega_0 \gg (\mathbf{F} \cdot \mathbf{r})_{k,0}$ , we transform (17) to the form

$$W = W_{0} \left[ 1 + \frac{C_{\mathbf{x}_{k}l_{k}}}{C_{\mathbf{x}_{0}l_{0}}} \sqrt{\frac{2l_{k}+1}{2l_{0}+1}} \frac{(\mathbf{Fr})_{k,0}}{\omega_{0}-\omega_{k}-\omega} \times (\gamma + \sqrt{1+\gamma^{2}}) \right]$$
(18)

In this case the correction to  $W_0$  is small, and is determined by the parameter  $F/F_0 \ll 1$  for  $\gamma \ll 1$  and  $(F/F_0)\gamma \ll 1$  for  $\gamma > 1$ .

In the immediate neighborhood of resonance  $[\epsilon^{(1)} \sim (\mathbf{F} \cdot \mathbf{r})_{k,0} \ll \omega]$  the second term in (17) gives the main contribution. In this case the probability of ionization is

$$W = A \frac{\Gamma_{1}^{2}/4}{(\omega_{0} - \omega_{k} - \omega)^{2} + \Gamma_{1}^{2}/4} (2\gamma)^{2} W_{0};$$
  

$$A = \frac{1}{2} \frac{2l_{k} + 1}{2l_{0} + 1} \left| \frac{C_{\varkappa_{k} l_{k}}}{C_{\varkappa_{0} l_{0}}} \right|^{2}, \quad \Gamma_{1} = 2 (\mathbf{Fr})_{k,0}.$$
(19)

If there are two levels in the atom with separation  $2\omega$ , a two-photon transition to the excited state is possible. In this case, far from resonance, the correction to the probability  $W_0$  is given by second-order perturbation theory. Near resonance  $(\omega_0 - \omega_k)/\omega \approx 2$ , and therefore in (11) the integral  $F_{n-1}^k(p)$  is replaced by  $F_{n-2}^k(p)$ . Furthermore, as will be shown in the Appendix, in this case the matrix element  $\frac{1}{2}(\mathbf{F}\cdot\mathbf{r})_{k,0}$  must everywhere be replaced by the matrix element for the transition with absorption of two quanta, i.e.,

$$\frac{(\mathbf{Fr})_{k,0}}{2} \rightarrow f_{k,0}^{(2)} = \frac{1}{2^2} \sum_{j} \frac{(\mathbf{Fr})_{k,j} (\mathbf{Fr})_{j,0}}{\omega_0 - \omega_j - \omega}.$$
 (20)

Besides this there is a shift of the levels in the electromagnetic field (if one photon is absorbed and the other is emitted); for example, the shift of the ground level  $\omega_0$  is given by (see Appendix)

$$\Delta \omega_{\mathbf{a}}^{(2)} = \frac{1}{2^2} \sum_{j} |(\mathbf{Fr})_{0,j}|^2 \left( \frac{1}{\omega_0 - \omega_j - \omega} + \frac{1}{\omega_0 - \omega_j + \omega} \right).$$
(21)

Then in (17) the quantity  $\epsilon^{(1)} = \omega_0 - \omega_k - \omega$  is replaced by

$$\varepsilon^{(2)} = (\omega + \Delta \omega^{(2)}) - (\omega_k + \Delta \omega_k^{(2)}) - 2\omega.$$
 (22)

The meaning of these replacements can be easily understood from diagrams. The diagrams for the shift (21) are shown in Fig. 1, and those for the absorption of two photons and transition from level  $\omega_0$  to level  $\omega_k$  in Fig. 2.

