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POLARIZATION OF HIGH HARMONICS GENERATED FROM A HYDROGEN ATOM IN A STRONG LASER FIELD

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Although the high-harmonic generation from an atom subject to a strong laser field has extensively been investigated both experimentally [1-3] and theoretically [4-8], most results have been obtained so far only with linearly polarized fields[1,4-6]. Recently a few experimental groups[2,3] have begun investigating the effects of intense elliptically polarized laser fields in order to clarify the physical nature of atom-laser processes, and to find optimal conditions for high-harmonic generation. In particular, the rotation of polarization of high harmonics generated from argon by intense ($> 10^{13} W/cm^2$), short ($\simeq 200 fs$), elliptically polarized laser pulses has just been measured[3]. This suggests a need for a theoretical model of the phenomenon. Emphasize that quantitative description of an atom in elliptically polarized electric field

$$\Delta V(x,y,z,t) = E_0 f(t)[z sin\omega_0 t + \varepsilon y cos\omega_0 t] \quad (1)$$

(where E_0 and ε are the amplitude and the ellipticity of the pulse, ω_0 and $f(t)$ denote the laser frequency and the envelope of the pulse) requires a nonperturbative approach for solving the time-dependent four-dimensional Schrödinger equation. In this report an approach to this problem is given using the method of global approximation on a subspace grid originally developed for the stationary Schrödinger equation[9] and applied to atoms in strong non-homogeneous fields[10]. In principle the method is closed to "discrete variable representation", "pseudospectral" and Lagrange-mesh methods applied to different stationary problems (a set of the corresponding references can be found in the resent paper[11] devoted to the Lagrange basis).

Following the key idea of the papers [9,12] I seek a solution in spherical coordinates, (R, θ, ϕ), as an expansion

$$\psi(R,\Omega,t) = \frac{1}{R}\sum_{\nu j}^{N} \varphi_\nu(\Omega)\,\varphi^{-1}_{\nu j}\,\psi_j(R,t), \quad (2)$$

$$\varphi_\nu(\Omega) = P_l^m(\theta)\begin{cases} \cos m\phi, & m > 0 \\ \frac{1}{\sqrt{2}}, & m = 0 \\ \sin m\phi, & m < 0, \end{cases}$$

where $P_l^m(\theta)$ are the associated Legendre polynomials, $\nu = \{l,m\}$ and $\varphi^{-1}_{\nu j}$ is the $N \times N$ matrix inverse to $\{\varphi_\nu(\Omega_j)\}$ defined on the grid $\Omega_j = \{\theta_j, \phi_j\}_1^N$, coinciding with the nodes of the Gauss quadrature over the variables θ and ϕ. In this way the problem is reduced to a system of Schrödinger-type equations with respect to the vector $\vec{\Psi}(R,t) = \{\lambda_j^{1/2}\psi_j(R,t)\}_1^N = \{\lambda_j^{1/2}\psi(R,\Omega_j,t)\}_1^N$ of unknown coefficients in expansion (2)

$$i\frac{\partial}{\partial t}\vec{\Psi}(R,t) = \hat{H}(R,t)\,\vec{\Psi}(R,t), \quad (3)$$

where

$$H_{kj}(R,t) = \{-\frac{1}{2}\frac{\partial^2}{\partial R^2} - \frac{1}{R} + \Delta V(R,\Omega_k,t)\}\delta_{kj} +$$
$$\frac{1}{2R^2\sqrt{\lambda_k\lambda_j}} \sum_{\nu=\{l,m\}}^{N} \varphi^{-1}_{\nu k}\, l(l+1)\,\varphi^{-1}_{\nu j},$$

λ_j are the weights of the Gauss quadrature, $\sum_{\nu=1}^{N} = \sum_{m=-(\sqrt{N}-1)/2}^{(\sqrt{N}-1)/2} \sum_{l=m}^{m+\sqrt{N}-1}$. In such an approach the atom-laser interaction $\Delta V(R,\Omega_k,t)$ (1) is diagonal, and the non-diagonal, proportional to $1/R^2$, time-independent part of the matrix H_{kj} may be reduced to a diagonal form, since the problem permits a splitting of the operator $\hat{H}(R,t)$. This circumstance has been used for developing an economic algorithm with the computational time proportional to the number of unknowns in equations (3). Details of the scheme will be presented in a separate paper.

