

Abstract Submitted
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Strong-field mechanism of molecular alignment based on transient nonadiabatic charge redistribution.¹ DMITRI ROMANOV, Department of Physics and Center for Advanced Photonics Research, Temple University, ROBERT LEVIS, Department of Chemistry and Center for Advanced Photonics Research, Temple University — A new mode of effective interaction of molecular rotational degrees of freedom with an intense, nonresonant, ultrashort laser pulse is explored. This new mode of impulsive-torque interaction replaces the traditional mechanism of molecular alignment based on linear anisotropic polarizability when the strong electric field of the pulse can cause transient nonadiabatic charge redistribution (TNCR) in larger molecules or molecular ions. We explore this new alignment mechanism on a generic model of a tight-binding diatomic molecule. The TNCR mode of effective laser interaction with the rotational degrees of freedom thoroughly changes the composition of the resulting rotational wavepacket and the dynamics of subsequent field-free alignment in the molecular ensemble. The rotational wavepacket emerging from the TNCR interaction is markedly contributed to by the plethora of states with higher rotational quantum numbers, in both perturbative and non-perturbative regime; the after-pulse alignment oscillations are out-of-phase with those resulting from the traditional interaction. This TNCR interaction mode opens a new class of alignment mechanisms associated with considerable nonresonant excitation or ionization of a molecule during the laser pulse.

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