PHYSICAL REVIEW A VOLUME 9, NUMBER 2 FEBRUARY 1974

Multiphoton excitation and ionization of He atoms in an intense laser beam

Man Mohan and S. N. Haque
Department of Physics and Astrophysics, University of Delhi, Delhi-7, India
(Received 29 November 1972; revised manuscript received 27 April 1973)

Transition amplitudes corresponding to bound-bound and bound-free multiphoton transitions for an He atom in the presence of an intense laser beam have been evaluated. Results show that for multiphoton excitation the transition amplitude for small values of N (the number of photons being absorbed) decreases with increasing intensity. However, for higher values of N the amplitude continues to rise with increasing intensity. A particular feature of the present method of calculating multiphoton ionization is that the transition amplitude increases linearly for small values of the intensity, but for higher values, the slope of the curve decreases, showing an important nonlinear effect.

INTRODUCTION

The theory of ordinary photoexcitation and photoionization in helium is an old problem in physics, which was treated successfully in the early days of quantum mechanics. Those calculations were based on the first-order perturbation theory and described the process of a bound electron absorbing a photon, and thereby being excited or emitted from the atom, depending upon the photon energy. In the interaction of a laser beam with an atom, however, the ordinary photoeffect is not possible. This is because the energy of a single photon of the laser beam is quite small as compared to the energy difference of any two quantum levels of the helium atom. In such a case only simultaneous absorption of many photons can result in the excitation or ionization of the atom. The problem of multiphoton ionization of helium using perturbation theory was done for the first time by Bebb and Gold.[1] At high intensity, however, perturbation theory is not applicable, as the perturbation energy due to light waves becomes comparable to the unperturbed Hamiltonian. In such cases the method developed by Reiss[2] for bound quantum systems is very useful. Essentially, Reiss's method involves a unitary transformation, which approximately removes the electromagnetic field from the problem. Furthermore, the accuracy of this technique increases with the increase in the number of photons involved in any given process.

In this paper we have used the Reiss method for the calculation of transition probabilities for multiphoton excitation and multiphoton ionization of an He atom in the presence of an intense laser beam.

In Sec. I, we have calculated the transition probability for transition from the $(1s^2)$ to the $(1s, 2p)$ state of an He atom by absorption of N photons of an intense field of frequency ω and vector potential A. The value of N is such that $E_f - E_i = N\omega$, where E_f and E_i are the energies of the final and initial state, respectively. (Throughout we have used $\hbar = c = 1$.)

In Sec. II, we present our calculation for the transition probability in the case of multiphoton ionization of an He atom by absorption of N photons. The value of N is such that $N\omega = \epsilon_I + k^2/2m$ is satisfied, where ϵ_I is the ionization energy of the atom and the last term is the kinetic energy of the emitted electron.

Our Eqs. (20) and (26) contain infinite sums for this purpose and to obtain rapidly converging results we have made use of Euler's transformation technique.[3] The value of the transition probability thus obtained is plotted against y (intensity parameter) for a particular value of N (N being the number of photons absorbed). An interesting point worth mentioning here is that for the transition $(1s^2)$ to $(1s, 2p)$ the transition probability for small values of N increases with intensity, attains a maximum, and then decreases for higher values of intensity. However, for higher values of N, the transition probability increases with an increase in the intensity, and shows no decreases, as for low values of N. Finally, the transition probability for multiphoton ionization shows an important nonlinear effect at high intensity.

In Sec. III we discuss our results in direct physical terms.

I. TRANSITION MATRIX ELEMENT FOR MULTIPHOTON EXCITATION OF AN He ATOM

The hydrogen atom and the hydrogenic ion resemble each other in that they are both one-electron systems that do not exhibit two-electron processes such as double excitation, double ionization, simultaneous ionization and excitation, etc. Here we have taken the hydrogenic model for the helium atom. The helium atom has two electrons, one of which is effected by interaction

with radiation, while the other remains in the $1s$ orbit of the atom: The operator that governs the interaction of radiation with the atom according to Reiss's approximation will be $e^{ie\vec{A}\cdot\vec{x}_1}$. Now, as we cannot make any distinction between the two electrons in an atom, it may happen that the first electron remains in the $1s$ orbit while the other electron makes the transition to the higher state. In such a case, only the operator $e^{ie\vec{A}\cdot\vec{x}_2}$ will be effected. To take account of the indistinguishability of the two electrons in the hydrogenic model for the helium atom, the matrix element will be the sum of the two parts, one containing the operator $e^{ie\vec{A}\cdot\vec{x}_1}$ and the other containing the operator $e^{ie\vec{A}\cdot\vec{x}_2}$.