For approximation of Eq.(3) over the radial variable R a quasi-uniform grid on the interval $R \in [0, R_m = 250]$ has been created by reflecting $R \to x$ of the initial interval to the $x \in [0,1]$ with the formula $R = R_m(e^{4x} - 1)/(e^4 - 1)$. The computations have been performed with a uniform grid of 1000 points for x ($\Delta x = 0.001$), much more accurate then the analogous grid for R variable ($\Delta R = 0.25$). The integration time step Δt was 1/1600 of an optical cycle. To prevent the artificial reflection of the electron from the grid boundary the wave function was multiplied after each time step by the "mask function" suggested in[6]. As input to the initial-value problem (3) the ground state of an atom has been used. The spectrum of the emitted radiation $|\, d_{z,y}(\omega)\,|^2$ in z and y directions has been evaluated by the Fourier transform for the induced dipole of the atom over the final five cycles[6]

$$|\, d_{z,y}(\omega)\,|^2 = |\, \frac{1}{T_2 - T_1}\int_{T_1}^{T_2} dt\, e^{-i\omega t}\int d\vec{R}\,\psi^*(\vec{R},t)\,(z,y)\,\psi(\vec{R},t)\,|^2. \quad (4)$$

The problem of a hydrogen atom in the field (1) is analyzed here for the laser intensity $I = E_0^2\, c/(8\pi) = 2 \times 10^{13}\, W/cm^2$ at wavelengths $1064nm$ and $532nm$. This is already in the non-perturbative region, and the limiting case of the linearly polarized field ($\varepsilon = 0$) for these parameters has been investigated by other authors with different approaches[5,6]. The form of the pulse $f(t)$ has also been chosen to coincide with that of[6], consisting of a linear ramp for the first five optical cycles, followed by 15 additional cycles with $f(t) = 1$, which corresponds to $\approx 70\, fs$ and $\approx 35\, fs$ duration of the pulse at $1064nm$ and $532nm$ respectively. The parameters of the laser field are also rather close to those of the experiment for detection of the polarization rotation of high harmonics[3].

The result of the calculations for the linearly polarized pulse ($\varepsilon = 0$) is given in Fig.1. In this case the azimuthal variable ϕ may be separated, and the two-dimensional basis $\varphi_\nu(\Omega)$ (2) is reduced to the set of the Legendre polynomials $P_l(cos\theta)$. This figure demonstrates very rapid convergence of the calculated intensities $| d_z(\omega) |^2$ over the number, $N = l_{max} + 1$, of the terms in expansion (2) at $532nm$. Increasing the laser wavelength to $1064nm$, the convergence becomes slower, but the computation with N=7 still gives rather realistic approximation. As shown in Fig.1, the calculated intensities agree well with the most accurate of previously published data[6]. Note that the remaining difference between these two results is within the framework defined by using the different grids over $R(x)$ and t variables in[6] ($\Delta R = 0.25, \Delta t = 2\pi/(800\omega_0)$) and in the present paper. The ionization rate per optical cycle evaluated here is also close to that of[6]. Most accurate values of the intensities $| d(n\,\omega_0) |^2$ for the odd harmonics calculated with $\Delta x = 0.0005$ and $\Delta t = 2\pi/(3200\,\omega_0)$ are presented in the Table.

In Figs.2 the intensities $| d_z(\omega) |^2$ and $| d_y(\omega) |^2$ of the harmonics radiated in z and y directions are presented for elliptically polarized fields ($\varepsilon \neq 0$). The calculations have been performed with $N = 49$, which corresponds to $-3 \leq m \leq 3, m \leq l \leq m + 6$. As illustrated above (see Fig.1,$N = 7$), the computation with this basis set is rather accurate at $532nm$ and gives a realistic picture of the total spectrum of harmonics generated at $1064nm$. These figures show considerable deviation of the ellipticities of the generated harmonics (for some n) from the ellipticity of the fundamental.

Finally, the quantities $\varepsilon' =| d_y(n\omega_0)/d_z(n\omega_0) |$ calculated for a few $\varepsilon \neq 0$ are presented in Fig.3 where satisfactory convergence over N is shown at $532nm$. This figure demonstrates clearly a significant alteration of harmonic polarization with respect to polarization of the fundamental laser field for $n = 3, 5$ at $532nm$ and for $9 \leq n \leq 17 (\varepsilon = 0.1)$ at $1064nm$. The computations show also the dependence of the values ε' on the wavelength and the ellipticity ε of the driving field. These results are in agreement with experiment [3] which measured the rotation of the harmonic polarization relative to the polarization of the driving field in argon, and support the conclusion of paper[3] that the effect is due to single-atom response to a strong laser field.

