

Coherent control and chaos in the multiphoton dissociation of diatomic molecules

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Extended Abstract

One of the most intriguing aspects of physics is the correspondence between quantum and classical theories [1]. Quantum-classical analogies are crucial for the applications of nonlinear classical mechanics to microscopic systems and for quantum chaos studies. On the other hand, the development of quantum control brings up the additional question to whether one can apply coherent control to classically chaotic systems [2]. An interesting case is the control of the ionization of the hydrogen atom by a two-color field. Sirko and Koch have interpreted this phenomenon in terms of classical common resonances [3]. Therefore, one might expect quantum-classical correspondence also for the control of dissociation of diatomic molecules by infrared pulses.

The driven Morse oscillator is a paradigm for the comparisons between quantum and classical mechanics [4–7]. Apart from its analytical properties, it accounts for real molecular anharmonicity and has a continuum energy sector, which allows for the description of the dissociation [8, 9]. For the multiphoton absorption of nonrotating HF [10], it has been found reasonable agreement between the two theories, however it has been recognized that quantum and classical results are similar only for a short interaction time [7, 11]. Recently, Constantinou and Nicolaides [12] have verified stabilization of the classical dissociation due to phase effects of two fields of commensurate frequencies. Those results open the possibility of further examination of the issue of classical correspondence with analogous quantum mechanical calculations.

In this work, we consider the quantum and classical dissociation dynamics of nonrotating diatomic molecules under infrared laser pulses. The molecule is assumed to be in a fixed vibrational level of its electronic ground-state, with adiabatic potential represented by the Morse oscillator. The laser-molecule interaction is given in the dipole approximation. The dipole function goes to zero for large nuclear separation, standing for neutral fragmentation. The classical calculations are performed considering Monte Carlo sampled trajectories. They are distributed according to the Husimi function of the correspondent vibrational state. This choice allows for an alternative repre-

sentation of the excited levels. The results are compared with the traditional one, which considers only trajectories with the energy of the eigenstate. To the quantum calculations, we apply the continuum expansion method, which involves Laguerre polynomials to describe the continuum coefficient. We have recently applied this technique to the study selective excitation of high overtones and to the investigation of multiphoton association [13, 14].

Two main physical quantities are used for the comparisons: the dissociation probability and the energy absorbed by the bound molecule. They can be defined unambiguously for both quantum and classical cases and are studied as a function of the laser parameters. The electric field is given by Gaussian pulses of commensurate frequencies. We investigate the role of amplitude, carrier frequency and relative phase on the dissociation probability. We also vary the initial vibrational state, with the objective of find reasonable quantum-classical correspondence. Through direct comparison of the quantum and classical theories, we investigate the role of relative phase and synchronization of multiple pulses on the dynamics. Special attention is paid to the analogies between coherent control and classical chaos.

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