Thus the matrix element[4,5] to be evaluated for transition from $\mathrm{He}(1s^2)$ to $\mathrm{He}(1s, 2p)$ by absorption of N photons of frequency ω and vector potential A is given by

$$M = (\psi_f, (e^{ie\vec{A}\cdot\vec{x}_1} + e^{ie\vec{A}\cdot\vec{x}_2})\psi_i), \tag{1}$$

where ψ_i and ψ_f are the initial and final states of the helium atom.

For the helium atom it is necessary to use an approximate wave function. The simplest wave function for the ground state is that obtained by Hylleras[6] using a variational method, i.e.,

$$\psi_i = \psi_{1s}(\alpha/r_1)\psi_{1s}(\alpha/r_2), \tag{2}$$

where $\alpha = 1.6875$.

Furthermore, it has been shown by Eckart[7] that a good approximation to the wave function of an excited state of helium (other than the s state) is obtained by taking a symmetrical combination of the product of two wave functions, one representing the ground state of an electron in the field of charge $\beta = 2$, and the other the excited state of an electron in the field of charge $\gamma = 1$. Thus, if we write $\psi_{np}(Z/r)$ for the wave function of a single electron in the nl state in the field of charge Z, we may take as a sufficiently good approximation for the wave function of an excited singlet state of helium (other than the s state) the form

$$\psi_f = (N_i/\sqrt{2})[\psi_{1s}(\beta/r_1)\psi_{np}(\gamma/r_2) + \psi_{np}(\gamma r_1)\psi_{1s}(\beta/r_2)], \tag{3}$$

the two electrons being distinguished by the numerals 1, 2. In Eq. (3), N_i represents the normalization constant.

For excited s states the wave functions used have been based on the form (3) but with ψ_{np} differing somewhat from the hydrogenic form. Thus, for $2\,^1s$, Marriott and Seaton[8] take in (3)

$$\psi_{20} = [N'/(4\pi a_0^3)^{1/2}]\,[e^{-\beta' r/a_0} - S(r/a_0)\,e^{-\gamma' r/a_0}], \tag{4}$$

where $N' = 0.568$, $\beta' = 1.136$, $\gamma' = 0.464$, and $S = 0.317$. These parameters were determined variationally, subject to the condition that the $2\,^1s$ function should be orthogonal to the Hylleras ground-state wave function (2).

As the ground-state and the final-state wave functions (2) and (3) are symmetric with respect to the interchange of coordinates r_1 and r_2, the matrix element (1) becomes

$$M = 2(\psi_f, e^{ie\vec{A}\cdot\vec{x}_1}\psi_i). \tag{5}$$

Substituting the values of ψ_i and ψ_f from (2) and (3) into (5), we have

$$M = N_i\sqrt{2}\left(\int d\vec{r}_2\psi_{1s}^*(\beta/r_2)\psi_{1s}(\alpha/r_2)\right) \times\left(\int d\vec{r}_1\,\psi_{np}(\gamma/r_1)\,e^{ie\vec{A}\cdot\vec{x}_1}\psi_{1s}(\alpha/r_1)\right). \tag{6}$$

In obtaining (6) we have utilized the orthogonality relation

$$\int \psi_{np}^*(\gamma/r)\,\psi_{1s}(\alpha/r)\,d\vec{r} = 0. \tag{7}$$

Performing the integration in the first part of (5), we have

$$I = \int d\vec{r}\,\psi_{1s}^*(\beta/r)\,\psi_{1s}(\alpha/r) = \frac{\delta(\alpha\beta)^{3/2}}{(\alpha+\beta)^3}. \tag{8}$$