The high efficiency of the approach developed here for the problem of high harmonic generation leads to its application to other problems of atomic interaction with elliptic driving fields (particularly for evaluation of the photoelectron spectra). Two circumstances, the rapid convergence over N and the proportionality of the computation time to the number of unknowns of (3), suggest the use of our approach with stronger fields and longer laser pulses. All the computations were performed on a DEC 3000 - M600 work-station.

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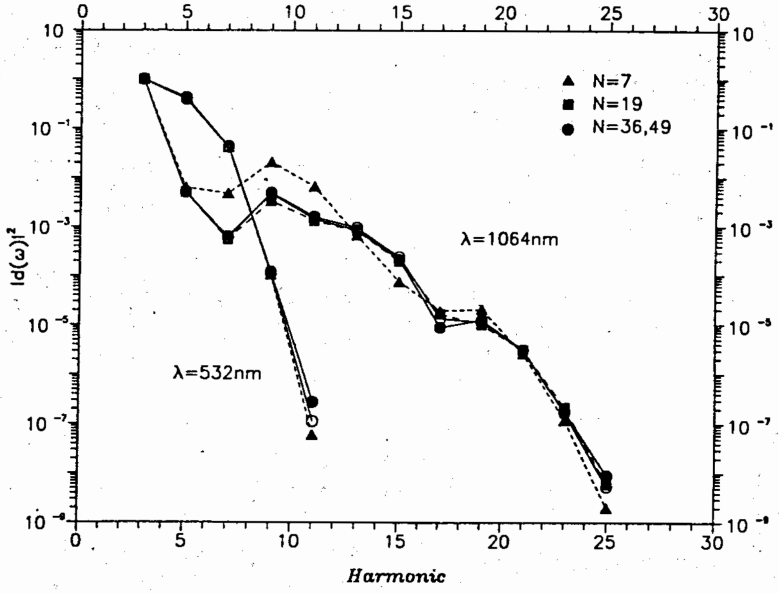

Fig. 1: The convergence of the harmonic intensities $| d(n\,\omega_0) |^2$ (where $n = 3, 5, ...$) with respect to the number, N, of the terms included in expansion (2). The first harmonics, $n = 1$, have been omitted, and the intensities have been normalized at the third harmonic. The previously published results[6] are also presented (open cycles) for comparison (by the courtesy of K.C. Kulander supplied us the data from Fig.1 of[6]).

