A linear polar molecule in a two-color laser field: a symmetry analysis

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Synopsis We describe the field-dressed rotational dynamics of a linear polar molecule in a a non-resonant two-color cw laser field, and analyze the orientation and alignment as the field parameters are varied.

An experimental technique to induce molecular orientation is based on the use of non-resonant two-color laser fields [1], which couples pendular states with different parity due to the interaction with the molecular hyperpolarizability. In this work, we investigate the rotational dynamics of a linear polar molecule in a a non-resonant two-color cw laser field. We consider non-resonant two-color laser fields having parallel polarizations in the regime where the time-average approximation does not hold [2]. To do so, we increase the frequencies of the laser fields till the time-average over the rapid oscillations of the laser field becomes non-negligible. Note that we still assume that the laser field does not drive any electric, vibrational or rotational transitions in the molecule, which is described within the rigid-rotor approximation. Thus, the field-dressed rigid-rotor Hamiltonian includes the interaction of the two-color electric field with the permanent electric dipole moment, the polarizability, and the hyperpolarizability of the molecule.

In the regime beyond the time-average approximation, we explore the symmetries of the two-color electric field given by the biharmonic function [3] and analyze their impact on the field-dressed rotational dynamics. We investigate the rotational dynamics of a rigid-rotor molecule in detail as the parameters of the two-color electric field, i.e., the frequencies of the two electric field components, their relative phase, and their relative strength, are varied. We show that the alignment and orientation are symmetric and antisymmetric, respectively, as a function of the relative phase of the two components of the electric field. We also provide the field parameters needed to reach the largest orientation.

References

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