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Intensity Effects and Absolute Phase Effects in Nonlinear Laser-Matter Interactions.

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1. Introduction

The generation of short pulses of electromagnetic radiations rely in many cases on nonlinear interactions, and, on the other hand, the resulting sources may have very large intensities, and can induce high-order nonlinear processes. Of course, these two faces of light-matter interactions are intimately connected, but they can usually be treated separately, depending on the emphasize put on one side or on the other. The subject of the present chapter belongs to the physics of „strong-field phenomena" taking place in laser-matter interactions, and it partly concerns quantum optics, too. A brief overview of theoretical methods for treating nonlinear light-matter interaction is given, and some characteristic examples are presented. We review the basic classical and quantum approaches in simple terms, by possibly avoiding involved mathematical derivations. We shall attemp to give a clear conceptual framework and illustrative examples, on the basis of which the main characteristics of the intensity and phase effects showing up in various extreme nonlinear light-matter interactions can be understood. We shall limit the discussion mostly to the single-electron picture, and genuine many-body effects will not be considered. On „strong-field phenomena" we mean here those phenomena whose main characteristics are governed by higher powers than linear of the laser intensity. Two typical examples for such processes are the nonlinear photoelectric effect and high-harmonic generation induced by strong laser fields. The former one has been the subject of an extensive research already from the beginning of the sixties of the last century, starting with the works of Keldish (1965) and Bunkin & Fedorov (1965). As for the later developments, see Farkas (1978), Krause et al. (1992), the book by Delone and Krainov (1994), and a recent review on multiphoton surface photoelectric effect by Ferrini et al. (2009). If one photon energy is not enough to ionize an atom (or deliberate an electron from a metal surface), then, in order to have a non-negligible ionization probability per unit time for many-photon absorptions, one needs a high-power source of the radiation. The high-harmonics of an incoming optical radiation stem from the laser-induced nonlinear polarization of the active constituents of matter, which, may for example be bound dipoles of atoms, molecules, solids or free electons in the conduction band of metals or in beams propagating in free space. Another wide class of strong-field phenomena consists of the laser-assisted processes, like, for instance x-ray scattering on atomic electrons in the presence of a high-power laser radiation. Here, in contrast to the induced processes, the primary process can of course take place without any nonlinearity, but the additional interaction modifies the spectrum of the scattered radiation, due to the appearence of side-bands separated by the laser frequency (see e.g. Puntajer & Leubner, 1990). The relative strengths of these side-bands are nonlinear functions of the laser intensity. In all the above-mentioned examples a considerable spectral broadening of the signals may take place, which results in temporal compression, i.e. short pulses are formed. Of course, the appearance and the quality of these pulses crucially depend on the phase relations of its Fourier components. In general, one may say that the generation of short pulses of electromagnetic radiation are based on nonlinear laser-matter interactions. As for the theoretical description of the mentioned, mostly very high-order processes, the greatest challenge is to work out non-pertubative methods, on the basis of which the interactions with the strong fields are taken into account up to arbitrary orders. This is needed because the perturbation theory may break down for large intensities, even to that extent that terms like 'perturbation' or 'higher-order', in fact, simply loose their usual contents. The optical tunneling offers itself as a good example for such a situation, when the electrons within one optical cycle leave the binding potential which is broken down by the high electric field of the laser radiation (Keldish (1965), Bunkin & Fedorov (1965)). Here one encounters with the fundamental question of wave-particle duality of both the electromagnetic radiation and the electron (see e.g. Büttiker & Landauer (1982) and Chiao (1998)). Another conceptually and also practically important question is to what extent can one control the phase of the ultrashort pulses, and, at all, what are the ultimate limits of phase stabilization in a given generating process? The systematic quantitative analysis of these problems is still missing, at least concerning the quantum uncertainties of the phases. One may expect that the usual high-intensity laser fields, being in a highly populated coherent state, can certainly be well represented in a satisfactory manner in the frame of external field approximation, i.e. in terms classical Maxwell fields of definite amplitudes and phases, or in terms of classical stochastic processes. On the other hand, in the generation of extreme pulses, like sub-femtosecond or attosecond pulses, it is an open question whether the fully quantum or the semiclassical description deliver the correct interpretation.

Within the limited space at our disposal, it cannot be our purpose to give an historical overview, and it would be impossible to review the presently ongoing research on strong-field phenomena. Besides studying the important early works, like Ritus & Nikishov (1979), Faisal (1987), Gavrila (1992), Mittleman (1993) and Delone & Krainov (1994), the interested reader can find further references in recent reviews, dealing with various aspects. We refer the reader e.g. to Brabec & Krausz (2000), Ehlotzky (2001), Agostini & DiMauro (2004), Salamin et al. (2006), Mourou et al. (2006) , Krausz & Ivanov (2009), Ehlotzky et al. (2009) and Gies (2009). In fact, the conceptual framework of the theory of strong-field phenomena has not changed very much in the last couple of decades. The main point in the theory has long been the following. In contrast to the usual texbook approach, where the light (or electromagnetic radiation, in general) is treated as a perturbation, the essence of all new methods is just to turn around this scheme, and consider the strong field as the dominating agent in the interaction. Besides direct numerical, *ab initio* calculations, the analytic approaches still play a very important role, because only this can give an intuitive understanding of the mentioned processes, and help to find the really important parameters to be controlled in the experiments. In the meantime, the laser technology has undergone a very fast development, which made it possibile to generate extreme radiation fields in well-controlled ways. Many processes have become accessible for direct experimental basic research, whose results, at the same time, have many potential applications in other branches of science and technology. We shall not touch this aspects, either. In the present chapter we attemp to give a summary of the characteristics of the simplest basic nonlinear processes taking place in strong-field laser-matter interactions. Based partly on our earlier and recent works, we shall present some typical examples, which may serve as guides towards a possibly intuitive insight into the broad field of research on strong-field light-matter interactions.

In Sections 2 and 3, on the occasion of a summary on nonlinear Thomson scatteing, high-harmonic generation and multiphoton ionization, we introduce the basic parameters (dimensionles intensity parameter, the ponderomotive energy shift, and the Keldish-γ or adiabaticity parameter) which govern the characteristics of the processes under discussion. The carrier-envelope phase effect will be studied in simple terms of classical electrodynamics. We shall briefly discuss the multiphoton analogon of the Kramers-Heisenberg dispersion formula. In Section 4 the nonlinear effect of the laser-induced oscillating double-layer potential on metal surfaces and the x-ray generation in the presence of a static homogeneous electric field will be touched. Here we shall introduce a new nonlinearity parameter, that seems to be a key element in the theoretical analysis of many recent experiments showing unexpectedly high laser-induced nonlinearities. We shall briefly discuss the question of optical rectification and the generation of quasi-static wake-fields, whose polarization can be manipulated by changing the carrier-envelope phase difference. Besides, the analogy between the usual Fourier-synthesis of the high-order harmonics stemming from a nonlinear scattering, and the interference of above-threshold electronic de Broglie wave components shall be emphasized. This latter effect may give a possibility for generation high-current attosecond electron pulses. In Section 5 some quantum statistical properties of short electromagnetic radiation pulses will be analysed.

2. Classical description of basic strong field phenomena. Carrier-envelope phase difference effects in ultrashort pulse interactions

In the present section we shall summarize the most important elements of the conceptual framework in which nonlinear laser-matter interactions can be theoretically considered. At the same time, this part serves as an introduction of the important parameters in terms of which the particular natures of these various processes can be quantified. In Table 1. we show the possible combinations of theories in the descriptions of photon-electron interactions. For short, on the word 'photon' we mean electromagnetic radiation in general.

The most fundamental illustrative cases are the nonlinear Thomson and Compton scattering, which in the cleanest way show some general basic characteristics of the nonlinear photon-electron interactions (see e.g. Brown & Kibble (1964), Leubner (1978, 1981), Bergou & Varró (1981b), Gao (2004), Lan et al. (2007), Boca & Florescu (2009), Mackenroth et al. (2010)). According to the description displayed in cell No. 2 in Table 1., consider the classical equation of motion of a point electron of charge $-e$ and rest mass m interacting in vacuum with a linearly polarized plane wave of electromagnetic radiation:

$$m\mathrm{d}(\gamma\mathbf{v})/\mathrm{d}t=-e[\mathbf{E}_0+(\mathbf{v}/c)\times\mathbf{B}_0],\quad \mathbf{E}_0=\varepsilon_0 F(\eta),\quad \eta\equiv t-\mathbf{n}_0\cdot\mathbf{r}(t),\quad \mathbf{B}_0=\mathbf{n}_0\times\mathbf{E}_0\,, \tag{1a}$$

$$F(\eta)=-\partial^2\Pi(\eta)/c^2\partial t^2,\quad \Pi(\eta)=(c/\omega_0)^2F_0f_0(\eta)\cos(\omega_0\eta+\varphi_0),\quad f_0(\eta)=g_0(\eta)\equiv\exp(-\eta^2/2\tau_0^2), \tag{1b}$$

Photon / **Electron**	Trajectory, Rays (Geometric Optics)	Field (Maxwell Theory)	Quantized Field (Photon Picture)
Trajectory, Current (Point Mechanics)	1.	2. Classical Electron Theory	3.Classical Current, Poisson Photon
Field, Current (Wave Mechanics)	4.	5. Semiclassical Description	6.Quantum Current, General Photon
Quantized Field (Negaton-Positon)	7.	8.	9. Quantum Electrodynamics

Table 1. Possible descriptions of photon-electron interactions. The most widely applied descriptions are displayed in cells Nos. 2. and 5. Concerning the electron motion, in each cells the relativistic description is also included. The cells Nos. 1., 4. and 7. are empty, because the direct interaction of point charged particles with rays of radiation are not considered.