In the general case of a resonance that arises from the absorption of N photons the integral  $F_{n-1}^{k}(\mathbf{p})$  in (11) is replaced by  $F_{n-N}^{k}(\mathbf{p})$  [it being assumed that  $N\omega/\omega_{0} \ll 1$ , since otherwise it is impossible to calculate the integral  $F_{n-N}^{k}(\mathbf{p})$  by the method of steepest descents]. The matrix element  $\frac{1}{2} (\mathbf{F} \cdot \mathbf{r})_{k,0}$  is replaced by  $f_{k,0}^{(N)}$  which corresponds to the diagram shown in Fig. 3, a:

$$\varepsilon^{(1)} \to \varepsilon^{(N)} = (\omega_0 + \Delta \omega_0) - (\omega_k + \Delta \omega_k) - N\omega,$$
  
$$\Delta \omega_{0; k} = \sum_{r=1}^{\lfloor N/2 \rfloor} \Delta \omega_{0; k}^{(2p)}, \quad [N/2] - \text{integer part of } N/2$$

where, for example, the level shift  $\Delta \omega_0^{(2\,p)}$  is described by the set of all diagrams with absorption of p photons and emission of p photons. The expression for the probability of ionization near the resonance from absorption of N photons takes the form (for justification of this formula see Appendix)

$$W = A \frac{\Gamma_N^2/4}{[(\omega_0 + \Delta\omega_0) - (\omega_h + \Delta\omega_h) - N\omega]^2 + \Gamma_{N, tot}^2/4} \times (2\gamma)^{2N} W_0, \qquad (23)$$

where  $\Gamma_N = 4f_{k,0}^{(N)}$ ;  $\Gamma_{N, \text{tot}} = \Gamma_N + \Gamma_r + \Gamma_i$ ; and the constant A is defined in (19).





In the general case, when the condition  $N\omega/\omega_0 \ll 1$  is not satisfied (but  $\omega/\omega_k \ll 1$ ), the probability of ionization is

$$W = -\frac{1}{2} \frac{\Gamma_N^2/4}{\left[(\omega_0 + \Delta\omega_0) - (\omega_k + \Delta\omega_k) - N\omega\right]^2 + \Gamma_{N, lot}^2/4} W_k,$$
(24)

where  $W_k$  is the probability of ionization from the level  $\omega_k$ .

In Eq. (23) we have included in the resonance width  $\Gamma_N$ , tot the terms  $\Gamma_r$  and  $\Gamma_i$  which are due to the radiative and ionization widths of the atomic levels (for not too large N we always have  $\Gamma_N \gg \Gamma_r$ ,  $\Gamma_i$ ).

If the electric field F(t) is not strictly monochromatic, but contains a small admixture of higher harmonics (this is often the situation in a laser beam), the resonance transition of the electron to the state k can occur with absorption of a single quantum of frequency N $\omega$ .<sup>3)</sup> In this case the matrix element  $f_{k,0}^{(N)}$  in (23) must contain an additional term  $\frac{1}{2} (\alpha_N \mathbf{F} \cdot \mathbf{r})_{k,0}$  (where  $\alpha_N$  is the fractional admixture of the harmonic of frequency  $N\omega$ ), corresponding to the diagram with absorption of one quantum of frequency N $\omega$ . Similarly, the energy shifts in each order of perturbation theory must contain terms corresponding to diagrams with the absorption and emission of quanta of frequency N $\omega$ . For example, the term added to  $\Delta \omega_0^{(2)}$ is of the form

$$\frac{1}{2^2} \sum_{j} |(\alpha_N \mathbf{F} \mathbf{r})_{0; j}|^2 \left( \frac{1}{\omega_0 - \omega_j - N\omega} + \frac{1}{\omega_0 - \omega_j + N\omega} \right) \cdot (25)$$

If the light beam contains an admixture of higher harmonics, then besides the indicated change in the resonance factor we must take into account the change of the probability of ionization from the ground level,  $W_0$ . The formula for  $W_0$  in this case has been derived by Perelomov and Popov;<sup>[9]</sup> they

<sup>&</sup>lt;sup>3)</sup>Strictly speaking, if the corresponding harmonics are present in the beam, there can be second order transitions [absorption of two quanta:  $[(N-1)\omega$  and  $\omega$ , or  $(N-2)\omega$  and  $2\omega$ , etc.], third-order transitions, and so on. We shall not analyze all of the possibilities in the general case here. For small N the number of such possibilities is small, and the contribution of each of them to the probability of ionization can be found easily by adding to  $f_{k,0}^{(N)}$  in (23) the contributions of the diagrams which correspond to the transition in question.

showed that the corrections to  $W_0$  owing to higher harmonics are important.<sup>4)</sup>

3. Let us discuss the physical meaning of (23). We see that if a resonance electronic transition occurs with the absorption of N photons of frequency  $\omega$ , for  $\gamma \gg 1$  a large factor  $(2\gamma)^{2N}$  appears. This is due to the fact that after the electronic transition the ionization effectively occurs from a higher level, where the barrier is narrower. The factor

$$\frac{\Gamma_N^2/4}{(\epsilon^{(N)})^2 + \Gamma_{N, tot}^2/4}$$

is proportional to the probability of the transition to the state k.