The relevant radial contribution from the second part of (6) is given by

$$II = \int_0^\infty dr\,r^2 R_{21}(\gamma/r)\,j_1(eAr)R_{10}(\alpha/r). \tag{9}$$

Using the relations

$$R_{21}(\gamma/r) = (\gamma/2a_0)^{3/2}(\gamma r/a_0\sqrt{3})\,e^{-\gamma r/2a_0}, \tag{9a}$$

$$R_{10}(\alpha/r) = (\alpha/a_0)^{3/2}\,2e^{-\alpha r/a_0}, \tag{9b}$$

and

$$j_l(eAr) = \left(\frac{\pi}{2eAr}\right)^{1/2}\sum_{k=0}^{\infty}\frac{(-)^k}{k!\Gamma(l+k+\frac{3}{2})}\left(\frac{eAr}{2}\right)^{2k+l+1/2}, \tag{10}$$

in expression (9), we have

$$II = \left(\frac{\pi}{2eA}\right)^{1/2}\sum_{k=0}^{\infty}\frac{(-)^k}{k!\Gamma(l+k+\frac{1}{2})}\left(\frac{eA}{2}\right)^{2k+l+1/2} \times\left(\frac{\gamma}{2a_0}\right)^{3/2}\frac{2\gamma}{3a_0}\left(\frac{\alpha}{a_0}\right)^{3/2}\frac{\Gamma(2k+l+4)}{(\alpha/a_0+\gamma/2a_0)^{2k+l+4}}. \tag{11}$$

Finally, utilizing the above and the orthogonality property of the spherical harmonics in the angular part of (6), we have for the matrix element M the following:

$$M=64\sqrt{3}N_i\frac{(\alpha\beta)^{3/2}}{(\alpha+\beta)^3}\frac{\gamma^{5/4}}{a_0^4}\sqrt{\pi}\;Y_1^{\pm1,0}(\theta_A,\phi_A)\,\alpha^{3/2}\left(\frac{\pi}{2eA}\right)^{1/2}\sum_{k=0}^{\infty}\frac{(-)^k}{k!\Gamma(k+\frac{3}{2})}\left(\frac{eA}{2}\right)^{2k+3/2}\frac{\Gamma(2k+5)}{(\alpha/a_0+\gamma/2a_0)^{2k+5}}\,. \quad (12)$$

We now use the dipole-approximation plane-wave form for eA

$$eA\simeq ea\cos\omega t=\tfrac{1}{2}a\beta e\,, \quad (13)$$

where

$$\beta=e^{i\omega t}+e^{-i\omega t}\,.$$

The intensity-dependence term in (12) is then

$$\left(\frac{\pi}{2eA}\right)^{1/2}\sum_{k=0}^{\infty}\left(\frac{eA}{2}\right)^{2k+3/2}=\frac{\sqrt{\pi}}{2}\sum_{k=0}^{\infty}(b\beta)^{2k+1}\left(\frac{3}{4a_0}\right)^{2k+1}, \quad (14)$$

where $b=\frac{1}{3}eaa_0$.

The factor β^{2k+1} in (14) can be expanded by the binomial theorem as

$$\beta^{2k+1}=\sum_{j=0}^{2k+1}\binom{2k+1}{j}e^{i(2k+1-2j)\omega t}\,. \quad (15)$$

In this process, as N photons are absorbed, we therefore use

$$2k+1-2j=-N$$

or

$$j=k+\tfrac{1}{2}+\tfrac{1}{2}N.$$

From (15) it is also clear that

$$2k+1\geq j$$

$$\geq k+\tfrac{1}{2}+\tfrac{1}{2}N$$

or

$$k\geq\tfrac{1}{2}(N-1)\,.$$

Thus (15) takes the form

$$\beta^{2k+1}=\sum_{k=\frac{1}{2}(N-1)}^{2k+1}\binom{2k+1}{k+\frac{1}{2}(N+1)}e^{-iN\omega t}\,. \quad (16)$$