where $\mathbf{v}(t)=d\mathbf{r}(t)/dt$ and c are the velocity of the electron and of the light field in vacuum, respectively, and $\gamma=[1-(v/c)^2]^{-1/2}$ is the usual relativistic factor. The amplitude of the electric field strength, the polarization vector and propagation vector are denoted by F_0, ε_0 and $\mathbf{n}_0$, respectively. In Eq. (1b) we have introduced the Hertz potential $\Pi(\mathbf{r},t)=\Pi(\eta)$, and the special case of a quasi-monochromatic wave of mean circular frequency $\omega_0=2\pi/T_0$ having a pulse envelope function $f_0(\eta)$, and the *carrier-envelope phase difference* (CEPD) φ_0. This latter quantity is often simply called *absolute phase*. In general, the pulse envelope function $f_0(\eta)$ has its maximum value 1 at $\eta=0$, and vanishes for $\eta\to\pm\infty$ (as an example, a Gaussian envelope function g_0 of full width at half maximum (FWHM) $2\tau_0(\log 2)^{1/2}$ is shown in the last equation of Eq. (1b)). The first equation of Eq. (1a) is a highly nonlinear second order ordinary differential equation for the electron's position $\mathbf{r}(t)$, because through the variable η the electromagnetic field strengths in their arguments contain this trajectory in a complicated manner. Thus, in general, we do not expect a simple harmonic motion, even if we set the envelope function constant ($f_0=1$), when we are dealing with an ideal monochromatic plane wave. This special case was first studied long ago by Halpern (1924) in the context of a possible description of Compton scattering. Later it turned out that the seemingly complicated equation in Eq. (1a) can be solved analytically for an arbitrary functional form of $f(\eta)$. It can be shown that $d\eta/d\tau=\gamma(1-\mathbf{n}_0\cdot\mathbf{v}/c)$ is a constant of motion (where $d\tau=dt/\gamma$ denotes the proper time element) thus, at the position of the electron the complete argument η of the wave is a linear function of the proper time of the electron. As an example, take an x-polarized wave ($\varepsilon_0=(1, 0, 0)$) propagating in the positive z-direction ($\mathbf{n}_0=(0, 0, 1)$). Then the equation of motion in Eq. (1a) can be brought to the set of equations

$$ma\,d^2x/d\eta^2=-eF(\eta),\quad a\equiv\gamma(1-v_z/c)=\text{const.},\quad d^2z/d\eta^2=(d/2c\,d\eta)(dx/d\eta)^2,\quad d^2y/d\eta^2=0, \tag{2a}$$

$$\gamma mc^2=mc^2+m(dx/d\eta)^2/2. \tag{2b}$$

The first (still exact) equation in Eq. (2a) is completely analogous to a simple Newton equation in a homogeneous time-varying field, which can be immediately integrated, and then, from the third equation, the function $z(\eta)$ can be determined. In Eq. (2b) we have shown the total energy of the electron, which naturally splits to the sum of the rest energy and a term analogous to the nonrelativistic kinetic energy. One has to keep in mind, however, that the trajectory still cannot be expressed by the true time t in a closed form. It is possible to give analytic expressions for $t(\eta)$, $x(\eta)$, $z(\eta)$ as functions of the retarded time η (or, equivalently, of the proper time τ). The „initial data" on the light-like hyperplane η=const. cannot directly correspond to true initial values, because the prescription $t-z/c$=const. cannot account for a true separation of the particles from a real interaction region, since this should be specified on the space-like plane t=const. (in order to secure causality). Concerning the solution of this problem see Varró & Ehlotzky (1992). In the *nonrelativistic limit* ($v/c \ll 1$, $\gamma\to1$, $a\to1$), in *dipole approximation* ($\eta\to t$) and *in a stationary field* we have the simple results

$$m d^2x/dt^2=-eF_0\cos(\omega_0 t+\varphi_0),\quad dx/dt=-v_{osc}\sin(\omega_0 t+\varphi_0)+v_0,\quad x=x_{osc}\cos(\omega_0 t+\varphi_0)+v_0 t+x_0, \tag{3a}$$

$$v_{osc}/c=\mu_0,\quad x_{osc}=\mu_0\lambda_0/2\pi,\quad \mu_0\equiv eF_0/mc\omega_0,\quad \mu_0=0.8528\times10^{-9}I_0[\mathrm{W/cm^2}]^{1/2}\lambda_0[\mu\mathrm{m}], \tag{3b}$$

$$\mu_0= 1.0426\mathrm{x}10^{-9}\,[I_0\,/(\mathrm{W/cm^2})]^{1/2}\,/\,[h\nu_0\,/\mathrm{eV}],\quad \mu_0=(I_0/I_c)^{1/2},\quad I_c=cE_c^2/8\pi,\quad E_c=2\pi e/r_0\lambda_0, \tag{3c}$$

where v_{osc} and x_{osc} are the velocity and coordinate amplitudes, respectively, of the electron oscillating in the laser field of intensity I_0 (measured in $\mathrm{W/cm^2}$) and of central wavelength $\lambda_0=2\pi/k_0=2\pi c/\omega_0=c/\nu_0$ (measured in microns, $\mu\mathrm{m}=10^{-4}\mathrm{cm}$). The *dimensionless intensity parameter* μ_0 introduced in the third equation of Eq. (3b) is the ratio of the amplitude of the velocity oscillation to the velocity of light. Though μ_0 is a classical parameter, in Eq. (3c) we have expressed it in terms of the associated photon energy $h\nu_0$, too, where h denotes the Planck constant. Besides, we have introduced the critical intensity I_c (for which $\mu_0=1$) associated to the critical field strength E_c, where $r_0=e^2/mc^2=2.818\times10^{-13}$cm is the classical electron radius. The work $eE_c\lambda_C/2\pi=h\nu_0$ done on the electron by this critical electric field (assumed here static) along the Compton wavelength $\lambda_C=h/mc=2.426\times10^{-10}$cm just equals the central photon energy $h\nu_0$. According to Eq. (3c), for a laser field of mean photon energy $h\nu_0$=1eV the critical intensity is $I_c=10^{18}\mathrm{W/cm^2}$. The parameter μ_0 quantifies the limits of validity of the approximations just applied. If μ_0 approaches unity, then v_{osc} gets close to the velocity of light and the nonrelativistic description is not valid any more. At the same time, the amplitude of the position oscillations becomes comparable with the wavelength, and the dipole approximation breaks down, too, thus the term $\mathbf{n}_0\cdot\mathbf{r}/c$ in the retarded time η cannot be neglected in the equation of motion. Just this kind of term is responsible for the generation of higher-order harmonics by a free electron in the presence of a strong laser field. In the nonrelativistic regime, under the action of the laser field, a pure harmonic oscillation is superimposed on the free inertial motion of the electron. According to classical electrodynamics (Jackson (1962)), for this *simple harmonic* motion the average power radiated per unit solid angle in the nth *higher-harmonic* is:

$$dP_n/d\Omega=(e^2\omega_0^2/2\pi c)(n\tan\theta)^2[\,J_n(\mu_0 n\cos\theta)\,]^2,\quad (e^2\omega_0^2/2\pi c)=\alpha(h\nu_0/T_0),\quad \alpha\equiv 2\pi e^2/hc, \tag{4}$$

where J_n denotes ordinary Bessel function of first kind of order n (see e.g. Gradshteyn and Ryzhik (2000)), and θ is the angle between the propagation direction of the emitted radiation with respect to the direction of oscillation (here: x-axis). In order to make the numerical estimates easier, in Eq. (4) we have also introduced the Sommerfeld fine structure constant $\alpha\approx1/137$. For instance, for a Ti:Sapphire laser the total power emitted (exclusively to the fundamental frequency) by a single electron becomes $\sim\alpha\mu_0^2(h\nu_0/T_0)\approx\alpha\mu_0^2\times10^{-4}$ Watt. This is because in the nonrelativistic case under discussion, we have $\mu_0 \ll 1$, i.e. the argument of the

Bessel function is much smaller than its order n = 1, 2, …, and the approximation $J_n(nz) \approx (nz/2)^n/n! \approx (z/2e)^{-n}$ is essentially exact. The ratio of the partial powers of the consecutive harmonics is to a good accuracy $(dP_{n+1})/(dP_n) \approx (\mu_0\cos\theta/2e)^2 <<< 1$, thus practically no harmonics are produced, except for the fundamental one. This simple example shows that the higher-harmonic generation on a free electron is an inherently relativistic effect.

In order to illustrate intensity and absolute phase effects in the relativistic regime, we present the solution of the initial value problem of the general equation in Eq. (2) for a stationary field $F(\eta)=F_0\cos(\omega_0\eta+\varphi_0)$. First, let us specify the position and velocity of the electron at the time instant t=0, in the case when $\varphi_0=0$, such that $x(0)=(dx/dt)(0)=z(0)=(dz/dt)(0)=0$, thus $\eta(0)=0$. In accord with an early work by Halpern (1924), on x-ray scattering, we obtain

$$x=\mu_0(\lambda_0/2\pi)[1-\cos(\omega_0\eta)],\quad z=(\mu_0/2)^2(\lambda_0/4\pi)[2\omega_0\eta+\sin(2\omega_0\eta)],\quad 2\omega t=2\omega_0\eta+\beta\sin(2\omega_0\eta), \tag{5a}$$

$$\omega\equiv \omega_0/[1+(\mu_0/2)^2],\quad \beta\equiv (\mu_0/2)^2/[1+(\mu_0/2)^2] < 1,\quad z(t)=\beta ct-(\lambda_0/2\pi)\Sigma_k[J_k(k\beta)/k]\sin 2k\omega t. \tag{5b}$$

The summation with respect to k in the last equation of Eq. (5b) runs through all the harmonic indeces k=1, 2, …, and J_k denotes the ordinary Bessel function of first kind of order k. From the parametric representations of $x(\eta)$ and $z(\eta)$ in Eq. (5a) one can immediately derive the well-know *„figure-8 shape"* for the electron trajectory if one transforms out the uniform motion in the longitudinal z-direction (propagation direction of the ideal laser field). Of course, the modulus of the velocity components are smaller than c for arbitrary high intensities. On the other hand, the energy in Eq. (2b) becomes $\gamma mc^2=mc^2[1+(\mu_0/2)^2(1-\cos(2\omega_0\eta)]$, where the term $(\mu_0/2)^2mc^2\equiv U_p$ is usually called *ponderomotive energy shift*. The velocity of the longitudinal *drift motion* βc approaches c from below in the extreme case when $\mu_0 >>> 1$, even if the initial velocity is zero. The frequency ω of the electon's fundamental oscillation in the laboratory frame is down-converted by a factor of $[1+(\mu_0/2)^2]$, but, on the other hand, the „renormalized spectrum" becomes very broad for large enough intensities. Really, by applying the asymptotic formula 8.455.1 of Gradshteyn & Ryzhik (2000), $\pi J_n(x) \approx [2(n-x)/3x]^{1/2}K_{1/3}\{[2(n-x)]^{3/2}/3x^{1/2}\}$, where $K_{1/3}$ denotes the modified Hankel function. With the help of this result we have $[J_n(n\beta)]^2 \sim \exp(-n/n_c)$ with $n_c = (3/8)^{1/2}(\mu_0/2)[1+(\mu_0/2)^2] >>> 1$ is the *critical harmonic number*. As for the longitudinal motion under discussion, the cut-off frequency $\omega_c=n_c\omega$ is of the order of $\mu_0\omega_0$, which may reach the x-uv region of the electromagnetic spectrum. For example, if μ_0=60 we have for the critical number $n_c \approx 16500$, and the spectrum starts from $\omega\approx \omega_0/900$ and ends around $\omega\approx 30\times\omega_0$, covering terahertz and uv radiations as well.

