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Pulsed excitation of quantum oscillator: model account for anharmonicity

F.B. ROSMEJ[1, 2] AND V.A. ASTAPENKO[3]

[1] Sorbonne University, Faculty of Science, UMR7605, case 128, 4 Place Jussieu, 75252 Paris, France
[2] LULI, Ecole Polytechnique, Atomic Physics in Dense Plasmas, Route de Saclay, 91129 Palaiseau, France
[3] Moscow Institute of Physics and Technology, 141701 Dolgoprudnyi, Russia

ABSTRACT: We proposed simple model for the account of quantum oscillator anharmonicity during its excitation by laser pulse with arbitrary parameters. This model is based on the correspondence between electric area of the pulse and its Fourier transform. To demonstrate our method we used formulas for Morse oscillator excitation probability from ground state by subcycle unipolar pulse derived in paper [1] in the framework of sudden perturbations approximation. Our approach permits the generalization of their results on the case of longer pulses with carrier frequency. Comparison between Morse oscillator excitation and the excitation of harmonic oscillator is also presented.

1. EXCITATION OF QUANTUM MORSE OSCILLATOR BY UNIPOLAR SUBCYCLE PULSE

Probability of quantum Morse oscillator excitation from ground state by unipolar subcycle pulse derived in the framework of sudden perturbation approximation [1] is equal to

$$W_{0\to n}^{(M)} = \frac{2\,s_n}{n!}\frac{\left|\Gamma(2\,s_0 - n - i\gamma)\right|^2}{\Gamma(2\,s_0)\Gamma(2\,s_0 - n + 1)}\left|\frac{\Gamma(1+i\gamma)}{\Gamma(1-n+i\gamma)}\right|^2 \tag{1}$$

here

$$\gamma = \frac{S_E\,Q\cos\theta}{\alpha} \quad s_n = \frac{\sqrt{2\,\mu U_0}}{\alpha} - n - 1/2 \tag{2}$$

Q is oscillator charge, μ is its mass, U_0 is the energy of the oscillator dissociation, θ is angle between oscillator axis and electric field of the pulse, α is parameter of Morse potential and

$$S_E = \int E(t)\,dt \tag{3}$$

is the electric pulse area,

$$S_E = \sqrt{2\pi}\,E_0\,\tau \tag{4}$$

is the electric pulse area of unipolar Gaussian pulse which is given by the expression

$$E(t) = E_0 \exp\left(-t^2/2\,\tau^2\right) \tag{5}$$

τ is pulse duration and E_0 is its amplitude.

In what follows we consider also oscillator excitation by Gaussian with carrier frequency ω_c:

$$E(t) = E_0 \exp\left(-t^2/2\tau^2\right)\cos(\omega_c t) \tag{6}$$

Our task is to generalize consideration of paper [1] on pulses with arbitrary duration and carrier frequency.

2. COMPARISON BETWEEN MORSE AND HARMONIC OSCILLATOR EXCITATIONS BY DIFFERENT PULSES

For harmonic quantum oscillator excitation probability from the ground state one has

$$W_n^{(H)} = \frac{\nu^n}{n!} e^{-\nu} \tag{7}$$

This expression follows from general Schwinger formula for quantum oscillator excitation by classical electric current [2]. Key dimensionless parameter ν in the case by electric field excitation is equal to [3]

$$\nu = \frac{Q^2}{2\mu\omega_0} \left|E(\omega_0, \tau)\right|^2 \tag{8}$$

here $E(\omega_0, \tau)$ is Fourier transform of electric field strength calculated at the eigenfrequency of harmonic oscillator.

The comparison between the probability excitation of Morse oscillator and harmonic oscillator as a function of pulse duration is shown in Fig.1. This figure shows the excitation of a harmonic oscillator by Gaussian pulses without a carrier frequency and with a carrier frequency and excitation oscillator by unipolar pulse (5) from ground oscillator state. Calculations are made for E_0=0.1 a.u., θ=0 and for following molecular parameters: α=2.1 a.u., U_0=0.11 a.u., μ=18380 a.u., Q=1 a.u. [1]