At exact resonance the characteristic time for the transition between the levels 0 and k is  $t_{tr} \sim \omega_0^{-1}(F_0/F)^N$ . In a ruby laser  $\omega \approx 1.79$  eV and  $F \approx 2.7 \times 10^7$  V/cm. Then for  $\omega_0 \sim 12$  eV we have  $t_{tr} \sim 10^{3N-14}$  sec. The increase of the probability of ionization near resonance is given by the ratio  $W/W_0 \sim (\omega F_0/\omega_0 F)^{2N}$ . The width of the resonance is proportional to the matrix element

$$f_{k,0}^{(N)} \sim \omega_0 (F/F_0)^N.$$

In spite of the fact that the formula (23) for the probability of resonance ionization has been derived for the case of a short-range potential V(r)in (1), it is also suitable for the description of the resonance ionization of atoms. [In an atom the potential V(r) has a Coulomb "tail" at infinity.] For not too large  $\gamma$  ( $1 \ll \gamma \ll \gamma_c$ ; for the definition of  $\gamma_c$  see <sup>[9]</sup>) it has been shown in <sup>[9]</sup> that inclusion of the Coulomb "tail" can be accomplished with perturbation theory applied to the action function, and the result reduces to the appearance of an additional factor  $(2F_0/F)^{2\lambda}$  in the expression for  $W_0$ ( $\lambda$  is the Coulomb parameter<sup>[5]</sup>). For large  $\gamma$  $(\gamma \gg \gamma_c)$  there is unfortunately no formula for W<sub>0</sub> with the Coulomb effect included. Therefore in this region we have no rigorous arguments for the justification of (23) and (24). There are, however, some qualitative considerations that favor the correctness of (24).

Let us break the true atomic potential up into two terms,  $V = V_S + V_l$ , where  $V_S$  contains only the short-range part and  $V_l$  the Coulomb "tail." In this case there is an equation for  $\psi(\mathbf{r}, t)$  analogous to (1), with V replaced by  $V_S$ , and the contribution of  $V_l$  is referred to the Green's function  $G(\mathbf{r}, t; \mathbf{r}', t')$  (but now the explicit form of G is of course unknown). Then in the integrand in (1) the function  $\psi(\mathbf{r}', t')$  can be put in the form (4), which is obviously sufficient for a foundation for (24), since the explicit form of  $W_k$  is not fixed in (24).

In conclusion the authors express their sincere gratitude to I. V. Obreimov for his interest in the work, and to B. L. Livshitz, A. M. Perelomov, and V. S. Popov for many discussions and helpful comments.

## APPENDIX

We here present the justification of Eq. (24) in the text. For this it is necessary to know the wave function of the electron when the distance between the levels in the atom is a multiple of the frequency of the external electromagnetic field. To solve the problem we consider the equation for the coefficients  $b_n(t) = a_n(t)e^{i\omega nt}$  [for the definition of  $a_n(t)$ see (2)]:

$$i\dot{b}_n(t) = -\omega_n b_n(t) - \sum_j (\mathbf{Fr})_{n,j} b_j(t) \cos \omega t.$$
 (A.1)

We look for the solution of (A.1) in the form

$$b_n(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} b_n(E) e^{iEt} dE.$$
 (A.2)

Initially (at time  $t = t_0$ ) the electron was in the state with energy  $\omega_0$ . Using the initial condition, we get from (A.1) a system of coupled equations in the energy representation:

$$(E - \omega_n) b_n(E) - \frac{1}{2} \sum_j (\mathbf{Fr})_{n, j} [b_j(E - \omega) + b_j(E + \omega)]$$
  
=  $-i\delta_{n0}e^{-iEt_0}.$  (A.3)

