Phase control of the interaction of short laser pulses with simple systems

Astapenko V.A., Romadanovskii M.S.

Moscow Institute of Physics and Technology

Modern laser technologies enable researchers by super-short laser pulses with controllable phase parameters such as carrier-envelope phase (CEP) and chirp of carrier frequency. Changing CEP or frequency chirp one can monitor the probability of light-induced processes or in other words to control radiative phenomena by phase modulation (phase control).

The paper is devoted to theoretical investigation of the phase control during the interaction between short laser pulses and simplest systems such as classical harmonic oscillator, classic Morse oscillator and two-level quantum system (TLS). We consider two types of exciting laser pulses. The first one has the electric field in the form

$$\mathbf{E}(t) = \mathbf{E}_0 \exp\left(-t^2/\Delta t^2\right)\cos(\omega t + \varphi_0), \qquad (1)$$

and another one in the form

$$\mathbf{E}(t) = \mathbf{E}_0 \exp\left(-t^2/\Delta t^2\right)\cos\left(\omega t + \kappa t^2\right), \qquad (2)$$

here φ_0 is CEP and κ is frequency chirp.

In the first order of the perturbation theory we obtain the following general expression for the population of the upper energy state of two-level system excited by laser pulse after it termination

$$N_2\left(T_{1,2} >> t >> \Delta t\right) = \left(\frac{d_0}{\hbar}\left|E(\omega_0)\right|\right)^2, \qquad (3)$$

here d_0 is transition dipole moment, ω_0 is own frequency of TLS, $T_{1,2}$ are relaxation times of population and coherency, $E(\omega)$ is Fourier transform of electric field strength in laser pulse. Substituting Fourier transform of electric field strength (1) into formula (3) one can find the following expression for population of upper TLS state after the laser pulse termination

$$N_2(\tau > \eta) \cong \frac{\pi}{4}(\xi\,\eta)^2\, G_{TS}(r,\eta)\left\{1 + \sec h\left(r\,\eta^2\right)\cos(2\varphi_0)\right\} \qquad (4)$$

here $\tau = \omega_0\, t$, $\xi = d_0\, E_0/\hbar\,\omega_0$, $\eta = \omega_0\,\Delta t$, $r = \omega/\omega_0$ are dimensionless parameters of laser pulse and TLS and $G_{TS}(r,\eta)$ is excitation spectral line shape of TLS:

$$G_{TS}(r,\eta) = \exp\left[-\frac{\eta^2\,(r-1)^2}{2}\right] + \exp\left[-\frac{\eta^2\,(r+1)^2}{2}\right]. \qquad (5)$$

From expression (4) one can see that the function $\sec h\left(r\,\eta^2\right)$ plays the role of phase modulation coefficient of TLS excitation by laser pulse (1). This function is represented in fig. 1.

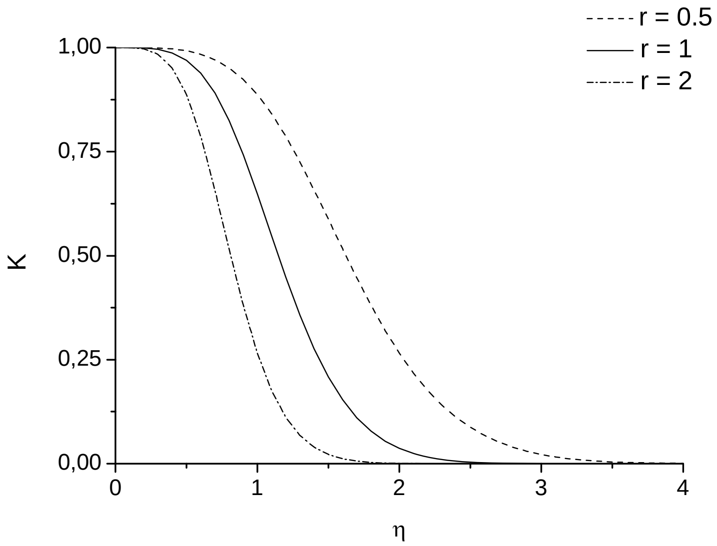

Fig. 1. Phase modulation coefficient as a function of dimensionless pulse duration in the perturbation theory limit

From fig. 1 it follows that in the perturbation case CEP control can be achieved only for subcycle laser pulses when $\eta < 2$. For longer pulses CE phase dependence of population N_2 is negligible.