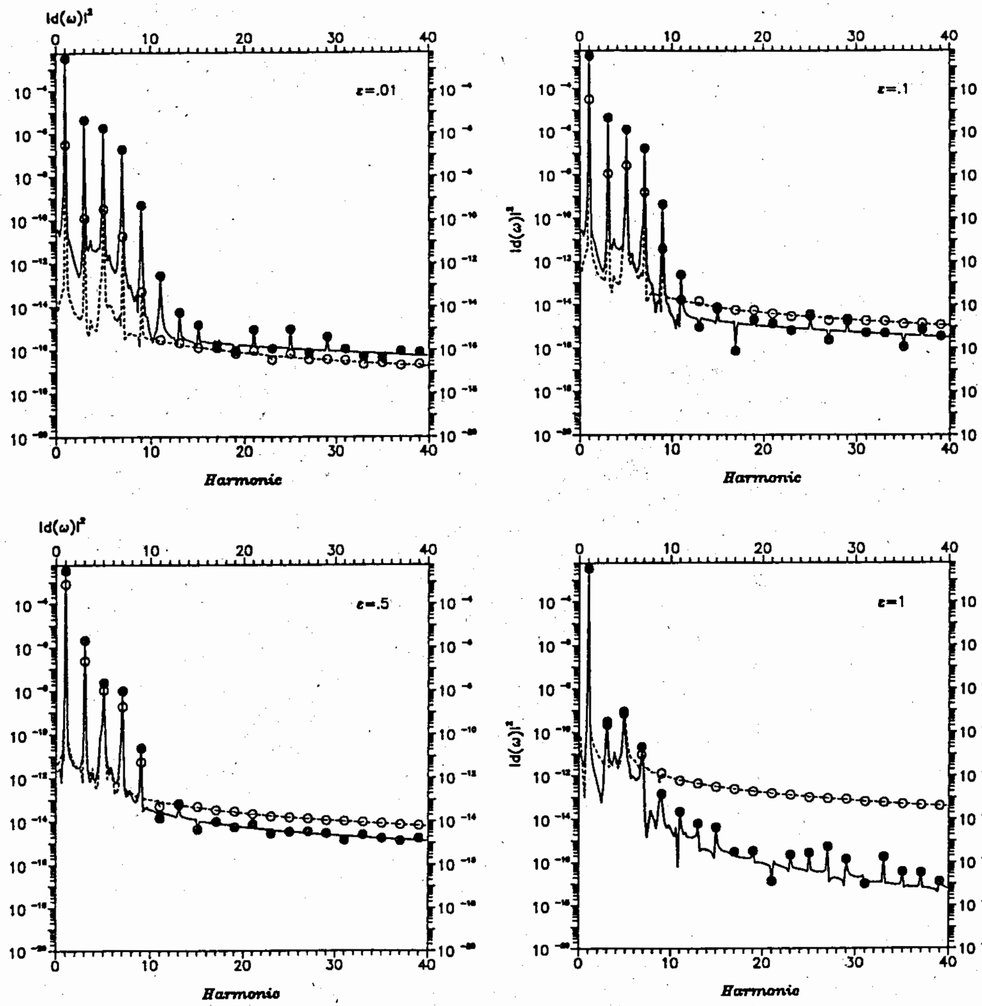

Fig. 2a: Harmonic spectra $| d_{(z,y)}(\omega) |^2$ at $532nm$ calculated for a few values ε with $N = 49$, $\Delta x = 0.001$ and $\Delta t = 2\pi/(1600\,\omega_0)$. The solid lines correspond to $| d_z(\omega) |^2$ (filled cycles mark the odd harmonics $\omega = n\,\omega_0$, $n = 1, 3, ...$) and the dashed ones - to $| d_y(\omega) |^2$ (open cycles mark the odd harmonics $\omega = n\,\omega_0$, $n = 1, 3, ...$).

