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MULTIPHOTON PROCESSES IN THE FOCUS OF AN INTENSE LASER BEAM WITH EXPANSION OF THE INTERACTIVE REGION TAKEN INTO CONSIDERATION

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A multiphoton process in the focus of laser radiation or near the focal points of individual modes is considered. It is shown that at large radiation densities saturation and expansion of the interactive region occurs, which substantially changes the form of the dependence of the number of events on the intensity of the field, $N(E) \sim E^k$; in particular, for very large fields $N \sim E^3$ and does not depend on the quantum nature of the process. Different forms of the field distribution are considered, namely, a cone with a focal constriction and a gaussian radial distribution. The results and conclusions of experiments on multiphoton ionization of atoms and molecules in a laser beam are reviewed. The possibility of using the results for a determination of the volume needed to initiate multiphoton ionization in a light spark is mentioned.

THE appearance of powerful nano- and picosecond lasers has increased interest in an investigation of the processes of multiphoton ionization, dissociation, etc. in the beam's focus. However, the abrupt inhomogeneity of the distribution of the intensity of laser radiation at the focus, at the focal points of individual modes, and at "hot" points requires special investigations in order to verify the possibility of estimating the dependence of the probability of events on the intensity of the field from the experimentally observed yield of events for a multiphoton process (ionization, dissociation, excitation, etc.).

In the present article the yield function for a multiquantum process in a focused light beam is obtained with expansion of the interactive region taken into account, and it is shown that such delocalization may significantly change the form of the observed result for the probability of an event as a function of the field intensity, and may even make it independent of the number of quanta required for the process.

Let us consider a multiquantum process in a focused beam. If the probability $w(\mathbf{r}, t)$ for the process per unit time at a given point of space is given, then the volume concentration of events happening (for example, ions which are produced) is given by

$$n(\mathbf{r},t)=n_a\left\{1-\exp\left(-\int_0^t w\,dt\right)\right\},$$

where n_a denotes the concentration of the initial atoms or molecules. This formula takes account of the decrease in the number of initial atoms, which is substantial at large field intensities, giving large probabilities for the process. In order to determine the total number of events it is necessary to integrate over the volume of the field distribution, for which it is necessary to specifically define the form of the function w and specify the field distribution.

Usually the probability w is assumed to have the form[1]

$$w = AE^{2k},$$

where k is the number of quanta which is sufficient for ionization (k may be smaller than $k_0 = \langle (I/\hbar\omega) + 1\rangle$ for a number of reasons: broadening of the upper levels in strong fields, transition through a resonance level, etc.), and E(t) is the amplitude of the light field (the envelope of the light beam).

We modulate the field distribution near the focus by converging and diverging beams with a constriction at the center due to the caustic of the focus or diffraction balancing. We consider three models of such a distribution for which there is a general relation for the field averaged over the cross section of the beam of radius a(z), $E \approx E_f a_f/a$, where E_f denotes the field at the minimum cross section of constriction of radius a_f. For example, for a focused multi-mode beam the volume of constriction (volume of the focus) is given by

$$V_f \approx \pi a_f^2 l_f,$$

where the transverse dimension of the focusing volume is $2a_f = f\varphi$ and the longitudinal dimension is $2l_f \approx a_f f/d \approx f^2\varphi/d$. Here f denotes the focal length of the lens, d is the diameter of the beam at the lens, and φ is the intrinsic divergence of the beam due to the choice of modes or due to diffraction. In this connection the aperture angle of the cone of focusing is $2\theta = d/f$. We note that in the case of a single mode the angle of divergence φ is close to the diffraction limit $\varphi \approx \lambda/d$, and the radius of the focusing volume is determined from the condition $\lambda/a_f \approx \theta$.

1. As the first model for the field distribution, let us consider a simplified double-cone geometry with a cylindrical constriction associated with a homogeneous distribution of the field over the cross section and with an abrupt drop at the edge of the beam. In this case the total number of events is given by

$$N = n_a\int_V\left\{1-\exp\left[-\int_0^t w\,dt\right]\right\}dV$$

$$= n_a V_f\left\{1-\exp\left[-A\int_0^t E_f^{2k}\,dt\right]\right\} + n_a\pi\,\mathrm{tg}^2\theta\int_l^\infty \{1-\exp[-B/z^{2k}]\}z^2\,dz,$$

$$B = A\int_0^t (E_f a_f/\mathrm{tg}\,\theta)^{2k}\,dt.$$

Introducing the variable $\xi = (l/z)^{2k}$, let us write the second term in the form

$$\frac{D}{2k}\int_0^1 [1-\exp(-x_f\xi)]\frac{d\xi}{\xi^{1+\beta}}, \quad \beta=\frac{3}{2k}<1, \quad D=\pi n_a l^3 \operatorname{tg}^2\theta;$$

$$x_f = B/l^{2k} \approx \int_0^t w_f\,dt$$

is the quantity which determines the total probability of ionization $W = 1 - \exp(-x_f)$ of an atom in the focusing region. It is easy to see that this integral can be expressed in terms of the confluent hypergeometric function $F(\alpha, \gamma, x)$ (see, for example,[2])

$$N = n_a V_f\left\{(1-e^{-x_f}) + \frac{1}{3}\left[F\left(-\frac{3}{2k}, 1-\frac{3}{2k}, -x_f\right)-1\right]\right\}.$$