The explicit calculation becomes much more complicated if the phase φ_0 is nonzero. The qualitative difference in this case is that, besides the oscillation around the position x=0, the transverse motion (along the polarization) aquires a drift $x_1(\sin\varphi_0)t$, too. The longitudinal motion is similar to that we have encountered in Eq. (5b), but now the drift part depends on the initial phase:

$$z(t)=\beta c(1+2\sin^2\varphi_0)t-(\lambda_0/2\pi)\Sigma_k(A_k\cos 2k\omega' t+B_k\sin 2k\omega' t), \tag{6}$$

where the Fourier coefficients A_k and B_k are combinations of Bessel functions, whose explicite form is not interesting here. Instead of proceeding further with the analytic formulae, we give in Figs. 1a-d and Figs. 2a-b some typical trajectories of an electron interacting with a strong laser pulse.

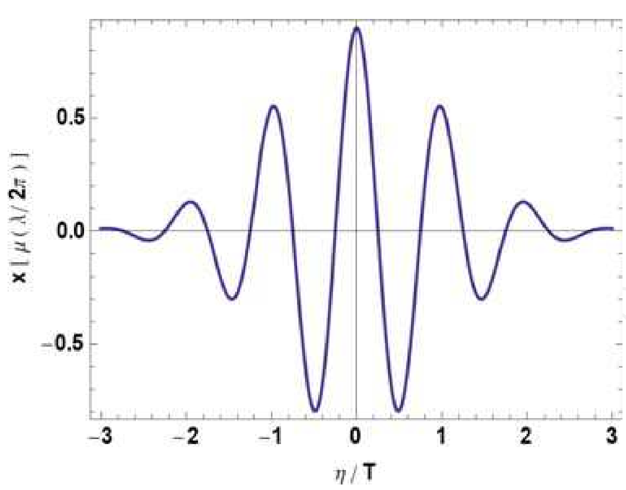

Fig. 1a. Shows the parametric variation of the electron's position along the polarization vector of the strong two-cycle laser field. We have taken a Gaussian pulse displayed in the last equation of Eq. (1b) with φ_0=0, and the intensity parameter was set μ_0=0.9, corresponding to the critical intensity $I_0 \approx 10^{18}$W/cm^2 for an optical field with $h\nu_0$=1eV.

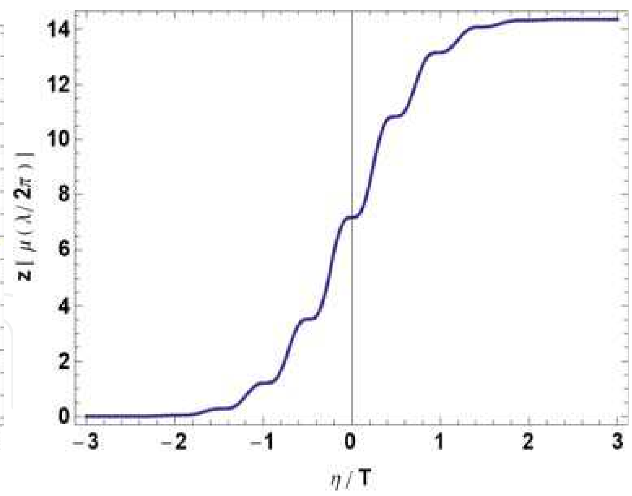

Fig. 1b. Shows the parametric variation of the electron's position along the propagation vector of a strong two-cycle laser field. This figure shows that the longitudinal motion has a systematic drift, whose velocity is essentially βc, as has been given in Eq. (5b). The parameters are the same as in Fig. 1a.

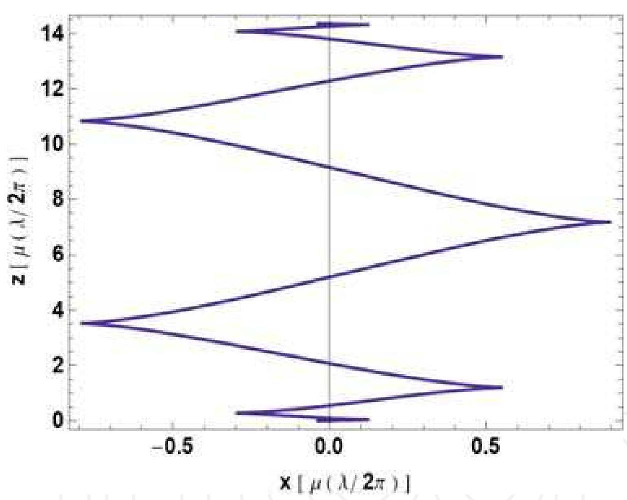

Fig. 1c. Shows the parametric variation of the electron's position along the polarization vector of the strong two-cycle laser field. We have taken a Gaussian pulse displayed in the last equation of Eq. (1b) with φ_0=0, and the intensity parameter was set μ_0=0.9, corresponding to the critical intensity $I_0 \approx 10^{18}$W/cm^2 for an optical field with $h\nu_0$=1eV.

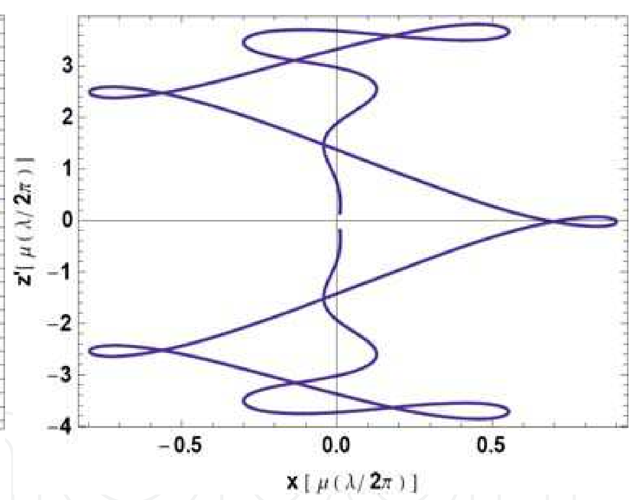

Fig. 1d. Shows the same as Figure 1c, but now the essentially uniform drift motion has been subracted from the longitudinal motion, thus the motion of the electron is viewed from a comoving frame. The coordinate z′ has been defined as $z' \equiv z - (\mu_0/2)^2(\lambda_0/4\pi)\times 2\omega_0\eta$, according to Eq. (5a). The curve starts slightly below the origin, towards negative z′ values, and then proceeds to positive values and finally gets back to the origin from above.

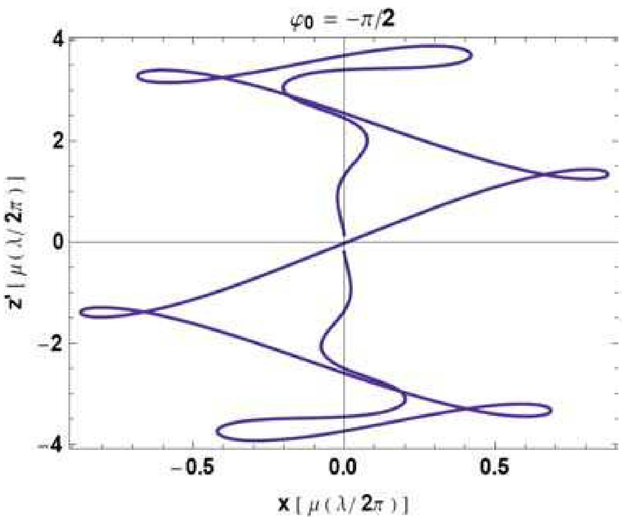

Fig. 2a. Shows the same as Fig. 1d for the reduced electron trajectory, but now the carrier envelope phase difference of the Gaussian pulse has been taken $\varphi_0=-\pi/2$, i.e. now we are dealing with a sine-pulse. The curve starts slightly below the origin, towards negative z′ values, and then proceeds to positive values and finally gets back to the origin from above.

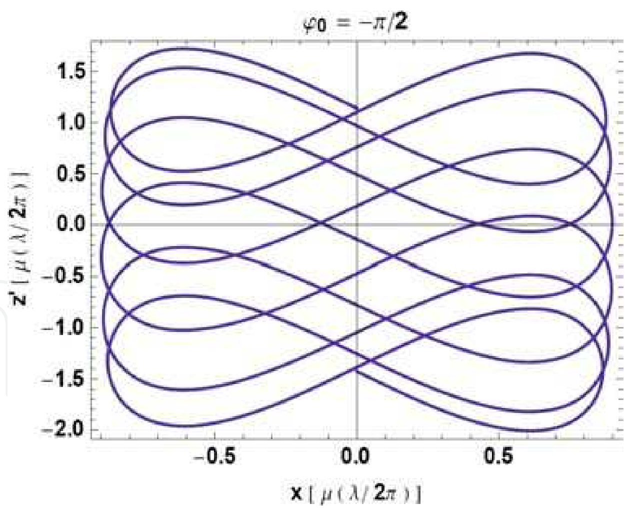

Fig. 2b. Shows the reduced electron trajectory around the maximum of a relatively long (~ 20 cycle) Gaussian sine-pulse (when $\varphi_0=-\pi/2$). The nearly uniform drift motion has not been transformed out completely, in order to illustrate that the „figure-8 trajectory" is never a stationary lemniscate. The curve starts at the point (0,−1.5) towards negative z′ values, and then proceeds to positive values and finally arrives at the ponit (0,+1.2).