For chirped pulse excitation one can obtain from (2), (3) the following expression

$$N_2 = \frac{\pi}{4}\frac{(\xi\eta)^2}{\sqrt{1+\alpha^2}} G(\alpha,\eta,r)\left[1 + K(\alpha,\eta,r)\cos\left(f(\alpha,\eta,r)\right)\right], \quad (6)$$

here

$$G(\alpha,\eta,r) = \exp\left\{-\frac{\eta^2(r-1)^2}{2(1+\alpha^2)}\right\} + \exp\left\{-\frac{\eta^2(r+1)^2}{2(1+\alpha^2)}\right\}, \quad (7)$$

$$K(\alpha,\eta,r) = \operatorname{sech}\left\{\frac{r\eta^2}{1+\alpha^2}\right\}, \quad (8)$$

$$f(\alpha,\eta,r) = \frac{\alpha\eta^2(1+r^2)}{2(1+\alpha^2)} - \operatorname{arctg}(\alpha) \quad (9)$$

$\alpha = \kappa\Delta t^2$ is dimensionless chirp of carrier frequency.

For arbitrary values of electric field strength the upper level population of TLS can be calculated numerically using the system of equations for Bloch vector $\mathbf{R} = (R_1, R_2, R_3)$:

$$\begin{cases} \dot{R}_1 = R_2 \\ \dot{R}_2 = -R_1 + 2\xi\exp\left(-\frac{\tau^2}{\eta^2}\right)\tilde{E}(\tau)R_3 , \\ \dot{R}_3 = -2\xi\exp\left(-\frac{\tau^2}{\eta^2}\right)\tilde{E}(\tau)R_2 \end{cases} \quad (10)$$

here $\tilde{E} = E/E_0$ is dimensionless field strength and using the relationship

$$N_2 = \frac{1 - R_3}{2}. \qquad (11)$$

Solution of (10) – (12) for laser pulse with controllable CEP (1) is shown in Fig. 2.

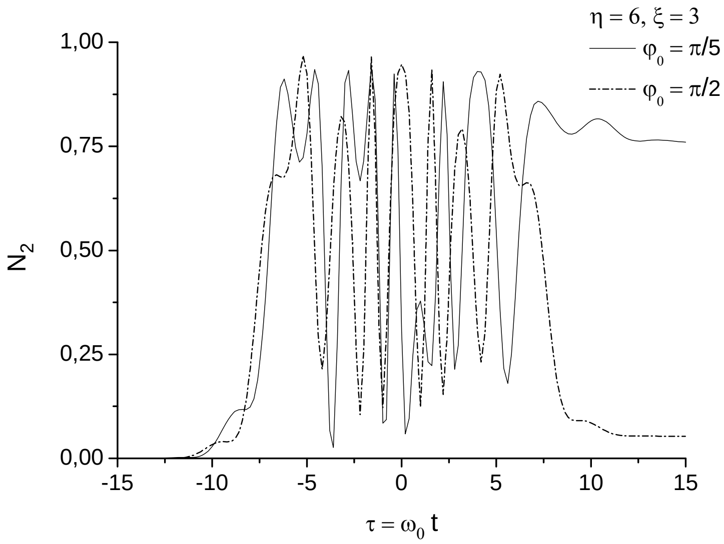

Fig. 2. Time dependence of upper level population of TLS for strong field excitation and near one-cycle laser pulse

From this figure one can see that upper level population after laser pulse termination can be minimized by proper choice of CEP value for short pulse duration and for high enough electric filed strength.

Figure 3 shows CEP dependences of population N_2 for various values of ξ.