From this formula it is clear that the dependence $N \sim E^{2k}$ is observed only for $x_f \ll 1$; in this connection

$$N \approx \frac{2k-2}{2k-3}\frac{\pi a^3}{\operatorname{tg}\theta} n_a A\int_0^t E_f^{2k}\,dt \sim E_f^{2k} t,$$

however, for $x_f \geq 1$ the dependence of N on E changes abruptly. Using the asymptotic behavior of the confluent hypergeometric function[2] for $x \gg 1$, we obtain

$$N \approx n_A V_f x_f^{3/2k} \approx n_A V_f (At)^{3/2k} E_0^3 \sim t^{3/2k} E_0^3,$$

i.e., the exponent of the power-law function $N(E_0)$ does not depend on the quantum nature of the process and is usually several times smaller than the number of quanta required for ionization of the majority of atoms by laser radiation ($k \sim 10$). In this connection the yield very weakly depends on the duration of the effect:

$$t^{3/2k} \sim t^{1/10}.$$

In the intermediate region $x_f \sim 0.3$ to 3 the effective exponent of the dependence $N \sim E^{2k_{eff}}$ may take intermediate values, $3/2 < k_{eff} < k$. Let us estimate the values of x at which k begins to change noticeably. If the dependence is given by

$$N \approx C(x - \gamma_2 x^2 + \ldots)$$

in an arbitrary case, then for $x \ll 1$ the deviation of k_{eff} from k is determined by the relation $\delta k/k \approx \gamma_2 x$. In fact, taking the logarithm of both sides of this dependence, we obtain

$$\ln N = \ln C + \ln(x - \gamma_2 x^2 + \ldots);$$

differentiating with respect to E for $x = AE^{2k}$ we have

$$\frac{\delta \ln N}{\delta \ln E} = \operatorname{tg}\psi = 2k_{\text{eff}} = 2k(1-\gamma x), \quad \text{i.e.} \quad \frac{\delta k}{k} \approx -\gamma_2 x.$$

In our case

$$\gamma = \frac{1}{2}\left(1-\frac{1}{4k}\right) \approx \frac{1}{2} \quad \text{for } k \gg 1;$$

therefore $\delta k/k \approx (1/2)x_f$, i.e., even for x_f of the order of tens of percent, appreciable changes of k should be observed.

In concluding this section we note the following general results: the asymptotic form $N \sim x_f^{3/2k} \sim E_f^3$ is characteristic of a conical shape, which one can easily verify, having set $x = x_f(a_f/a)^{2k}$ in the expression

$$N = n_0\int_0^\infty (1-e^{-x})a^2 dz$$

and having cut off the integration over the volume, $N \approx n_0 \int_0^{a_1} a^2 dz$, at an upper limit corresponding to $x_1 \approx 1$, and neglecting the remaining part in view of the abrupt drop in the value of the integrand $1 - e^{-x}$ for $x \ll 1$.

One can interpret the considered effect in a simple fashion. If the field intensity at the focus $E_f \gg E_{cr}$ (where $AE_{cr}^{2k}t \sim 1$, i.e., $E_{cr} \approx 1/(At)^{1/2k}$) and if for a radius r of the cross section $E(r) \approx E_f a_f/r$, then one can obtain the region of strong ionization by assuming $E(r_{max}) \approx E_{cr}$; hence $r_{max} \approx E_f a_f/E_{cr}$ defines the volume of the strongly ionized region:

$$V \approx \frac{1}{2}\pi r_{max}^2 z_{max} \approx \frac{\pi r_{max}^3}{2\operatorname{tg}\theta} \approx \frac{\pi a_f^3}{2\operatorname{tg}\theta} \sim E_f^3 t^{3/2k},$$

in agreement with the results obtained.

This simplified model gave the basic relationships characteristic of multiphoton processes in the focus of a laser. Now let us consider the case of a smoother variation of the field.