Substituting (16) and (14) into (12), we have for M

$$M=64(3\pi)\,\alpha^3\beta^{3/2}\gamma^{5/4}\frac{1}{(\alpha+\beta)^3}\;\frac{1}{a_0^4}\,y_1^{\pm1,0}(\theta_A,\phi_A)\sum_{k=\frac{1}{2}(N-1)}^{2k+1}\binom{2k+1}{k+\frac{1}{2}(N+1)}\left(\frac{3b}{4a_0}\right)^{2k+1}\frac{(-)^k}{k!\Gamma(k+\frac{5}{2})}\;\frac{\Gamma(2k+5)}{(\alpha/a_0+\gamma/2a_0)^{2k+5}}\,e^{-iN\omega t}\,. \quad (17)$$

If the index of summation in (17) is shifted so that the sum starts with zero index, we have then

$$M=64\,(3\pi)\,\alpha^3\beta^{3/2}\gamma^{5/4}\frac{1}{(\alpha+\beta)^3}\frac{1}{(\alpha+\frac{1}{4}\gamma)^4}\,(-)^{(N-1)/2}\left(\frac{3b}{(2\alpha+\gamma)}\right)^N$$
$$\times\sum_{k=0}^{\infty}(-)^k\left(\frac{3b}{2\alpha+\gamma}\right)^{2k}\binom{N+2k}{k}\{[\tfrac{1}{2}(N-1)+k]^2+3[\tfrac{1}{2}(N-1)+k]+2\}\,e^{-iN\omega t}\,. \quad (18)$$

The T-matrix element for transition from the He($1s^2$) to the He($1s$, $2p$) state can now be easily written as

$$T=(E_{1s^2}-E_{1s,2p})\,64\sqrt{(3\pi)}\;\alpha^3\beta^{3/2}\gamma^{5/4}\frac{1}{(\alpha+\beta)^3}\;\frac{1}{(\alpha+\frac{1}{2}\gamma)^4}\,y_1^{\pm1,0}(\theta_A,\phi_A)\left(\frac{3}{2\alpha+\gamma}\right)^N\tau\,, \quad (19)$$

where τ is defined as the "reduced" transition amplitude depending on the intensity only. It is given by

$$\tau=b^N\sum_{k=0}^{\infty}(-)^k\,b^{2k}\left(\frac{3}{2\alpha+\gamma}\right)^{2k}\binom{N+2k}{k}\{[\tfrac{1}{2}(N-1)+k]^2+3[\tfrac{1}{2}(N-1)+k]+2\}\,, \quad (20)$$

where $b=\frac{1}{3}eaa_0=\frac{1}{2}y$.

II. TRANSITION MATRIX ELEMENT FOR MULTIPHOTON IONIZATION OF AN He ATOM

The matrix element to be evaluated for the transition of one of the electrons from the ground state of the He atom to the continuum state is given by the same expression as (1), i.e.,

$$M=(\psi_f,(e^{ie\vec{A}\cdot\vec{x}_1}+e^{ie\vec{A}\cdot\vec{x}_2})\psi_i)\,, \quad (1)$$

where ψ_i and ψ_f are the initial and final states of the helium atom, and further we have assumed a hydrogenic model for the He atom. For the initial state, we use the same Hylleras,[6] one-parameter variational wave function, i.e.,

$$\psi_i = \psi_{1s}(\alpha/r_1)\,\psi_{1s}(\alpha/r_2), \tag{1'}$$

where $\alpha = 1.6875$. As the electron is finally emitted from the atom, therefore, we use for the final state the symmetrical combination of the product of the two wave functions, one representing the ground-state wave function of the He^+ ion with charge $\beta = 2$, and the other the wave function for the free state of an electron, i.e.,

$$\psi_f = 2^{-1/2}[\psi_{1s}(\beta/r_1)\,\psi_K(r_2) + \psi_{1s}(\beta/r_2)\,\psi_K(r_1)], \tag{21}$$

where $\psi_{1s}(\beta/r)$ with $\beta = 2$ is the ground-state wave function of the He^+ ion and $\psi_K(r)$ is the wave function of the free state. The free-state wave function is given by