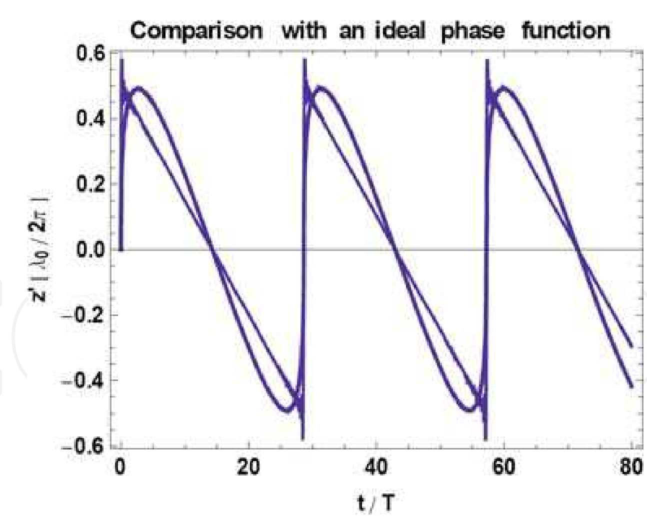

Fig. 3. The real time-dependence of the longitudinal position $z'(t)\equiv-[z(t)-\beta ct]$ of the electron as given in Eq. (5b), where we have subtracted the uniform drift motion $\beta c t$. The trajectory is compared with the ideal phase function („saw-tooth"), which is given by the famous Fourier series $\Sigma_k(\sin kx)/k=(\pi-x)/2$ on the open interval $(0, 2\pi)$. From the first equation of Eq. (5b), the „saw-tooth" is represented by the sum $\Sigma_k[\sin(2k\times0.11(t/T))]/k$. which corresponds to the $\mu_0=15$ value of the dimensionless intensity parameter.

In order to consider the general role of the ponderomotive energy shift $(\mu_0/2)^2mc^2=U_p$ introduced after Eq. (5b), we write the relativistic equation of motion in a covariant form, which is equivalent to Eq. (1a) and the relativistic work theorem. From Eq. (1a) we obtain $m d\mathbf{u}/d\tau=(e/c)(u_0\mathbf{E}+\mathbf{u}\times\mathbf{B})$ and $m du_0/d\tau=(e/c)\mathbf{u}\cdot\mathbf{E}$, where $(u_0, \mathbf{u})=\{u^\mu\}=\{dx^\mu/d\tau\}$ is the four-velocity, $\{x^\mu\}=(ct, \mathbf{r})$ is the four-position, and $d\tau=dt/\gamma$ denotes the proper time element of the electron. We adopt the signature (+ — — —), and then the metric tensor g becomes $g_{00}=-g_{ii}=1$ for i=1, 2, 3, and the off-diagonal elements are zero, i.e. $g_{\mu\nu}=0$ for $\mu\neq\nu$, for μ, ν =0, 1, 2, 3. With the usual summation convention, the four-product of two vectors $\{a^\mu\}$ and $\{b^\nu\}$ reads $a\cdot b\equiv g_{\mu\nu}a^\mu b^\nu=a^\mu b_\mu=a^0b^0-\mathbf{a}\cdot\mathbf{b}$, and the gradient vector is defined as $\partial\equiv\{\partial_\mu\}\equiv\{\partial/\partial x^\mu\}$. With these notations the covariant equation of motion of the electron in the laser field becomes:

$$du_\mu/d\tau=(e/mc)F_{\mu\nu}u^\nu\ ,\ \ F_{\mu\nu}=\partial_\mu A_\nu-\partial_\nu A_\mu\ ,\quad du_\mu/d\tau=-(e/mc)dA_\mu/d\tau+(e/mc)u^\nu(\partial_\mu A_\nu), \qquad (7a)$$

$$u_\mu=V_\mu-(e/mc)A_\mu\ ,\ \ dV_\mu/d\tau=(e/mc)V^\nu(\partial_\mu A_\nu)-(e/mc)^2A^\nu(\partial_\mu A_\nu), \qquad (7b)$$

where we have introduced the field tensor $F_{\mu\nu}$, the vector potential A_μ and the part V_μ of the four velocity (soon we shall assume that this is a „slowly-varying" quantity). We emphasize that at this stage Eqs. (7a-b) do not contain any approximations, the above manipulations do not rely on the assumption that V_μ is slowly-varying on any space-time scale. If we take the average over the oscillations of the vector potential in the second equation of Eq. (7b), then, because $\langle V_\mu\rangle=\langle u_\mu\rangle$, we have

$$d\langle V_\mu\rangle/d\tau=-(e/mc)^2\partial_\mu\langle A^\nu A_\nu\rangle\ ,\ \ m d\langle u_\nu\rangle/d\tau=\partial_\nu[\mu^2(\mathbf{r},t)mc^2/2],\ \ m d\langle\mathbf{v}\rangle/dt=-mc^2\nabla\mu^2(\mathbf{r})/2. \quad (7c)$$

In Eq. (7c) we have introduced the generalized intensity parameter μ , in terms of which the ponderomotive energy is expressed as $U_p(\mathbf{r}, t)= \mu_0^2 f(\mathbf{r}, t)(mc^2/4)$, where $\mu_0\equiv eF_0/mc\omega_0$ is the maximum value of the dimensionless intensity parameter, and $f(\mathbf{r}, t)$ is a (slowly-varying) spatio-temporal intensity profile function of the laser beam. The third equation in Eq. (7c) is a (nonrelativistic) special case of the second, more general equation. Here the ponderomotive energy is represented by a conservative (static) potential. It is important to note that, regardless of the sign of the charge of a particle, the ponderomotive potential of the laser field is repulsive towards the lower intensity values (because it is proportional with the square of the charge and with the negative gradient of the intensity distribution). This means, for instance, that an ionized electron experiences a force pushing it out of the interaction region. As a consequence, for instance, the measured energy spectrum of the ionized electrons are different to that they have their „place of birth" inside the laser beam. Since $2mc^2$=1MeV, the maximum energy increase may be of this order if $\mu_0\approx2$, i.e. if the maximum intensity $I_0\approx2\times10^{18}$ W/cm^2 for an optical field ($h\nu_0\approx$1eV). This kind of ponderomotive energy increase is still considered as a way towards laser acceleration of charged particles. It is clear that even at nonrelativistic intensities, the ponderomotive energy shift may considerably be larger than the photon energy. For an optical field of 2×10^{13} W/cm^2 intensity the ponderomotive energy shift is $U_{pmax}\approx$10eV.

At the end of the present section we present a simple classical example to illustrate the possibility of linear absolute carrier-envelope phase difference effects. Let us take the Newton equation of a charged particle bound in a linear potential, and driven by a laser pulse represented by a Gaussian envelope, and study the Fourier spectrum of its position:

$$d^2x/dt^2 + \Gamma dx/dt\ + \Omega^2x=-(e/m)F_0\exp(-t^2/\tau^2)\cos(\omega_0 t+\varphi_0) =-(e/m)F(t), \qquad (8a)$$

$$x(\omega)|^2 = (e/m)^2|F(\omega)|^2/[(\omega^2 - \Omega^2) + \Gamma^2], \quad |F(\omega)|^2 = |C(\omega)|^2[\cosh(2\tau^2\omega\omega_0) + \cos(2\varphi_0)], \quad (8b)$$

$$M(\omega) \equiv [(|x(\omega)|^2)_{max} - (|x(\omega)|^2)_{min}]/[(|x(\omega)|^2)_{max} + (|x(\omega)|^2)_{min}] = 1/\cosh(2\tau^2\omega\omega_0), \quad (8c)$$

where the subsidiary quantity $C(\omega)$ introduced in Eq. (8b) does not depend on φ_0 , its detailed form is unimportant here. In Eq. (8c) we have defined the *modulation function* $M(\omega)$, which is an analogon of the *visibility* used in optics. According to Figs. 4a-b-c, the modulation is negligible except for the very low-frequency part of the spectrum, where it can be considerable if $\omega/\omega_0 < 0.2$ for a three-cycle pulse, for instance. However, this is just that region where the signal (in the present case; the induced dipole) is very small. In the

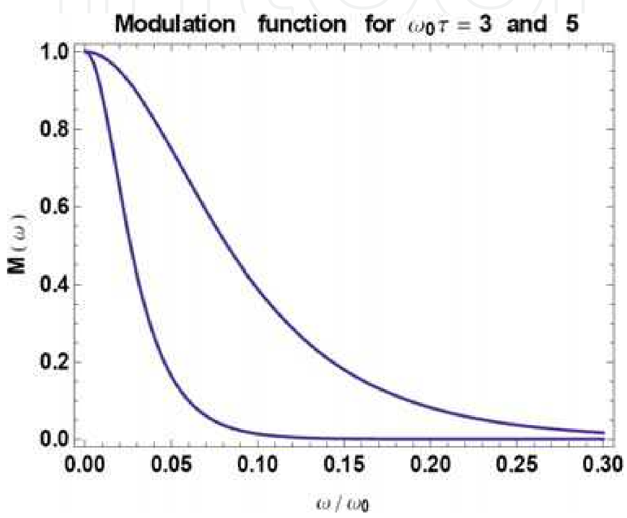

Fig. 4a. Shows the linear modulation function for two few-cycle Gaussian pulses according to the formula given by Eq. (8.c). The upper and the lower curves refer to the values $\omega_0\tau$=3 and $\omega_0\tau$=5, respectively.

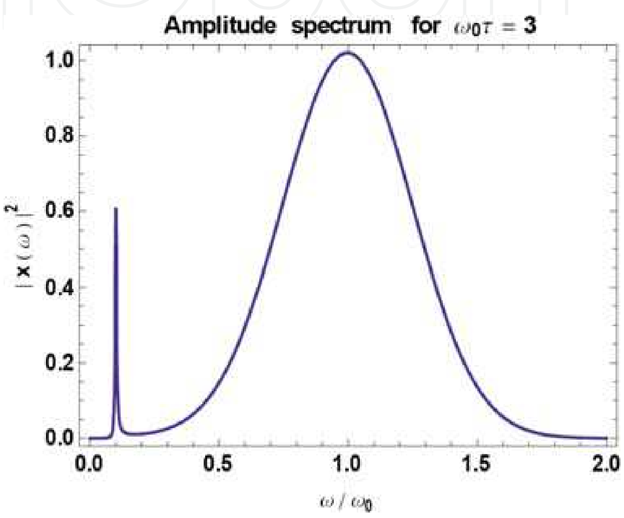

Fig. 4b. Shows the amplitude (dipole) spectrum of the linear oscillator (see Eq. (8b)) with eigenfrequency $\Omega = 0.1\omega_0$ and damping constant $\Gamma = 0.005\omega_0$.

