Classical and quantum dissociation of a laser driven Morse molecule

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The multiphoton vibrational dissociation of Morse molecules has been examined nonperturbatively using Hamilton and Schroedinger time-dependent equations, for a range of laser pulse parameters. The time-dependent Schrodinger equation is solved by the state-specific expansion approach including the effects of the continuous spectrum. The focus of the work is on the dependence of the dissociation probability and rate on laser frequency and the effects of laser intensity, pulse shape and duration on it. The molecule is considered initially at its ground state. The comparison between the classical and quantum aspects of these dependencies raises fundamental issues of quantum-classical correspondence for open systems such as the interrelation of classical and quantum resonances and their influence on escape process as well as the impact of classical phase space features (i.e. sticking regions) on quantum dynamics. It is shown, first, that classical dynamics predicts the red shift phenomenon, according to which the maximum of the dissociation probability occurs at the 85the first excited state. Actually, the whole dependence of the quantum dissociation probability on laser frequency in this frequency regime is sufficiently reproduced by classical dynamics, surprisingly even for quite low laser intensities where only a quantum resonant peak is present. Second, it is found that the enhancement of stickiness effects, observed as the laser frequency increases, affects both classical and quantum dissociation rates in a similar fashion, demonstrating thus their impact on quantum dynamics. Finally, it is shown that both classical and quantum calculations predict strong influence of the pulse ramp on time on the dissociation probability only at frequencies lower than the frequency of maximum dissociation probability. A classical interpretation of this effect is given and its quantum mechanical implications are discussed.