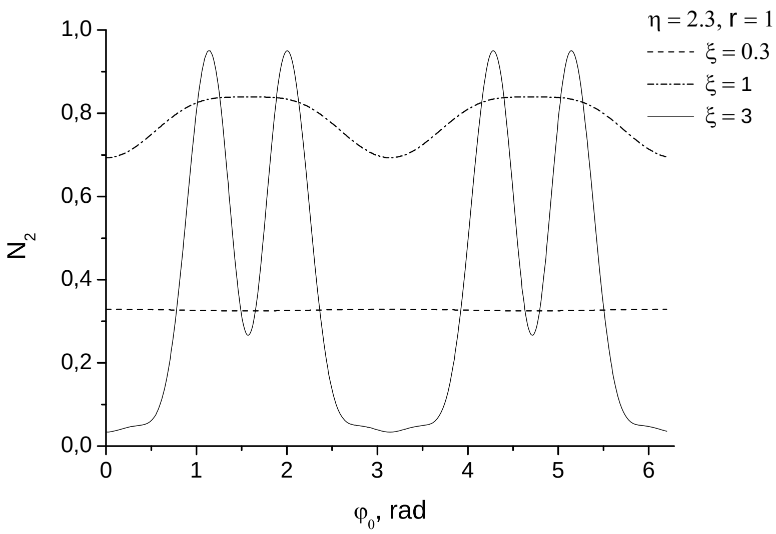

Fig. 3. CEP dependence of upper level population after laser pulse termination

Dependences of phase modulation coefficient upon the dimensionless pulse duration for various values of dimensionless field strength are shown in fig. 4.

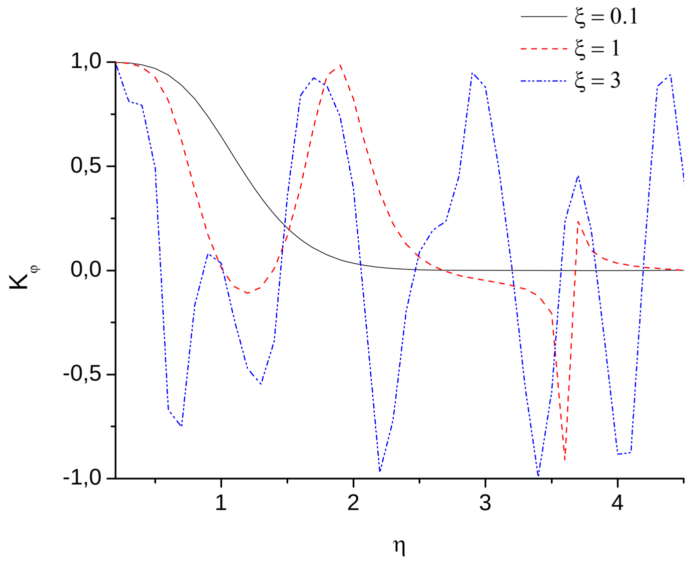

Fig. 4. Phase modulation coefficient as a function of laser pulse duration

In this figure the solid line coincides with the result for perturbation theory limit given by formula (4). Thus in the range $\xi < 0.1$ one can use formula (4) to calculate CEP control of TLS excitation by super-short laser pulse. For higher ξ values phase modulation coefficient oscillates as a function of pulse duration.

To analyze the Morse oscillator behavior under the action of strong laser field (1), (2) we solve numerically the following equation

$$\ddot{\rho}_{\tau^2} = \exp(-2\rho) - \exp(-\rho) + \gamma \tilde{E}(\tau, \varphi_0), \qquad (12)$$

here $\rho = \alpha x$ is dimensionless oscillator coordinate, α is parameter of Morse potential, $\gamma = \alpha q E_0 / m\omega_0^2$ is dimensionless field strength. Fig. 5 shows calculated CEP dependence of radiation power after Morse oscillator excitation by laser pulse (1) for one-cycle pulse duration.