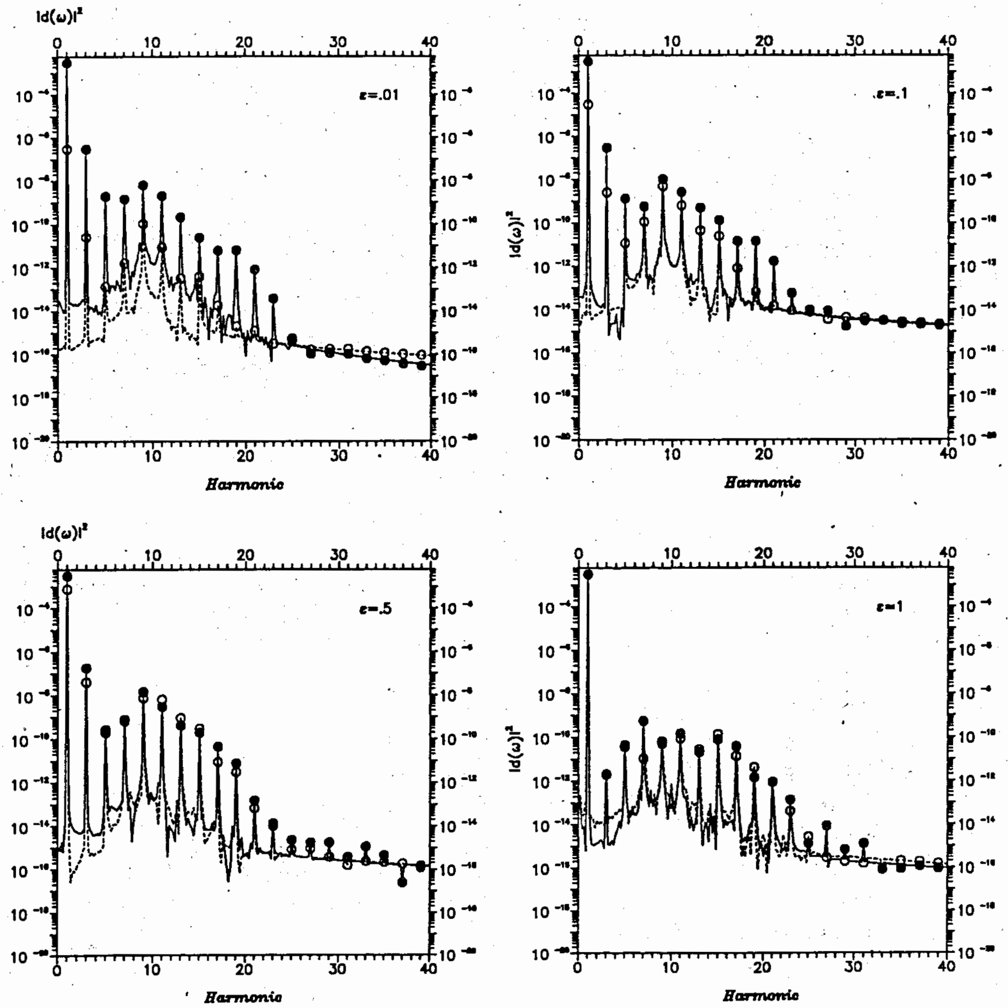

Fig. 2b: Same as in Fig.2a but at $1064nm$.

Table 1: The harmonic intensities $| d_z(n\omega_0) |^2$ calculated at $N = 49, \Delta x = 0.0005$ and $\Delta t = 2\pi/(3200\,\omega_0)$ for the case $\varepsilon = 0$. The last digits of the values may be subject to computational errors.

532nm	n	1	3	5	7	9
	$\|d_z\|^2$	3.328×10^{-3}	4.428×10^{-6}	1.818×10^{-6}	1.96×10^{-7}	5.15×10^{-10}
1064nm	n	1	3	5	7	9
	$\|d_z\|^2$	3.098×10^{-3}	3.027×10^{-7}	1.561×10^{-9}	2.14×10^{-10}	1.46×10^{-9}
	n	11	13	15	17	19
	$\|d_z\|^2$	4.81×10^{-10}	2.42×10^{-10}	6.7×10^{-11}	3.6×10^{-12}	2.4×10^{-12}

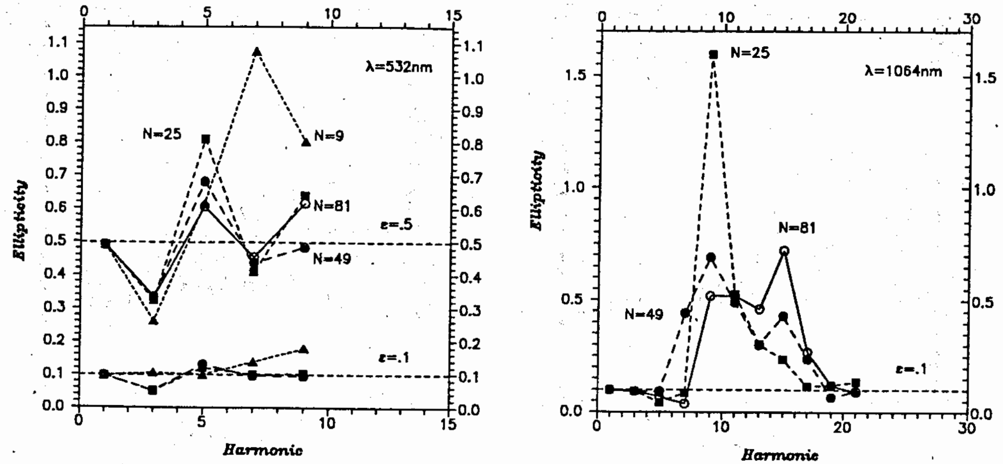

Fig. 3: The calculated ellipticities $\varepsilon'(n) = | d_y(n\,\omega_0)/d_z(n\,\omega_0) |$ of generated harmonics for a few ellipticities ε of the fundamental laser field (1).

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Мележик В.С. E4-96-47

Поляризация высоких гармоник, излучаемых атомом водорода в сильном лазерном поле

С помощью непертурбативного метода глобальной аппроксимации на подпространственной сетке исследован спектр высоких гармоник атома водорода в сильном ($>10^{13}$ Вт/см^2), эллиптически поляризованном лазерном поле. Обнаружена существенная перестройка поляризации гармоник относительно поляризации лазера.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 1996

Melezhik V.S. E4-96-47

Polarization of High Harmonics Generated from a Hydrogen Atom in a Strong Laser Field

The high harmonic spectrum of a hydrogen atom subject to an intense ($>10^{13}$ W/cm^2), elliptically polarized laser field is analyzed with a nonperturbative method of global approximation on a subspace grid. Considerable alteration of harmonics polarization with respect to laser polarization is found.

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

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