$$\psi_K(r) = 4\pi \sum_{l_f=0}^{\infty} \sum_{m_f=-l_f}^{l_f} (i)^{l_f} e^{i\eta_{l_f}} R^c_{l_f}(\gamma'/k_f r) \times y^{m_f}_{l_f}(\theta, \phi)\, y^{m_f}_{l_f}(\theta_K, \phi_K), \tag{22}$$

where

$$R^c_{l_f}(\gamma'/k_f r) = N^c_{l_f}(\gamma')\,(2k_f r)^{l_f} e^{-ik_f r} \times F(l_f + 1 + i\gamma' \mid 2l_f + 2 \mid 2ik_f r),$$

$$N^c_{l_f}(\gamma') = \frac{|\Gamma(l_f + 1 - i\gamma')|}{(2l_f + 1)!}\, e^{\pi\gamma'/2},$$

$$\gamma' = \frac{1}{k_f a_0},$$

and

$$\eta_{l_f} = \arg\Gamma(l_f + 1 - i\gamma'),$$

while k_f and (θ_K, ϕ_K) stand for the wave vector and azimuthal angles of the emitted electron.

Now using the same argument as in Sec. I we get the T-matrix element for the multiphoton ionization of He from the ground state, as

$$T = \sum_{l_f=0}^{\infty} (-)^{l_f} e^{-i\eta_{l_f}}\, y^{m_f}_{l_f}(\theta_K, \phi_K)\, A_{l_f}, \tag{23}$$

where

$$A_{l_f} = (\Delta E)\, 16\pi^2 \left(\frac{\alpha}{a_0}\right)^{3/2} \frac{(\alpha\beta)^{3/2}}{(\alpha+\beta)^3}\, y^{m_f}_{l_f}(\theta_A, \phi_A)\, e^{\pi\gamma'/2}\, (2k_f)^{l_f} \times \left(\frac{3b}{4}\right)^N \sum_{k=0}^{\infty} (-)^k \left(\frac{3b}{4}\right)^{2k} \binom{N+2k}{k} \frac{1}{\Gamma[\frac{1}{2}(N-l_f)+k_f+1]}\, \frac{1}{\Gamma[\frac{1}{2}(N+l_f)+\frac{3}{2}+k_f]} \times \left(\frac{1}{a_0}\right)^{l_f+3} \sum_{\gamma'=0}^{N-l_f+1+2k} (-)^{N-l_f+1+2k} \frac{\Gamma(N-l+2+2k)}{\Gamma(\gamma'+1)\Gamma(N-l+2+2k-\gamma')} \times \frac{1}{|\Gamma(l_f+1-i\gamma')|} (\alpha - ia_0k_f)^{-N-2k-2-i\gamma'+\gamma'} (\alpha + ia_0k_f)^{-l_f-1+i\gamma'-\gamma'}.$$

Here ΔE is the energy difference between initial and final states.

Therefore,

$$|T|^2 = \sum_{l_f=0}^{\infty} \sum_{l'_f=0}^{\infty} (-)^{l_f} e^{-i\eta_{l_f}} (-)^{l'_f} e^{+i\eta_{l'_f}}\, y^{*m_f}_{l'_f}(\theta_K, \phi_K)\, y^{m_f}_{l_f}(\theta_K, \phi_K)\, A^*_{l'_f} A_{l_f}. \tag{24}$$

Since we are interested in the total ionization rate, we integrate (24) over all angles assumed by k_f, i.e., over $d\Omega_K$. Using the orthogonality relation for the spherical harmonics,[1] we have

$$\int d\Omega_K\, |T|^2 = \sum_{l_f=0}^{\infty} |A_{l_f}|^2. \tag{25}$$

In the accompanying graphs we have plotted τ_1 the "reduced" transition amplitude against y for a particular value of N, where τ_1 contains only the intensity-dependent term of A_{l_f} and is given by

$$\tau_1 = \frac{|A_{l_f}|}{(\Delta E)\, 16\pi^2 (\alpha/a_0)^{3/2}\, y^{m_f}_{l_f}(\theta_A, \phi_A)}\, \frac{(\alpha+\beta)^3}{(\alpha\beta)^{3/2}}. \tag{26}$$

