Off-resonant vibrational excitation: Orientational dependence and spatial control of photofragments - DTU Orbit (27/09/2019)

Off-resonant and resonant vibrational excitation with short intense infrared (IR) laser pulses creates localized oscillating wave packets, but differs by the efficiency of the excitation and surprisingly by the orientational dependence. Orientational selectivity of the vibrational excitation of randomly oriented heteronuclear diatomic molecules can be obtained under simultaneous irradiation by a resonant and an off-resonant intense IR laser pulse: Molecules with one initial orientation will be vibrationally excited, while those with the opposite orientation will be at rest. The orientation-dependent response to the IR fields is due to the anharmonicity of the potential. A subsequent ultraviolet laser pulse in resonance at the outer turning point of the vibrational motion can then dissociate the oscillating molecules, all with the same orientation, leading to spatial control of the photofragment distribution. (C) 2000 American Institute of Physics.

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