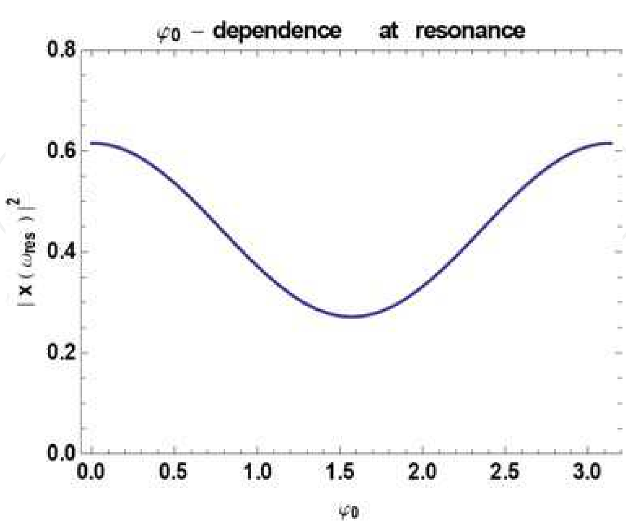

Fig. 4c. Shows the variation of the resonant value of the modulus square of the resonant dipole moment (at frequency $\omega = \Omega = 0.1\omega_0$) as a function of the carrier-envelope phase difference φ_0 , according to Eq. (8b).

linear regime, the spectrum of the response follows the spectrum of the excitation according to this simple model, as is shown in Fig. (4b). If the system has a low-frequency resonance, then both the signal and the modulation become large enough, as can be seen in Figs. 4b-c.

3. Multiphoton ionization and the multiphoton Kramers–Heisenberg formula for high-harmonic generation.

The usual dispersion relations to light scattering are derived in the linear regime, where the incoming radiation is treated as a weak perturbation in comparison of the atomic binding potential. By restricting the present analysis to nonrelativistic intensities, we start with the Schrödinger equation of a single electron interacting jointly with the atomic potential $V(\mathbf{r})$, with the laser field, characterized by the dipole term $e\mathbf{r}\cdot\mathbf{F}(t)$, and with the spontaneously emitted radiation, whose effect is also described by a dipole term of the form $e\mathbf{r}\cdot\mathbf{F}'(t)$:

$$[\mathbf{p}^2/2m + V(\mathbf{r}) + e\mathbf{r}\cdot\mathbf{F}(t) + K(\mathbf{r},t)]\,|\Psi(t)\rangle = i\hbar\partial_t\,|\Psi(t)\rangle, \quad \mathbf{F}(t)=\varepsilon F_0(t)\cos(\omega_0 t+\varphi_0), \tag{9a}$$

$$K(\mathbf{r},t)= e\mathbf{r}\cdot\mathbf{F}'(t), \quad \mathbf{F}'(t)=i\boldsymbol{\varepsilon}'(2\pi\hbar\omega'/L^3)^{1/2}[a\exp(-i\omega' t)-a^\dagger\exp(+i\omega' t)], \tag{9b}$$

where $\mathbf{p}=-i\hbar\partial/\partial\mathbf{r}$ is the electron's momentum operator and L^3 is the quantization volume. The frequency, the polarization vector and the quantized amplitude of the emitted photon are denoted by ω', $\boldsymbol{\varepsilon}'$ and a, respectively. The quantized amplitudes satisfy the usual commutation relation $[aa^\dagger - a^\dagger a]=1$. The system starts at some initial time t_0 in the product state $|\Psi(t_0)\rangle=|\Psi_0\rangle=|\psi_0\rangle|0'\rangle$, and evolves, according to the above Schrödinger equation, to a final state $|\Psi_0(t_1)\rangle$. Here $|\psi_0\rangle$ is the initial electron state, and $|0'\rangle$ symbolizes the vacuum state, where the prime refers to the scattering mode that is initially empty. If we neglect the spontaneous emission during the interaction, then Eq. (9a) can describe multiphoton excitation or ionization, or multiphoton direct or inverse Bremsstrahlung processes. Following the general method due to Varró & Ehlotzky (1993), we can go over to the Kramers-Henneberger frame, and receive a integral equation for the evolution operator of the complete system. By projecting to a final state $\langle\Psi_1|$, we obtain for the S-matrix elements:

$$\langle\Psi_1|\Psi_0(t_1)\rangle=\delta_{10}-(i/\hbar)\int dt\langle\Psi_1|U_0^+(t)W(t)U_0(t)|\Psi_0\rangle$$

$$-(i/\hbar)^2\int dt\int dt'\langle\Psi_1|U_0^+(t)W(t)U_0(t)\,U_0^+(t')W(t')U_0(t')|\Psi_0\rangle+\langle\Psi_1|R_0(t_1)\rangle, \tag{10a}$$

where $U_0(t)$ denotes the unperturbed propagator of the electron, and $|R_0(t_1)\rangle$ represents the third iteration of the integral equation. In deriving Eq. (10a) we have used that $\langle\Psi_1|$ and $|\Psi_0\rangle$ are orthogonal eigenstates of the unpertubed Hamiltonian. The effective interaction represented by the operator $W_\alpha(t)$ has the form

$$W(t)\equiv V_{cl}(\mathbf{r},t)+ K(\mathbf{r}+\boldsymbol{\alpha},t), \quad V_{cl}(\mathbf{r},t)\equiv V(\mathbf{r}+\mathbf{r}_{cl}(t))-V(\mathbf{r}), \quad m d^2\mathbf{r}_{cl}(t)/dt^2=-e\mathbf{F}(t), \tag{10b}$$

where $\mathbf{r}_{cl}(t)$ is the classical position of the electron satisfying the Newton equation. According to Eqs. (3a-c), for a stationary field with $\varphi_0=0$ (or in other words, for smooth switching–on and –off of the laser field) the trajectory simply reads $\mathbf{r}_{cl}(t)=\boldsymbol{\alpha}(t)\equiv\alpha_0\varepsilon_z\sin\omega_0 t$, $\alpha_0\equiv\mu_0\lambda_0/2\pi$. Owing to the periodic modulation of the space-translated atomic potential $V_{cl}(\mathbf{r},t)$, already the first nontrivial term on the right hand side of Eq. (10a) contains all the higher harmonics of the incoming laser radiation:

$$V_{cl}(\mathbf{r},t)=\Sigma_n V_n(\mathbf{r})\exp(-in\omega_0 t), \qquad V_n(\mathbf{r})=\Sigma_l V_n^{(l)}(r)P_l(\cos\theta), \tag{11}$$

where $P_l(\cos\theta)$ is the l-th Legendre polynomial and θ is the polar angle of the electron's position with respect to the polarization vector of the laser field. The detailed form of the „multipole-multiphoton expansion coefficients" $V_n^{(l)}(r)$ have been presented by Varró & Ehlotzky (1993).

Before entering into a short analysis of high-harmonic generation (HHG) in the frame of our semiclassical method, a brief summary may be in order on the famous model by Keldish (1965) on the *multiphoton ionization* and the *optical tunneling* or *cold emission of electrons induced by strong laser fields* (see Bunkin & Fedorov (1965), Farkas et al. (1983,1984), Chin et al. (1983), Farkas & Chin (1984) and Walsh et al. (1994)). Leaving out the interaction term K responsible for the spontaneous emission of radiation, the second term on the right hand side in Eq. (10a) can be approximated in the following way:

$$T_{fi}^{(1)} = -(i/\hbar)\int dt\, \langle \mathbf{p}\,|\, U_p^\dagger(t)U_2^\dagger(t)U_r^\dagger(t)V(\mathbf{r})\,|\,\psi_0\rangle \exp[(i/\hbar)(p^2/2m-E_0)t], \tag{12a}$$

$$U_r(t)=\exp[(ie/\hbar\omega_0)F_0\mathbf{r}\cdot\boldsymbol{\varepsilon}\cos(\omega_0 t)], \quad U_p(t)=\exp[-(i/\hbar)\mathbf{p}\cdot\alpha_0\boldsymbol{\varepsilon}_z\sin\omega_0 t], \tag{12b}$$

$$U_2(t)=\exp[-(i/\hbar)\Delta E_2 t - i(\Delta E_2/2\hbar\omega_0)\sin(2\omega_0 t)], \qquad \Delta E_2=\mu_0^2 mc^2/4, \tag{12c}$$

$$T_{fi}^{(Keldish)} = -(i/\hbar)\int dt \int d^3r\, \psi^*_{\mathbf{p}}(\mathbf{r},t)V(\mathbf{r})\psi_0(\mathbf{r},t) = -2\pi i \Sigma_n T_{fi}^{(n)}\delta[E_p-(E_0-\Delta E_2+n\hbar\omega_0)], \tag{12d}$$

where $\mu_0 \equiv eF_0/mc\omega_0 = 10^{-9}I_0^{1/2}/E_{ph}$ (with $E_{ph}\equiv\hbar\omega_0/\mathrm{eV}$ being the photon energy measured in eV) is the same dimensionless intensity parameter which has already been introduced in Eq. (3c) in the course of the classical analysis. We have also encountered the quantity $\Delta E_2=\mu_0^2 mc^2/4=U_p$, which is just the ponderomotive energy shift, defined after Eq. (5b). In the Keldish model the final state of the electron is taken to be a Volkov state $\psi_{\mathbf{p}}(\mathbf{r}, t)$ of some average energy $E_p= p^2/2m$, which is a dressed state with respect to the interaction with the laser field. In the initial state the interaction with the laser is not incorporated, so it is an unperturbed atomic ground state $\psi_0(\mathbf{r}, t)=\psi_0(\mathbf{r})\exp[-(i/\hbar)E_0 t]$. The explicit form of a nonrelativistic Volkov state reads (where $\mathbf{A}(t)$ is the vector potential of the field $\mathbf{F}(t)$):

$$\psi_{\mathbf{p}}(\mathbf{r},t)=\langle \mathbf{r}\,|\,U_r(t)\,|\,\mathbf{p}\rangle \exp\{-(i/\hbar)\int dt'[\mathbf{p}-(e/c)\mathbf{A}(t')]^2/2m\}, \quad \mathbf{F}(t)=-\partial\mathbf{A}(t)/c\partial t, \tag{13a}$$

$$\exp[-(i/\hbar)\mathbf{p}\cdot\alpha_0\boldsymbol{\varepsilon}_z\sin\omega_0 t]=\exp[-i(\mu_0\mathbf{p}\cdot\boldsymbol{\varepsilon}_z/\hbar k)\sin(\omega_0 t)]=\Sigma_n J_n(\mu_0\mathbf{p}\cdot\boldsymbol{\varepsilon}_z/\hbar k)\exp[-(i/\hbar)(n\hbar\omega_0)t], \tag{13b}$$