Let us consider the general case in which the distance between levels is close to N $\omega$ . In the region far from resonance the N-photon process is determined by N-th order perturbation theory. In particular, the correction to the ground-state wave function contains a term  $\sim [\omega_0 - \omega_k - N\omega]^{-1}\psi_k^0$ . If, however, the difference between the levels is equal to N $\omega$ , this term becomes large. Under these conditions an actual transition between the levels 0 and k is possible, with the absorption (or emission) of N photons. For times larger than the characteris-

<sup>&</sup>lt;sup>4)</sup>The essential difference between the expression (23) for the probability of resonance ionization and the corresponding formula in the paper by Keldysh [<sup>3</sup>] is that in our case the width  $\Gamma_N$  of the resonance arises owing to nonlinear effects in the interaction with the external field  $\mathbf{F}(t)$ , whereas in [<sup>3</sup>] the broadening of the resonance line is due only to the radiative ( $\Gamma_r$ ) and ionization ( $\Gamma_i$ ) widths of the atomic levels. Moreover, in our approach the level shift  $\Delta \omega_{0;k}$  in the electric field arises automatically.

tic time of the transition there is strong mixing of the states 0 and k, so that on the average the probabilities of finding the electron in these two levels are about equal. Thus near resonance the coefficients  $b_0(t)$  and  $b_k(t)$  are of the order of unity, and are large in comparison with  $b_n(t)$   $(n \neq k, 0)$ .

Using the system (A.3), we express by means of recurrence relations the coefficients  $b_0(E)$  and  $b_k(E - N\omega)$  in terms of all the others. We then get a system of two equations:

$$\begin{bmatrix} E - \omega_0 - \sum_{p=1}^{\infty} \Delta \omega_0^{(2p)}(E) \end{bmatrix} b_0(E) - \sum_{j=0, 2, \dots} f_{0, k}^{(N+j)}(E) b_k(E - N\omega) = -ie^{-iEt_0} + \xi(E) , (A.4)$$

$$-\sum_{j=0,2} f_{k,0}^{(N+j)}(E) b_0(E) + \left[ E - N\omega - \omega_k - \sum_{p=1}^{\infty} \Delta \omega_k^{(2p)}(E) \right]$$
$$\times b_k(E - N\omega) = \mathfrak{n}(E),$$

where, for example, for N = 2

$$\Delta \omega_0^{(^{\mathcal{C}}}(E) = \frac{1}{2^2} \sum_j |(\mathbf{Fr})_{0,j}|^2 \left( \frac{1}{E - \omega_j - \omega} + \frac{1}{E - \omega_j + \omega} \right),$$
$$f_{0,k}^{(2)}(E) = \frac{1}{2^2} \sum_j (\mathbf{Fr})_{0,j} (\mathbf{Fr})_{j,k} \frac{1}{E - \omega_j - \omega}. \quad (A.5)$$

The functions  $\xi(E)$  and  $\eta(E)$  which occur in the right members of the system (A.4) contain  $b_0$  and  $b_k$  with other arguments, but still they occur with factors ~  $F/F_0 \ll 1$ . Their contribution to  $b_0(E)$  and  $b_k(E - N\omega)$  near resonance is small, and we shall not take them into account in what follows.

The determinant of the system (A.4) is (when we take into account the main terms with respect to  $F/F_0$ ):

$$D = \left[ E - \omega_0 - \sum_{p=1}^{[N/2]} \Delta \omega_0^{(2p)}(E) - x_1^{(N)}(E) \right]$$
$$\times \left[ E - \omega_0 - \sum_{p=1}^{[N/2]} \Delta \omega_0^{(2p)}(E) - x_2^{(N)}(E) \right], \quad (A.6)$$

where [N/2] is the integer part of the number N/2, and

$$\begin{aligned} x_{1;2}^{(N)}(E) &= -\frac{1}{2} \varepsilon^{(N)}(E) \pm \Omega^{(N)}(E), \\ \Omega^{(N)}(E) &= \left[ (\frac{1}{2} \varepsilon^{(N)})^2 + |f_{0,k}^{(N)}(E)|^2 \right]^{\frac{1}{2}}, \\ \varepsilon^{(N)}(E) &= \left( \omega_0 + \sum_{p=1}^{\lfloor N/2 \rfloor} \Delta \omega_0^{(2p)}(E) \right) \\ &- \left( \omega_k + \sum_{p=1}^{\lfloor N/2 \rfloor} \Delta \omega_k^{(2p)}(E) \right) - N \omega. \end{aligned}$$
(A.7)

