Control of Transitions by Broadband Pulse Shaping in Stimulated Raman Spectroscopy

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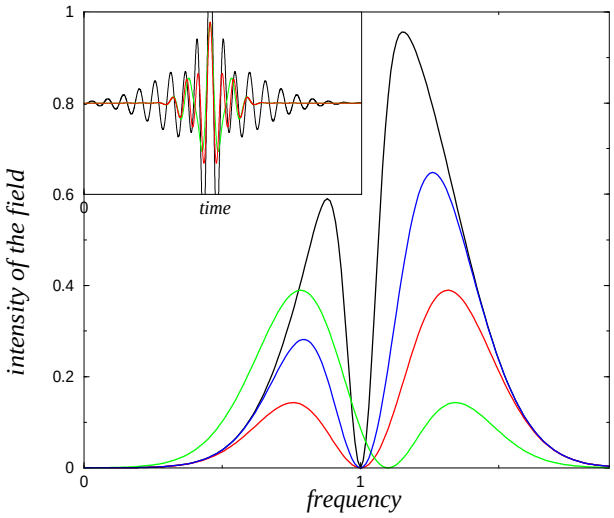
Control of atoms and molecules is of increasing demand in modern natural sciences. In stimulated Raman spectroscopy the control of vibrational degrees of freedom in molecules in liquid and gas phase is experimentally achieved by means of ultrafast optical pulse shaper and evolutionary learning algorithm [1, 2]. In this work a semiclassical model is used to investigate the possibility of selectively exciting one of two closely spaced, uncoupled Raman transitions. The condition for the bandwidth of the intense pump pulse that creates the Raman coherence is that it is much broader than frequency separation of the corresponding transitions. The duration of the pulse is about a typical molecular vibrational period (impulsive regime of interaction).

Pulse shapes are found that provide either enhancement or suppression of particular vibrational excitations [3]. Introduced in the frequency domain, they give high field intensity at the frequency of the transition to be excited and zero intensity at the frequency of the transition to be suppressed.



Such shapes results in no excitation of one of the transitions and effective excitation of another in the weak field regime. In strong fields numerical analysis reveals

that, to selectively excite one of two transitions with the designed pulse shapes it is necessary to suitably choose the field amplitude. Numerical solution with the proposed pulse shape is obtained taking into account the coupling between two, two-level systems. It shows that the mechanism of the selective mode excitation may relay on the coupling between vibrational modes within a molecule. If the direct excitation of a particular vibrational mode is weak owing to a weak oscillator strength, the excitation may be enhanced through the coupling to the other Raman active vibrational modes.

References

[1] B.J. Pearson, J.L. White, T.C. Weinacht, and P.H. Bucksbaum, Phys. Rev. A **63**, 063412 (2001).

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[3] S.A. Malinovskaya, P.H. Bucksbaum, and P.R. Berman, Phys. Rev. A 2003.