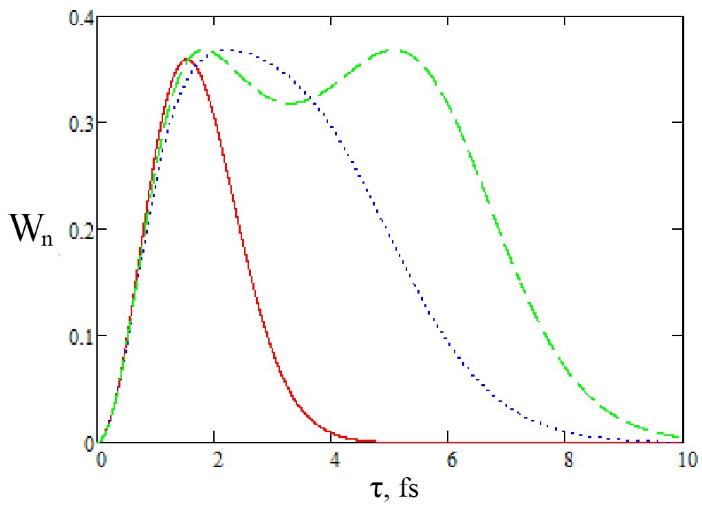

Fig.1. Excitation probability of Morse oscillator on 0⟶1 transition as function of pulse duration: solid line – calculation using formulas (1)-(2), dotted line – calculation using Schwinger formula (7) for Gaussian pulse with carrier frequency equal to eigenfrequency of oscillator, dashed line – calculation using Schwinger formula for Gaussian pulse with zero carrier frequency

It can be seen from this figure that the above dependences for the excitation of the Morse oscillator and the harmonic oscillator coincide with each other only for short pulse durations, when the approximation of sudden perturbations is valid.

3. MODEL

Our model is based on the following correspondence between electric pulse area and the Fourier transform of electric field strength in the pulse:

$$S_E \to \left|E(\omega_{0M})\right| \tag{9}$$

here we introduce eigenfrequency of Morse oscillator according to the equality

$$\omega_{0M} = \sqrt{\frac{2U_0}{\mu}}\,\alpha\,. \tag{10}$$

This definition follows from the harmonic limit of Morse potential. For above parameters of Morse oscillator one has: ω_{0M}=0.198 eV.

Note that correspondence (9) is rigorous for harmonic oscillator.

In what follows we use expressions (9) and (10) substituted in formulas (1)-(2) for the calculation of the excitation probability of Morse oscillator from its ground state.

Using proposed model one can describe Morse oscillator excitation without framework of sudden perturbations approximation i.e. for any pulse durations and for the excitation by general pulse type (6) with carrier frequency.

4. RESULTS AND DISCUSSIONS

Results of calculations of τ-dependence (dependence on pulse duration) of excitation probability of Morse and harmonic oscillators to different excited states are presented in Fig. 2-3.

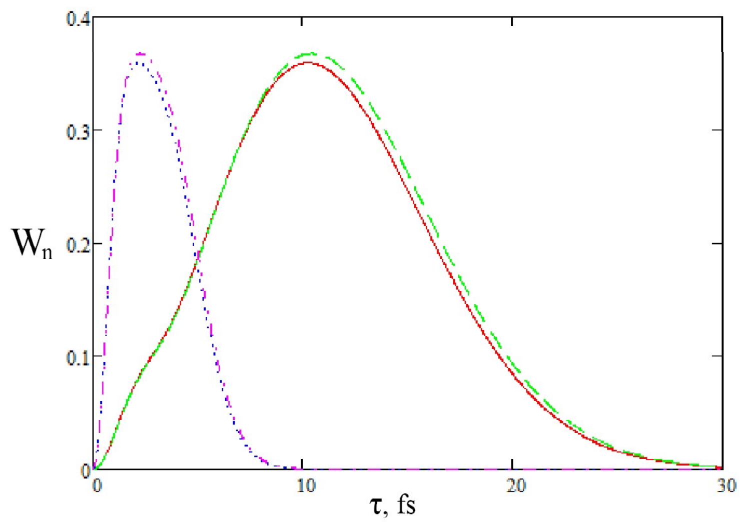

Fig.2. Excitation of 0⟶1 transition: solid line – Morse oscillator, E_0=0.03 a.u., dotted line – Morse oscillator, E_0=0.1 a.u., dashed line – harmonic oscillator, E_0=0.03 a.u., dotted-dashed line – harmonic oscillator, E_0=0.1 a.u., $\omega_c=\omega_0$

One can see from Fig.2 that in the case of the 0→1 transition excitation there is practically no difference between Morse and harmonic oscillators for small and large values of pulse amplitude. This is due the fact that for small excitation energy oscillator Morse is equivalent to harmonic oscillator while for high excitation level there is a significant difference between these oscillators as one can see from Fig.3.

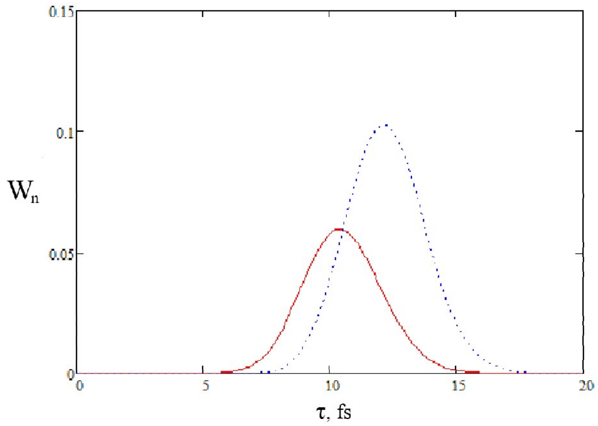

Fig.3. Excitation of 0→15 transition: solid line – Morse oscillator, dotted line – harmonic oscillator, E_0=0.1 a.u., $\omega_c=\omega_0$

Note that the maximum of the excitation probability for harmonic oscillator shifts to longer pulse durations in comparison with oscillator Morse for the excitation into high energy level.