2. In the case of a smooth focal constriction, the relation for the radius of the cross section is usually given by

$$a^2(z) = a_f^2 + z^2 \operatorname{tg}^2\theta,$$

where a_f denotes the minimum radius at $z = 0$ and θ is the angle of the focusing cone. Then

$$N = n_0 \cdot 2\pi\int_0^\infty \{1-\exp[-B/a^{2k}(z)]\}a^2 dz, \quad B = Aa_f^{2k}\int_0^t E_f^{2k}\,dt = x_f a_f^{2k}.$$

Introducing the substitution

$$z = a_f \operatorname{tg}\eta / \operatorname{tg}\theta,$$

we find

$$N = \frac{\pi^{3/2}a_f^3 n_a}{\operatorname{tg}\theta}\sum_{n=1}^\infty \frac{(-1)^{n+1}}{n!}x_f^n\frac{\Gamma(kn-3/2)}{\Gamma(kn-1)}.$$

The large value of k permits one to use the asymptotic expression for the Γ function, which gives $N \sim x - (1/2)x^2\sqrt{(2k-4)/(4k-4)} + \ldots$, hence the coefficient $\gamma_2 = 1/2\sqrt{2}$ for $k \gg 1$, i.e., $\delta k/k \approx x/2\sqrt{2}$.

3. Let us consider beams with a smooth radial distribution. A Gaussian distribution $E = E_0(z)e^{-r^2/2a^2(z)}$ is most often assumed, which corresponds to a single-mode distribution of the field in a confocal resonator for a given radius of the cross section

$$a^2(z) = a_f^2 + z^2 \operatorname{tg}^2\theta.$$

First let us calculate the linear yield $\partial N/\partial z$ of events for a given $a(z)$, which coincides with the yield for the frequently used Gaussian distribution of a cylindrical beam:

$$\frac{\partial N}{\partial z} = n_a\int_0^\infty \{1-e^{-y}\}2\pi r\,dr,$$

where

$$x_0(z) = A\int_0^t E_0^{2k}(z)\,dt, \quad y = x_0\exp[-r^2 k/a^2(z)],$$

and $E_0(z)$ is the field on the axis of the beam ($E_0(z) = E_f a_f/a$). Introducing the variable y, we obtain

$$\frac{\partial N}{\partial z}=\frac{\pi n_a a^2}{k}\int_0^{x_0}(1-e^{-y})\frac{dy}{y}=\frac{n_a\pi a^2}{k}\{C+\ln x-\mathrm{Ei}(-x)\}$$

$$=\frac{n_a\pi a^2}{k}\sum_{n=1}^{\infty}\frac{x_0^n(-1)^{n+1}}{n!n}$$

according to the definition of the exponential integral (see, for example,[2]). The asymptotic behavior of this series for $x_0 \gg 1$ follows from the asymptotic expansion of the function $\mathrm{Ei}(-x) \approx e^{-x}/x$, which causes it and the Euler constant C to be small in comparison with the logarithm, i.e.,

$$\frac{\partial N}{dz}\approx n_a\frac{\pi a^2}{k}\ln x_0 \quad \text{for} \quad x_0 \gg 1.$$

For small values of x_0 we have

$$\frac{\partial N}{\partial z}\approx n_a\frac{\pi a^2}{k}\left(x_0-\frac{x_0^2}{4}\right),$$

i.e., for a Gaussian cylindrical beam $\delta k/k \approx (1/4)x_0$. Integration of $\partial N/\partial z$ with respect to z gives

$$N=\frac{n_a\cdot 2\pi a_f^3}{k\,\mathrm{tg}\,\theta}\sum_{n=1}^{\infty}\frac{x_f^n(-1)^{n+1}\Gamma(kn-3/2)}{n!n\Gamma(kn-1)};$$

for $a^2 = a_f^2 + z^2\tan^2\theta$ and $z = a_f \tan\eta/\tan\theta$. The asymptotic form of this expression is the same as that considered above—the result is typical for a conical geometry (the shape of the constriction does not affect the asymptotic behavior).

The obtained results are also applicable for multiphoton breakdown or multiphoton priming of optical cascade breakdown in a beam from powerful pico and nanosecond lasers. (For example, in the case of breakdown propagation of the wavefront for $E_f > E_{\text{threshold}}$, the radius of the spark cone is determined from the relation $a = E_f a_f/E_{\text{threshold}}$, i.e., the length of the spark $z = a/\tan\theta \approx E_f a_f/E_{\text{threshold}}$, $\tan\theta \sim E_f/E_{\text{threshold}}$.)

CONCLUSION

From the above discussion it follows that the spatial and temporal structure of focused laser radiation may substantially change the dependence of N on E because the observed effect is only an averaged, "rounded" result of individual spikes at specific points of the modes for which one may have $x \gtrsim 1$ whereas the average value $\langle x\rangle < 1$. Thus, in order to interpret experiments on multiphoton ionization it is necessary to take into account that a decrease $k_{\text{eff}} < k_0 = I/\hbar\omega$ may occur not only as a result of a broadening of the upper levels or the presence of a resonance, but also how much will occur as a consequence of the effect considered in the present article.

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