where we have explicitely written out the Fourier expansion coefficients of the exponential in Eq. (12b), by using the Jacobi-Anger formula for the generating function of the ordinary Bessel functions (Gradshteyn & Ryzhik (2000)). This is one of the key mathematical formula with the help of which the strength of the *multiphoton side-bands* of additional energies $n\hbar\omega_0$ can be calculated. It is clearly seen that the appearance of the side-bands is a result of the *sinusoidal modulation of the phase of the Volkov state* given in Eq. (13a). The multiphoton ionization process can be viewed as a transition from a side-band to the free electron mass shell corresponding to free propagation. This is expressed by the delta functions in the incoherent superposition of the multiphoton transition amplitudes, describing the conservation of energy:

$$p_n^2/2m = n\hbar\omega_0 - (|E_0| + \Delta E_2) > 0, \quad n = n_{min} + n_{excess} = 1, 2, 3, \dots, \tag{14}$$

where n_{min} is the minimum number of quanta for the deliberation of the electron from the binding potential, and n_{excess} denotes the number of excess quanta, which may be absorbed additionally in the continuum. This latter phenomenon is called *above-threshold ionization* (ATI). The above-threshold electron spectra of nonlinear photoionization induced by *relatively long laser pulses*, analysed thoroughly e.g. by Krause et al. (1992), Agostini (2001), Paulus and Walther (2001), and recently by Banfi et al. (2005) and Ferrini et al. (2009) for multiphoton surface photoelectric effect, have common features with the corresponding high-harmonic spectra. The initial fall-off, the (occasionally rising) plateau and the sharp cut-off are present in each cases. Just for illustration, in Fig. 5 we show the structure of a typical ATI spectrum, which in many cases very much resembles to the HHG spectrum.

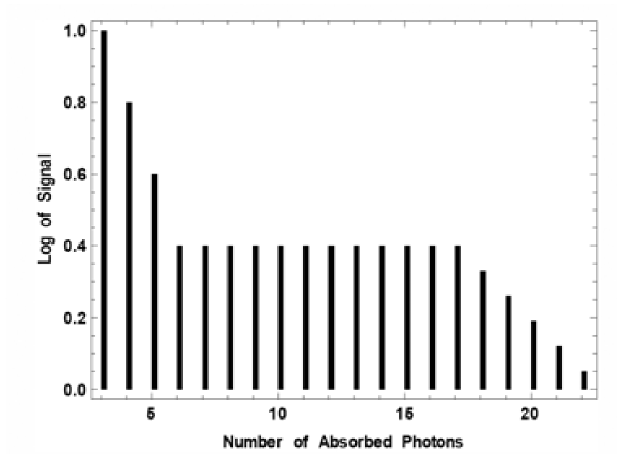

Fig. 5. Shows schematically a typical above-threshold ionization (ATI) electron spectrum. The initial fall-off, the plateau and the cut-off are usually also present in the higher-harmonic generation (HHG) processes (in which case the harmonic order is drawn on the abcissa).

From Eq. (14) it is seen that the ionization potential is increased by the ponderomotive energy shift, which, by the increase of the intensity, may well become even much larger than the photon energy. In such cases the channel corresponding to the minimum number of photons gets closed, and the phenomena of *peak supression* enters in the scene. Keldish (1965) has approximately calculated the *total ionization probability*, by using the method of stationary phase of the classical action in the phase of the electron's Volkov state. On the basis of his method, from Eqs. (12d) and (13a-b), the total probability can be approximated by the formula (see also Landau & Lifshitz (1978)):

$$w \sim \exp[-(2A/\hbar\omega_0)f(\gamma)], \quad f(\gamma) \equiv (1+1/2\gamma^2)\mathrm{Arsh}\gamma - (1+\gamma^2)^{1/2}/2\gamma, \quad \gamma \equiv \omega_0(2mA)^{1/2}/eF_0 \equiv 2\omega_0\tau, \tag{15}$$

where $A=|E_0|$ is the ionization potential (work function), and τ is the tunnel time. *The Keldish γ parameter* defined in Eq. (15) can also be expressed in terms of the dimensionless intensity parameter introduced already in Eq. (3b): $\gamma=2(A/2mc^2)^{1/2}/\mu_0$. We emphasize that the above approximate formula can be realistic strictly in the case when the ionization energy is much larger than the photon energy, $A=|E_0|>>\hbar\omega_0$. In the meantime the formula has been considerably refined, in particular by Ammosov, Krainov & Delone (1998), but we shall not enter into these more sophisticated methods. We also note that the tunneling time τ introduced here corresponds to the concept of tunneling time by Büttiker and Landauer (1982). The numerical value of the Keldish γ can be calculated according to the following formula: $\gamma=2\times10^6 E_{ph}(A/I_0)^{1/2}$, where the photon energy E_{ph} and the ionization potential (work function) measured in electron volts, and the peak intensity I_0 measured in W/cm^2. For example for $E_{ph}=1$, $A=25$, and $I_0=10^{12}$ we have $\gamma=10$. On the basis of Eq. (15), in the two extreme cases $\gamma >> 1$ („large frequency and small intensity": „multiphoton regime") and $\gamma << 1$ („small frequency and large intensity": „tunnel regime") we can give a simple physical interpretation of Eq. (15):

$$w \sim [\mu_0^2(mc^2/8A)]^{A/\hbar\omega_0} \sim (I_0)^n \ \text{if} \ \gamma >> 1, \qquad w \sim \exp[-4(2mA^3)^{1/2}/3\hbar e F_0] \ \text{if} \ \gamma << 1, \tag{16}$$

In the first formula of Eq. (16) we mean on the power index n the minimum number of photons needed for the ionization, $n\hbar\omega_0 >\approx A$. The „n-power-law" corresponds to the fall-off regime shown in Fig. 5. The tunnel regime $\gamma < 1$ can be reached with intensities higher than 10^{14} W/cm^2 in the above numerical example. In Figs. 6a and 6b we show in a special case the intensity dependence of the Keldish gamma parameter and the relative probability, respectively.

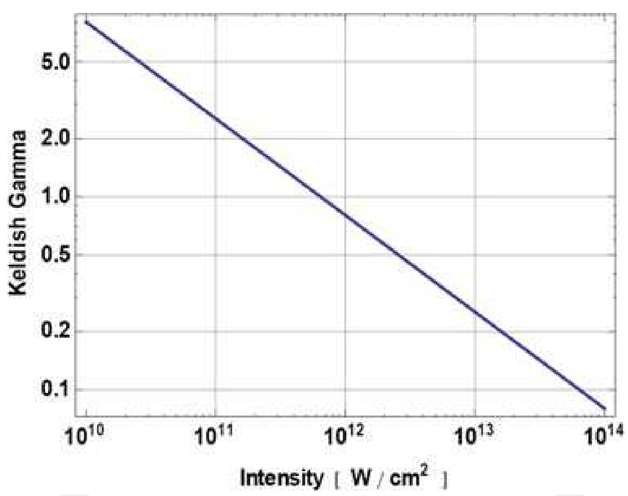

Fig. 6a. Shows the intensity dependence of the Keldish γ parameter, defined in Eq. (15), for the special values A=16eV and $\hbar\omega_0$=0.1eV, which correspond to the ionization of Argon atoms by a CO_2 laser. The transition region is located around the intensity value 10^{12} W/cm^2, where the value of the γ parameter becomes about unity.

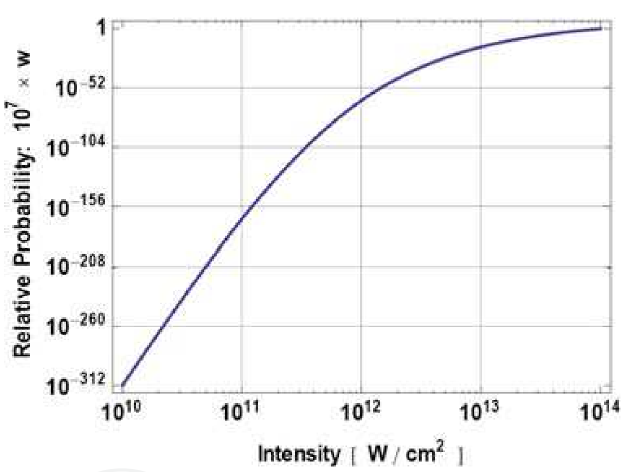

Fig. 6b. Shows the intensity dependence of the Keldish relative probability, according to Eq. (15), for the special values A=16eV and $\hbar\omega_0$=0.1eV, which correspond to the ionization of Argon atoms by a CO_2 laser. Around the intensity value 10^{12} W/cm^2, the dependence changes from the perturbative behaviour to the tunneling regime, as can also be estimated from Eq. (16).

The general equation for the matrix elements for multiphoton transitions, Eqs. (10a-b), can also be used to calculate the production rates of high-harmonics, as has been shown by Varró & Ehlotzky (1993). In contrast to the standard method based on the determination of the laser-induced nonlinear atomic dipole moment, this description relies on scattering theory, and it yields a possible *multiphoton generalization of the Kramers-Heisenberg dispersion* *relation*. By performing the time integral, we have the energy conservation for an n-th-order process $\hbar\omega' = n\hbar\omega_0 - (E_f - E_0)$, thus the formalism describes also the possibility that the final energy E_f of the atomic electron does not coincides with its initial energy E_0 . On the basis of Eqs. (10a-b) the differential cross-sections for such processes read

$$d\sigma_n/d\Omega = r_0^2(\omega'/\omega_0)^3(2a_0/\alpha_0)^2|M_n|^2,\quad r_0=e^2/mc^2,\ a_0=\hbar^2/me^2,\quad M_n=(\varepsilon_z\cdot\varepsilon')\delta_{f0}\delta_{n1} + S_1 + S_2\,, \tag{17a}$$

$$S_1 \equiv +i\Sigma_k[\langle\psi_f|V_n|E_k\rangle\langle E_k|(\mathbf{r}\cdot\varepsilon')|\psi_0\rangle(E_k - E_0 + \hbar\omega' + i\varepsilon)^{-1}]\,, \tag{17b}$$

$$S_2 \equiv +i\Sigma_k[\langle\psi_f|(\mathbf{r}\cdot\varepsilon')|E_k\rangle\langle E_k|V_n|\psi_0\rangle(E_k - E_0 + \hbar\omega' - n\hbar\omega_0 + i\varepsilon)^{-1}]\,, \tag{17c}$$

where in the summation over the intermediate states the positive infinitesimal ε has been introduced. It can be shown that the cross-section given by Eqs. (17a-b-c) reduces to the well-known linear dispersion formula. In Varró & Ehlotzky (1993) we have calculated the matrix elements analytically, and applied to the calculation of the production rates on three kinds of noble gas atoms, and obtained reasonable agreement with the experimental observations.