III. DISCUSSION

We have presented our numerical results in Figs. 1 and 2 as the variation of the "reduced" transition amplitude against the parameter y. Our aim is to examine the behavior of the transition amplitude with intensity and to look for possible deviation from the perturbation theory, which predicts the latter to be a function of Nth power of the intensity for the Nth-order process. In the expression for the transition amplitude in Eqs. (19) and (23), intensity-dependent factors have been separated out and renamed as "reduced" transition amplitude. The photointensity term here appears as y and is related to the conventional form by the expression

$$y^2 = (I/I_0)(4/E_\lambda^2),$$

where I is the flux of the photon in W/cm^2, I_0 is 1.4014×10^{17} W/cm^2, and E_λ is the energy of the photon in atomic units. The condition for the validity of the perturbation theory is that

$$y \leq 1, \quad \text{i.e.,} \quad I \leq I_0 \tfrac{1}{4} E_\lambda^2,$$

which, in our case, is 10^{14} W/cm^2. If we examine Fig. 1 for small values of N, we find the increase of log τ with increasing y to be straight line, but the nature of the curve changes in the neighborhood of $y \simeq 1$ and the transition amplitude decreases with the increase of y. For large values of N, on the other hand, the transition amplitude increases and finally becomes constant and resembles the ionization curve. This happens because of the influence of the intense external field on the multiphoton process in the atom.[5,9,10] Arytyunyant *et al.*[9] and Vornov *et al.*[10] have estimated the decrease of ionization potential owing to external field and have thus concluded that at high intensity there is smearing of the upper energy levels of the atom, leading to overlapping and merging of the levels into a quasicontinuum spectrum. For example, to reduce the ionization potential by 1 eV or to reduce the needed number of quanta for transition by one, i.e., $\Delta I \simeq h\omega \simeq 1$ eV, the external field required is $E \sim 10^7$ V/cm. Therefore, for the intensities that are under consideration, the $2p$ state of the He atom acts as a quasicontinuum state and causes the transition amplitude to increase with intensity. However, if we evaluate the transition amplitude for the transition He($1s^2$) to He($1s, 2s$), we will not get the same behavior as described above; here the amplitude will decrease with the intensity, because the $2s$ state of the He atom is quite far off from the continuum, as compared with $2p$ state, and does not overlap with the continuum for the high intensity under consideration.

Another interesting feature revealed by Fig. 1 is the dominance of a large number of photon transitions over the smaller one. The existence of such a type of anomaly has been confirmed by Koval'skaya *et al.*[11] from their experiment on photocurrent instability and multiphoton processes in $CdSnP_2$ using a Q-switched glass laser. They found that the probability of three-photon absorption in semiconductors with complicated band structure can exceed the probability of the two-photon process. Similar types of anomalies were also observed recently in gallium phosphide by Pyshkin *et al.*[12]

In Fig. 2, where the reduced transition amplitude for ionization has been plotted against y, the deviation from perturbation theory is not so radical. Though the behavior here is not exactly a straight line, the deviation is small. If we take $x = \frac{1}{2}y = (\frac{1}{3} eaa_0)^2$, then it is found that

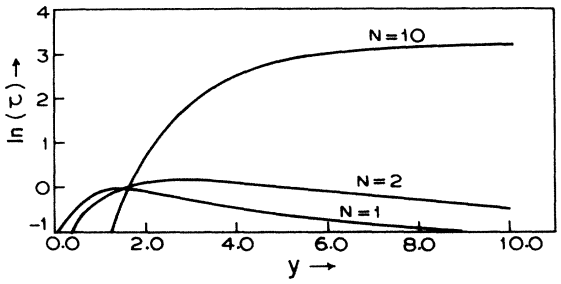

FIG. 1. "Reduced" transition amplitude for the transition He($1s^2$) He($1s,2p$), as a function of the intensity parameter for various photon multiplicities.