The dependence on E in the functions

$$x_{1;2}^{(N)}(E), \quad \Omega^{(N)}(E), \quad \varepsilon^{(N)}(E), \quad \sum_{p=1}^{\lfloor N/2 \rfloor} \Delta \omega_{0;k}^{(2p)}(E)$$

can be neglected, and we can consider them at the points  $E = \omega_0 = \omega_k + N\omega$ , since in the region of a resonance that occurs with the absorption of N photons all of these functions are very small in comparison with  $\omega_0$ ,  $\omega_k$ ,  $\omega$ , and they change appreciably in ranges of the order of  $\omega_0$ ,  $\omega_k$ ,  $\omega$ . Then the matrix elements  $f_{0,k}^{(N)}$  ( $E = \omega_0$ ) and  $\Delta \omega_{0,k}^{(2p)}$  ( $E = \omega_0$ ) are equal to the  $f_{0,k}^{(N)}$  and  $\Delta \omega_{0,k}^{(2p)}$ , defined in the text of this paper.

From the system (A.4) we find

$$b_{0}(E) = -\frac{i}{D} e^{-iEt_{0}} \left( E - N\omega - \omega_{k} - \sum_{p=1}^{[N/2]} \Delta \omega_{k}^{(2p)} \right),$$
  
$$b_{k}(E - N\omega) = -iD^{-1}e^{-iEt_{0}} \int_{k,0}^{(N)}.$$
 (A.8)

Integrating over the energy in (A.2), we get the expressions for the amplitudes:

$$a_{0}^{(N)}(t, t_{0}) = \frac{1}{2\Omega^{(N)}} \exp\{i[\Delta\omega_{0}t - (\omega_{0} + \Delta\omega_{0})t_{0}]\}$$

$$\times [x_{1}^{(N)} \exp\{ix_{2}^{(N)}(t - t_{0})\} - x_{2}^{(N)} \exp\{ix_{1}^{(N)}(t - t_{0})\}],$$

$$a_{k}^{(N)}(t, t_{0}) = \frac{f_{k,0}^{(N)}}{2\Omega^{(N)}} \exp\{i[\Delta\omega_{k}t - (\omega_{0} + \Delta\omega_{0})t_{0}]\}$$

$$\times [\exp\{-ix_{1}^{(N)}t_{0} - ix_{2}^{(N)}t\} - \exp\{-ix_{1}^{(N)}t - ix_{2}^{(N)}t_{0}\}].$$
(A.9)

The expressions (A.9) are valid only in the neighborhood of the resonance, when  $a_0^{(N)} \sim a_k^{(N)} \sim 1$ . Using (A.9) and the formula (4) for  $\psi(\mathbf{r}, t)$  and repeating the arguments given in the text for the derivation of (19) for N = 1, we can in an analogous way derive the general expression (23).

<sup>1</sup>G. S. Voronov and N. B. Delone, JETP 50, 78 (1966), Soviet Phys. JETP 23, 54 (1966).

<sup>2</sup>G. S. Voronov, G. A. Delone, N. B. Delone, and O. V. Kudrevatova, JETP Letters 2, 377 (1965), transl. 2, 237 (1965).

<sup>3</sup> L. V. Keldysh, JETP **47**, 1945 (1964), Soviet Phys. JETP **20**, 1307 (1965).

<sup>4</sup>A, I. Nikishov and V. I. Ritus, JETP 46, 776 (1964), Soviet Phys. JETP 19, 529 (1964).

<sup>5</sup>A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, JETP 50, 1393 (1966), Soviet Phys. JETP 23, 924 (1966).

<sup>6</sup>A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, JETP 51, 309 (1966), Soviet Phys. JETP 24, 207 (1967). <sup>7</sup>A. Gold and H. Bebb, Phys. Rev. Letters 14, 60 (1965).

<sup>8</sup> L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963, page 173.

<sup>9</sup>A. M. Perelomov and V. S. Popov, JETP 52, 514 (1967), Soviet Phys. JETP 25, 336 (1967).

Translated by W. H. Furry 92