4. The nonlinear effect of the laser-induced oscillating double-layer potential on metal surfaces. X-ray generation in the presence of a static homogeneous electric field.

In case of multiphoton photoelectric effect of metals, Farkas & Tóth (1990) and Farkas et al. (1998) measured very high-order above-threshold electrons coming from metal targets. The theoretical interpretation of these results has been first given by the present author in Varró & Ehlotzky (1998) and recently in Kroó et al. (2007). In Varró et al. (2010) on the basis of the so-called laser-induced oscillating double-layer potential model the spontaneous emission of radiation by metallic electrons in the presence of electromagnetic fields of surface plasmon oscillations. Our original model belongs to a wider class Floquet-type analyses (see e.g. Kylstra (2001)), and considers the inelastic electron scattering on the *oscillating double-layer potential generated by the incoming laser field at the metal surface. The* model has already been succesfully used to interpret the experimental results on very high order surface photoelectric effect in the near infrared (Farkas & Tóth (1990)) and in the far infrared regime (Farkas et al., 1998). In this description the basic interaction leading to very high nonlinearities is caused by the collective velocity field of the oscillating electrons near the metal surface, within a layer of thickness smaller that the penetration depth δ. Because the quasistatic velocity field is screened inside the metal, the thickness of the layer is taken as the Thomas-Fermi screening length $\delta_s=1/k_F$ where $k_F=(6\pi n_e e^2/E_F)^{1/2}$. The wave function of an electron will then obey the two Schrödinger equations

$$(p^2/2m - V_0 - V_D\sin\omega_0 t)\Psi_I = i\hbar\partial_t\Psi_I\quad (z<0)\,,\qquad (p^2/2m + V_D\sin\omega_0 t)\Psi_{II} = i\hbar\partial_t\Psi_{II}\quad (z>0), \tag{18a}$$

where the subscript *I* refers to the interior region (metal) and *II* to the exterior region (vacuum), respectively. Following Varró & Ehlotzky (1998), the amplitude of the collective velocity field

$$V_D = 2\pi n_e e^2\alpha_0\delta_s = \mu_0(\omega_p/4\omega_0)(\delta_s/\delta)(2mc^2),\qquad \omega_p=(4\pi n_e e^2/m)^{1/2}\,,\qquad V_D = 3\mu_0\times 10^4\,\mathrm{eV}, \tag{18b}$$

where ω_p is the plasma frequency, $\alpha_0 \equiv \mu_0 \lambda_0 / 2\pi$, and δ denotes the skin-depth of the metal taken at the laser frequency ω_0. The interaction with the other agents like surface plasmon fields should in principle still be taken into account, but since their direct effect is much smaller than that of the induced collective velocity field, we have not displayed these direct interaction in Eqs. (7a-b). Thus in Eqs. (7a-b) we only display the Schrödinger equation relevant in the final state interaction, where additional energy redistribution takes place due to the interaction with the induced near field. The outgoing electron current components for which the momenta $p_n=[2m(n\hbar\omega_0 - A)]^{1/2}$ are real ($n \geq n_0=4$ in the case under discussion), corresponding to n-order multiplasmon absorption, have been obtained from the Fourier expansion of Ψ_I and Ψ_{II} . The unknown multiphoton reflection and transmission coefficients, R_n and T_n , respectively, can be determined from the matching equations, i.e. from the continuity of the wave function, $\Psi_I(0,t)=\Psi_{II}(0,t)$ and of its spatial derivative, $\partial_z\Psi_I(0,t)=\partial_z\Psi_{II}(0,t)$ which relation must hold for arbitrary instants of time. The resulting two coupled infinite set of linear algebraic equations for R_n and T_n can be numerically solved without any particular difficulty, moreover, it is possible to derive quite accurate analytic approximate formulas, too. According to these results, the current components normalized to the incoming current can be expressed as

$$j_t(n)=(p_n/q_0)\,|T_n|^2, \quad |T_n|^2 \approx J_n^2(a) \quad (n \geq n_0), \quad a \equiv 2V_D/\hbar\omega_0\,, \tag{19}$$

where $q_0=(2mE_F)^{1/2}$ is the average of the initial momenta. For instance, in case of $I_0=2\times10^8 W/cm^2$ we have $2V_D=11eV$, and $a=2V_D/\hbar\omega_0=7$ for a Ti:Sapphire laser of photon energy 1.56eV. We have taken for the ratio $(\delta_s/\delta)=2\times10^{-2}$, i.e. for the $\delta=22.5nm$ skin-depth of gold metal the screening length is about $\delta_s=0.4nm$. Though the standard nonlinearity parameter, $\mu_0=10^{-5}$, is very small at such intensities, the parameter „a" is that large that it can cause 7th order nonlinearities with sizeable probability. This numerical example clearly shows that already at very low intensities used in several experiments the *new nonlinearity parameter* „a" introduced in Eq. (19) has a much larger value than the argument of the Bessel function in Eq. (13b). On the basis of this remarkable quantitative difference, our theory based on introducing the laser-induced near-field is capable of accounting for the basic features of the recently measured unexpectedly broad above-threshold electron spectra. Because of the smallnes of μ_0, in the frame of the standard *nonperturbative* Volkov description there is no chance to interpret several recent and earlier multiphoton experiments. It is interesting to note that these high nonlinearities found in the experiments cannot be accounted for by including the effect of field enhancement due to the generation of surface plasmon oscillations. Though this effect results in a two order of magnitude increase of the intensity parameter, owing to the compression of the radiation field at the metal surface, it is still not enough to have the standard nonperturbative description to work.

The double-layer near-field model has also been applied to consider the generation of x-rays by irradiating metal surfaces with a powerful laser beam in the presence of a static electric field (Varró et al. (1999)). A similar effect has been recently studied by Odžak & Milošević (2005) in their work on high-order harmonic generation in the presence of a static electric field.

Recently, in a series of papers (see Varró (2004) and Varró (2007a-b-c)) the author has studied the scattering of a few-cycle laser pulse on a thin metal nano-layer, and the effect of the carrier-envelope phase difference has been investigated. The analysis has been extended to consider plasma layers, too, in the relativistic regime. Perhaps the most surprising effect which came out from the analysis, was the generation of phase dependent wake-fields. This wake-fields are reflected quasi-static.

By analogy, one may think that if the phases of the *above-threshold electron de Broglie waves* generated at the metal surface are locked (i.e. the difference of the phases of the neighbouring components is a smooth, possibly a constant function of the order, namely the number of absorbed photons), then the Fourier synthesis of these components yields an *attosecond electron pulse train* emanating perpendicularly from the metal surface, quite similarily to the generation of attosecond light pulses from high harmonics (which, on the other hand, are propagating in the specular direction). This expectation is quite natural, because the spacing of the electron peaks in the frequency space is just the optical frequency $h\nu_0/h=\nu_0$, like in the case of high-harmonic generation. The idea has been worked out quite recently by Varró & Farkas (2008a), where further references can be found concerning the general question attosecond electron pulses. Concerning these developments, we refer the reader to the above-mentioned papers.

5. Some quantum phase and other statistical properties of few-cycle and attosecond pulses

It is a natural, and both conceptually and practically important question that to what extent can one control the phase of the ultrashort pulses, and, at all, what are the ultimate quantum limits of phase stabilization in a given generating process? The systematic quantitative analysis of these problems is still missing, at least concerning the quantum uncertainties of the phases. One may expect that the usual high-intensity laser fields, being in a highly populated coherent state, can certainly be well represented in a satisfactory manner in the frame of external field approximation, i.e. in terms classical Maxwell fields of definite amplitudes and phases, or in terms of classical stochastic processes. On the other hand, in the generation of extreme pulses, like sub-femtosecond or attosecond pulses, it is an open question whether the fully quantum or the semiclassical description should be used for the correct interpretation. The reader can judge the importance of such questions by remembering the chapters devoted to the classical theory of coherence in the book by Born & Wolf (2009), for instance. Concerning the general description of quantum coherence and correlations of the electromagnetic radiation, we refer the reader to the books by Klauder & Sudarshan (1968), Scully & Zubairy (1997), Loudon (2000) and to the book by Schleich (2001) on quantum optics in phase space. In this context one has to keep in mind that, after all, though we have been speaking of „photon absorption" or „photon emission" in „multiphoton processes", either the classical or semiclassical framework has been used, in which the word „photon" has simply no meaning. As is general in the study of nonlinear laser-induced processes, the appearance of the side-bands in the electron energy in the phase of the wave function, $\exp[-(i/\hbar)(E_{el}-E'_{el})t-in\omega_0 t]$, is transformed to the mathematically identical form $\exp[-(i/\hbar)(E_{el}-E'_{el}+n\hbar\omega_0)t]$. The delta functions $\delta(E_{el}-E'_{el}+n\hbar\omega_0)$ appearing in the transition probabilities are said to be responsible for the energy conservation $E'_{el}=E_{el}+n\hbar\omega_0$, and then this balance equation is considered as „n-photon absorption" by the electron. On the other hand, in the very sense, no true light quanta has been considered, but rather mere classical Maxwell fields.

Recently we have analysed in quite details the interaction of strong quantized radiation fields with free electron wave-packets (see Varró (2008b-c) and Varró (2010b)), and found that the entangled photon-electron states, developed due to the interaction, are closely related to the number-phase minimum uncertainty states of the photon field. We have also proved (Varró (2010)) that the strong-field interactions necessarily lead to entangled states and the appearance of entropy remnants of a photon-electron system. In a series of papers Fedorov et al. (2006) have shown that the short-pulse and strong-field breakup processes may be considered as a route to study entangled wave-packets. For the other extreme of weak, single-photon fields we have recently presented a method to treat intensity-intensity correlations by simple classical probability theory (Varró (2008c)). The study of the interaction of electrons with quantised radiation field dates back to our earlier works (Bergou & Varró (1981a-b)), in which the multiphoton Bremsstrahlung processes and the nonlinear Compton scattering were described on the basis of an algebraic treatment introduced by us. These analyses has served as a "microscopic foundation" for the semiclassical treatments. Here we mention only one genuine quantal effect, namely that in Compton scattering the frequency ω'_n of the scattered radiation depends on the change in the occupation of the modes of the incoming radiation:

$$\omega'_n=(n\omega_0+\omega_c\mu_0^2\delta/4)/[1+(2n\omega_0/\omega_c+\mu_0^2/2)\sin^2(\theta/2)]\,, \tag{20}$$

where $\omega_c\equiv mc^2/\hbar$ is the Compton frequency, θ is the scattering angle and $\delta\equiv(N_f-N_i)/N_i$ is the "depletion factor", which is a relative change in the photon occupation number. The "quantum intensity parameter" μ_0 can be defined in a similar manner as in the semiclassical theory, in Section 2, such that in the present definition we *formally* associate an amplitude $A_0=(c/\omega_0)(2\pi\hbar\rho)^{1/2}$ for the vector potential, where $\rho=\langle N_i\rangle/L^3$ is the initial photon density.