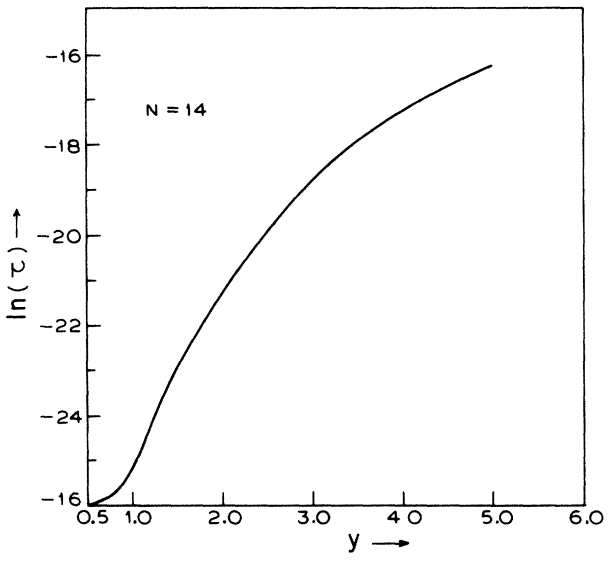

FIG. 2. "Reduced" transition amplitude corresponding to the multiphoton ionization, as a function of the intensity parameter for $N = 14$; photons are being absorbed.

$$\frac{\partial \ln |T|^2}{\partial \ln x} = N - \Delta ,$$

where Δ depends on N and y and increases with an increase of y. Thus the nonlinear effect at high intensity appears to be due to a deviation of the power-law dependence of transition probability on intensity of something less than the lowest order of transition. Furthermore, it is likely that other processes (e.g., saturation of multiquantum transitions, saturating intermediate resonances, spatial and temporal structure of the laser beam, etc.) than the one considered here, might show stronger deviations from the perturbation behavior. Much work has also been done in the field of multiquantum process on the experimental side. Bystora *et al.*[13] and Agostine *et al.*[14] have observed the multiphoton ionization effect in alkali-metal vapors and noble gases. Three-photon ionization of a helium atom in the excited $2s$ state has been studied by Bakos *et al.*[15] In their experiment, He atoms of a discharge afterglow plasma in the excited metastable states 2^1s and 2^3s were exposed to focused radiation of a Q-switched ruby laser. The ions produced in the radiation were registered with a probe. Since the ion signal is directly connected with the cross section, the plot of the probe signal with the intensity gives directly the variation of the multiphoton ionization cross section with intensity. It is found that the variation of amplitude signal with intensity is a linear one for the field interval $(1-3)\times 10^5$ V/cm. This behavior is also the same as is expected theoretically at low intensity. Recently an experiment on the light intensity dependence of the multiphoton-ionization probability in the resonance case for a high-intensity laser beam has been done by Bakos, Kiss, Szabo, and Tendler.[16] They have measured the general intensity dependence of the five-photon ionization of a triplet metastable helium atom, by the method of Langmuire probe using radiation of a Q-switched Nd glass laser with frequency tuning. It was found that at high intensity, the probe signal varies nonlinearly with intensity, as expected, and as also described earlier. This type of nonlinear effect has also been observed by various other authors.[9]

Finally, the study of multiphoton transitions in the optical band using laser radiation is of great interest from two points of view. It is possible to use this method, first, to investigate the transition between states having identical parity, i.e., transition that is forbidden in the single-photon process. For example, this property is utilized by Burell and Kunze[17] in their experiment on two-photon absorption and stimulated Raman scattering on an excited helium atom, which is useful for the possible measurement of high-frequency electric field in the plasma.

Second, if the structure of the levels and the matrix element of the transitions in investigations are known, then the investigation of multiphoton processes can yield additional information on the statistical properties of laser emission,[18,19] e.g., the detection of singly stimulated two-photon emission from metastable deuterium atoms by Braundich and Lambropoulos,[18] who have provided the experimental tool for the study of the effects of the coherence properties of radiation on two-photon process. Furthermore, the effect of intense external field on an atom, which results in lowering of the ionization potential, and hence the reduction of the needed number of quanta, is of practical interest for photon ionization of excited atoms, atoms on surface media and in the process of dissociation, for in these cases the work function is several times smaller than the ionization energy of atoms.[9]