Spectra of excitation probability, i.e. the probability dependence on pulse carrier frequency are shown in Fig.4-6 for excited states with low and high energies.

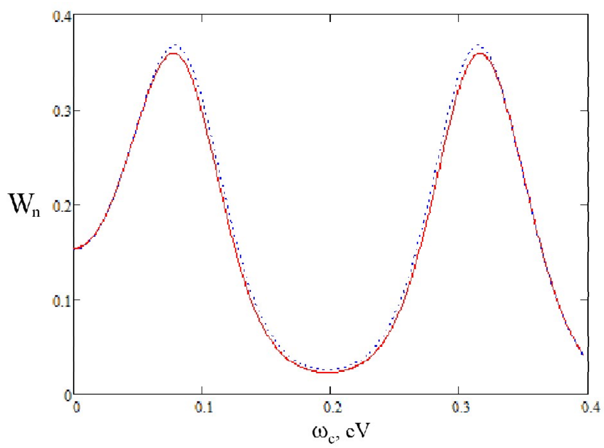

Fig.4. Excitation of 0→1 transition, t=7.2 fs, E_0=0.1 a.u.: solid line – Morse oscillator, dotted line – harmonic oscillator

One can see from Fig.4 that there is practically no difference between Morse and harmonic oscillators for the excitation into low energy level while for high level excitation this difference is significant (Fig.5, 6).

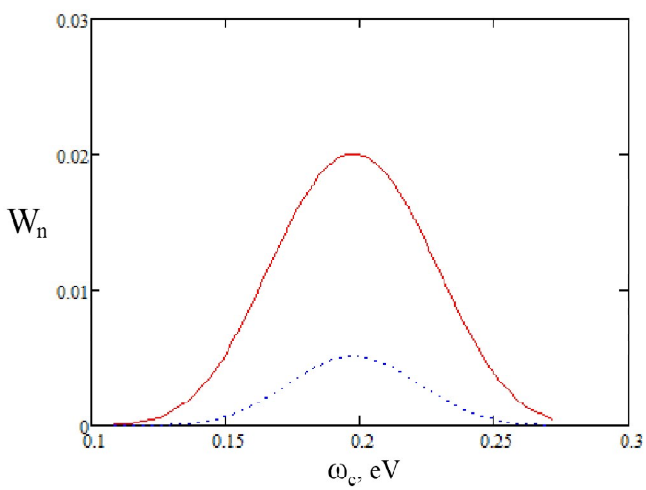

Fig.5. Excitation of 1→12 transition, τ=7.2 fs, E_0=0.1 a.u.: solid line – Morse oscillator, dotted line – harmonic oscillator

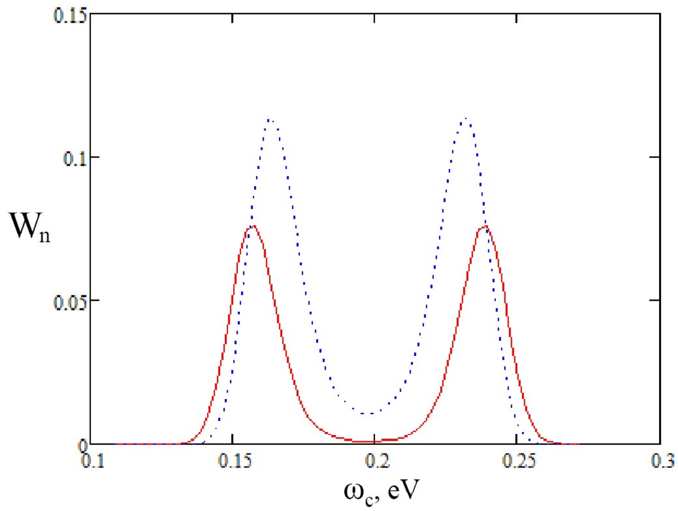

Fig.6. Excitation of 0→12 transition, τ=14.4 fs, E_0=0.1 a.u.: solid line – Morse oscillator, dotted line – harmonic oscillator

Figures 5 and 6 demonstrate also the transition from weak perturbation into the regime of strong perturbation during oscillator excitation by laser pulse. In the latter case minimum occurs instead the maximum in spectrum of excitation probabilities and two side maxima appear.

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