In the following we present the simplest quantal description of high-harmonic generation in Thomson (Compton) scattering, where we describe the electron by a classical current density $\mathbf{j}(\mathbf{r},t)$, which is induced by the incoming strong laser field. This part corresponds to the description listed in cell No. 3 in Table 1. According to Eq. (3a) we take a nonrelativistic oscillation caused by a moderately intense field, i.e. $\mathbf{v}(t)=-\varepsilon_0 v_{osc}\sin(\omega_0 t+\varphi_0)$ and $\mathbf{r}(t)=\varepsilon_0 x_{osc}\cos(\omega_0 t+\varphi_0)$, thus we do not consider the effect of the possible drift motion (in fact, for an electron being initially at rest, this assumption does not correspond to any approximation, except that the special case of a smooth switching-on and -off is considered). The equation of motion of the quantized readiation field reads:

$$i\hbar\partial_t|\Phi\rangle=H_{int}|\Phi\rangle,\quad H_{int}=-(1/c)\int d^3r\,\mathbf{j}(\mathbf{r},t)\cdot\mathbf{A}_Q(\mathbf{r},t),\quad \mathbf{j}(\mathbf{r},t)=-e\mathbf{v}(t)\delta(\mathbf{r}-\mathbf{r}(t)), \tag{21a}$$

$$\mathbf{A}_Q(\mathbf{r},t)=\Sigma_{\mathbf{k}s}[\mathbf{g}(\mathbf{k},s;\mathbf{r},t)a_{\mathbf{k}s}+\mathbf{g}^*(\mathbf{k},s;\mathbf{r},t)a^\dagger_{\mathbf{k}s}], \tag{21b}$$

$$\mathbf{g}(\mathbf{k},s;\mathbf{r},t)\equiv c(2\pi\hbar/\omega_k L^3)^{1/2}\varepsilon(\mathbf{k},s)\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega_k t),\quad \omega_k=c|\mathbf{k}|,\quad [a_{\mathbf{k}s},a^\dagger_{\mathbf{k}'s'}]=\delta_{\mathbf{k}\mathbf{k}'}\delta_{ss'}\,, \tag{21c}$$

where $\mathbf{A}_Q(\mathbf{r},t)$ is the vector potential of the whole quantized radiation field decomposed into the vector plane wave modes represented by $\mathbf{g}(\mathbf{k},s;\mathbf{r},t)$. The exact solution of Eq. (21a) can be expressed with the help od the displacement operators $D[\alpha_{\mathbf{k}s}(t)]$ for each mode, by properly determining the parameters $\alpha_{\mathbf{k}s}(t)$, as is shown in the following equation:

$$|\Phi(t)\rangle=\Pi_{\mathbf{k}s}D[\alpha_{\mathbf{k}s}(t)]\,|\Phi(t_0)\rangle,\quad \alpha_{\mathbf{k}s}(t)=(i/\hbar c)\int dt\int d^3r\,\mathbf{j}(\mathbf{r},t)\cdot\mathbf{g}^*(\mathbf{k},s;\mathbf{r},t). \tag{22}$$

We summarize some basic properties of the displacement operator $D(\alpha)$ and the coherent states denoted by $|\alpha\rangle$, which are generated from vacuum under the action of the classical current. In order to simplify these formulae we do not display the mode index {**k**, s} (where **k** is the propagation vector and s is the polarization index of the scattered radiation):

$$D(\alpha)\equiv\exp(\alpha a^\dagger-\alpha^* a), \qquad D(\alpha)|0\rangle=|\alpha\rangle=\Sigma_k\alpha^k/(k!)^{1/2}|k\rangle\exp(-|\alpha|^2), \qquad a|\alpha\rangle=\alpha|\alpha\rangle, \tag{23}$$

Finally we note that the the displacement property $D^\dagger(\alpha)aD(\alpha)=a+\alpha$ means a shift of the quantized amplitudes. By putting explicit form of the electron current in the integral on the right hand side of Eq. (22), and using the Jacobi-Anger formula for the generation of the ordinary Bessel functions, we obtain:

$$\alpha_{\mathbf{k}s}(t)= (i/\hbar)ce\mu_0(2\pi\hbar/\omega_k L^3)^{1/2}(\varepsilon_0\cdot\varepsilon(\mathbf{k},s))\Sigma_n\{nJ_n[\mu_0\lambda_0(\varepsilon_0\cdot\mathbf{k})]/\mu_0\lambda_0(\varepsilon_0\cdot\mathbf{k})\}\int dt\exp[i(\omega_k-n\omega_0)t]. \tag{24}$$

The time integral for large interaction times give a disctrete sequence of resonant frequencies $\omega_{\mathbf{k}}=n\omega_0$, which are just the high-harmonic frequencies. In general, if the quantized field is initially on the vacuum state (thus we do not consider induced processes), i.e. $|\Phi(t_0)\rangle=|0\rangle$, then the quantum state $|\Phi(t)\rangle$, developing due to the interaction with the oscillating electron, will be a multimode coherent state. Because of the frequency condition coming from the resonant time integral in Eq. (24), each harmonic components will be in a coherent state, regardless of their average occupation (which is governed by the size of the Bessel functions). The expectation value of the energy of a particular component is given by the expression:

$$\langle(n\hbar\omega_0)a^\dagger_{n\omega0\mathbf{s}}a_{n\omega0\mathbf{s}}\rangle=r_0^2|\mathbf{s}\times\varepsilon_0|^2 n^4[2J_n(z)/z]^2, \quad \mathbf{s}\equiv\mathbf{k}/|\mathbf{k}|, \quad z\equiv n\mu_0(\mathbf{s}\cdot\varepsilon_0). \tag{25}$$

The formula, Eq. (25) is equivalent with the the classical formula for the high-harmonic production in nonlinear Thomson scattering, however, it contains only the first moment of the photon distribution, so it should be considered as a mean value. Of course, each harmonics of the scattered quantum field is loaded by inherent fluctuations, thus, the phases of the components are uncertain to a lesser or larger extent.

The simplest description of the phase uncertainties of a quantum field can be formulated in terms of the Susskind and Glogower „cosine" and „sine" operators, C and S, respectively, which are defined in analogy with the cosine and sine of a phase of the complex number (see e.g. Varró (2008c)). By introducing the *formal* polar decomposition of the quantized amplitudes, $a=E(a^\dagger a)^{1/2}$, $a=(a^\dagger a)^{1/2}E^\dagger$, we define

$$C\equiv(E+E^\dagger)/2, \quad S\equiv(E-E^\dagger)/2i, \quad \Delta C^2\equiv\langle(C-\langle C\rangle)^2\rangle, \quad \Delta S^2\equiv\langle(S-\langle S\rangle)^2\rangle, \tag{26}$$

where the variances ΔC^2 and ΔS^2 characterize the phase uncertainties. In Fig. 7 we have plotted the sum of this variances as a function of frequency in a Gaussian few-cycle pulse (which may result from high-harmonic generation, resulting in a quantum state like $|\Phi(t)\rangle$ above in Eq. (22)).

The Figure 7 clearly shows that the quantum phase uncertainty is quite steeply increasing as the spectral components are getting off the cental peak, and finally the uncertainty saturates, and takes its maximum value 1. It should be kept in mind that in general the nonlinear sources cannot be described by classical currents. According to our recent study(see Varró (2008b) and Varró (2010))., the joint quantum interaction with localized electron wave packets result in the generation of phase eigenstates of Jackiw type, thus they have very different correlation properties in comparison with coherent states, which are set to be the closest analogons of classical stable fields.

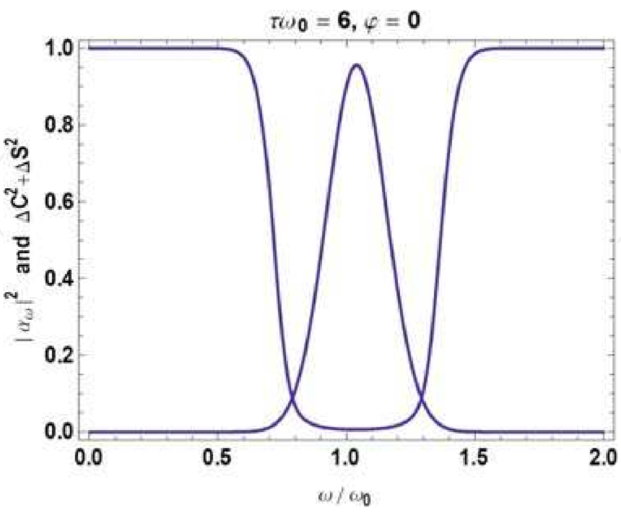

Fig. 7. Shows the normalized quantum amplitude distribution $|\alpha_\omega|^2$ of a 3—cycle femtosecond pulse and the dependencece of the quantum phase uncertainty $\Delta C^2+\Delta S^2$ associated to each spectral components with normalized frequency ω/ω_0. The pulse has been represented by a continuous Gaussian multimode coherent state, like that given in Eq. (22). The spectrum is peaked around ω_0 , where the quantum phase uncertainty has its minimum (it is essentially very close to zero).

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Pulsed lasers are available in the gas, liquid, and the solid state. These lasers are also enormously versatile in their output characteristics yielding emission from very large energy pulses to very high peak-power pulses. Pulsed lasers are equally versatile in their spectral characteristics. This volume includes an impressive array of current research on pulsed laser phenomena and applications. *Laser Pulse Phenomena and Applications* covers a wide range of topics from laser powered orbital launchers, and laser rocket engines, to laser-matter interactions, detector and sensor laser technology, laser ablation, and biological applications.

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