[1]H. B. Bebb and A. Gold, Phys. Rev. 143, 1 (1966).
[2]H. R. Reiss, Phys. Rev. A 1, 803 (1970); Phys. Rev. Lett. 25, 1149 (1971); Phys. Rev. D 3533 (1971).
[3]P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), Vol. I.
[4]M. R. C. McDowell and J. P. Coleman, *Introduction to the Theory of Ion-Atom Collisions* (North-Holland, Amsterdam, 1970), p. 324.
[5]Man Mohan and R. K. Thareja, Phys. Rev. A 7, 34 (1973); J. Phys. B 5, L134 (1972); J. Phys. B 6, 806 (1973); Man Mohan, J. Phys. B 6, 1218 (1973).
[6]E. A. Hylleras, Z. Phys. 54, 347 (1929).
[7]C. Ackart, Phys. Rev. 36, 878 (1950).
[8]X. Marriott and M. J. Seaton, Proc. Phys. Soc. A70, 296 (1957).
[9]I. N. Arutyunyan and G. A. Askaryan, Zh. Eksp. Teor. Fiz. Pis'ma Red. 12, 378 (1970) [Sov. Phys.—JETP Lett. 12, 259 (1970)].
[10]G. S. Vornov, G. A. Delone, H. B. Delone, and O. Y. Kudrevatova, Zh. Eksp. Teor. Fiz. Pis'ma Red. 2, 377 (1955) [Sov. Phys.—JETP Lett. 2, 237 (1965)]; T. B. Bystrova, G. S. Voronov, G. A. Delone, and N. B. Delone, Zh. Eksp. Teor. Fiz. Pis'ma Red. 5, 223 (1967) [Sov. Phys.—JETP Lett. 5, 178 (1967)]; G. S. Voronov and N. B. Delone, Zh. Eksp. Teor. Fiz. 50, 78 (1966) [Sov. Phys.—JETP 23, 54 (1966)]; G. S. Voronov, G. A. Delone, and N. B. Delone, Zh. Eksp. Teor. Fiz. 51, 1860 (1966) [Sov. Phys.—JETP 24, 1122 (1967)]; G. S. Voronov, Zh. Eksp. Teor. Fiz. 51, 1496 (1966) [Sov. Phys.—JETP 24, 1009 (1967)].
[11]V. S. Koval'skaya, N. A. Ferdman, E. I. Leonov, V. M. Orlov, N. A. Groyunova, S. L. Pyshkin, and S. J.

Radautsan, Zh. Eksp. Teor. Fiz. Pis'ma Red. 12, 399 (1970) [Sov. Phys.—JETP Lett. 12, 275 (1970)].
[12]S. L. Pyshkin, N. A. Ferdman, S. I. Radautsan, V. A. Kovarsky, and E. V. Vitiu, in Proceedings of the Sixth International Conference on Quantum Electronics, Kyoto, 1970 (unpublished).
[13]T. B. Bystrova, G. S. Voronov, G. A. Delone, and N. B. Delone, Zh. Eksp. Teor. Fiz. Pis'ma Red. 5, 223 (1967) [Sov. Phys—JETP Lett. 5, 178 (1967)].
[14]P. Agostini, G. Barjot, G. Mainfray, C. Manus, J. Morellec, and J. Thebault, Phys. Lett. A 31, 367 (1970); and in Ref. 12, p. 92.
[15]J. Bakos, J. Kantor and A. Kiss, Zh. Eksp. Teor. Fiz. Pis'ma Red. 12, 371 (1970) [Sov. Phys—JETP Lett. 12, 255 (1970)].
[16]J. Bakos, A. Kiss, L. Szabo, and M. Tendler, Phys. Lett. A 41, 163 (1972).
[17]C. F. Burrell and H. J. Kunze, Phys. Rev. Lett. 29, 1445 (1972).
[18]P. Braunlich and P. Lambropoulos, Phys. Rev. Lett. 25, 135 (1970); Phys. Rev. Lett. 25, 986 (1970).
[19]J. Ducuing and N. Bloembergen, Phys. Rev. 133, A1493 (1964).