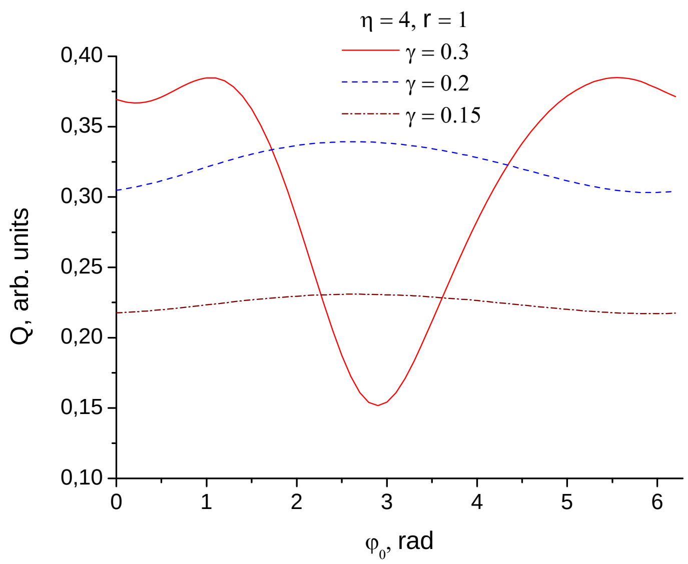

Fig. 5. CEP dependence of Morse oscillator radiation power after laser pulse excitation

From fig. 5 one can see that even for one-cycle laser pulses ($\eta = 4$) CEP control can be achieved only for high enough field strength $\gamma > 0.2$. In the case of weak electric field strength $\gamma < 0.1$ harmonic approximation is valid for the description of CEP control of Morse oscillator excitation.

The excitation energy ε of Morse oscillator under the action of one-cycle laser pulse as a function of field strength parameter γ is shown in fig. 6 for various CEP values (D is bound energy of Morse oscillator).

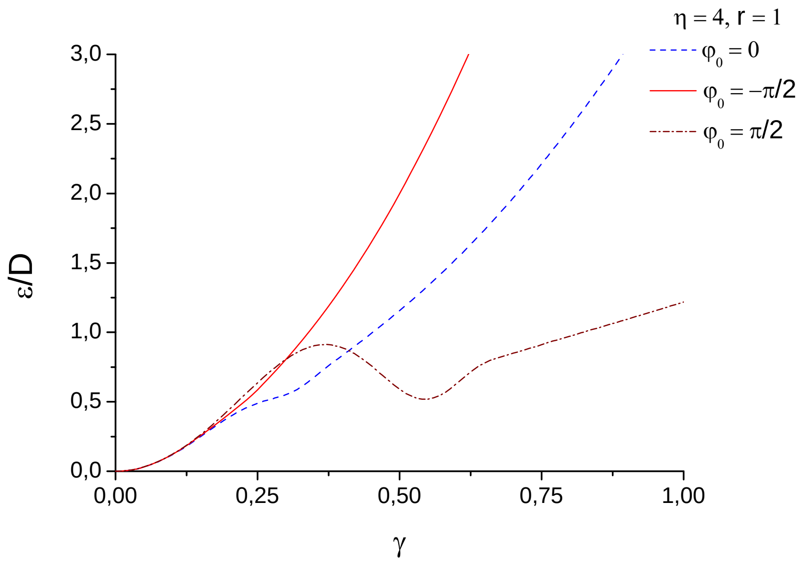

Fig. 6. Excitation energy of Morse oscillator as a function of dimensionless field strength

It should be mentioned that for $\varepsilon \geq D$ the dissociation of Morse oscillator takes place. Thus dissociation may be controlled by monitoring CEP value. From fig. 6 one also can see another interesting feature of Morse oscillator phase control, namely, the up-down asymmetry of the excitation process $\varepsilon(\varphi_0 = \pi/2) \neq \varepsilon(-\varphi_0 = \pi/2)$ arisen because of the lack of inverse symmetry of Morse potential.

References

1. Arustamyan M.G., Astapenko V.A. *Phase control of two-level system excitation by short laser pulses.* Laser Physics, 2008, v.18, No6, pp.768-773.

2. Arustamyan M.G., Astapenko V.A. *Phase control of oscillator excitation under the action of ultra-short laser pulses.* Laser Physics, 2008, v.18, No9, pp.1031-1036.

3. Astapenko V.A., Romadanovskii M.S. *Two level system excitation under the action of chirped laser pulse* Laser Physics, 2009, v.19, No5 (in press).