Controlling the rotational dynamics of molecules using combined laser pulses and static electric fields

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We present a theoretical study of mixed-field-orientation and alignment experiments of linear and asymmetric top polar molecules. In these experiments, pendular states were created by means of linearly polarized strong laser pulses combined with tilted weak electric fields. Within the rigid rotor approximation, we assume that the dc field couples with the dipole moment and the ac field by means of the polarizability anisotropy, and solve the time-dependent Schrödinger equation taking into account the time profile of the alignment pulse [1, 2]. Our results show that the adiabaticity of the mixed-field orientation depends on the avoided crossings that the adiabatic states suffer, the energy splitting of the states in a J-manifold at weak laser intensities, as well as, on the formation on the quasidegenate doublets at stronger ac-fields. These pendular doublets result in the transfer of population from a single occupied field-free rotational state into two strongly oriented and anti-oriented pendular states, reducing the overall orientation. Hence, we probe that, in general, the weak dc field orientation is not adiabatic and that a time-dependent description of this process is mandatory. Specifically, we compare our numerical results with the experimental observations of the mixed-field orientation of OCS, and we obtain a very good agreement for several field configurations [3]. For asymmetric top molecules, we show that the field-dressed dynamics is more complicated due to the level structure and avoided crossings, and that the conditions to achieve the adiabatic dynamics become more difficult to reach [2, 4]. For perpendicular fields, one gets pure alignment, our calculations reproduce the pendular wave-packet dynamics of the OCS molecule due to a picosecond laser pulse [5].

References

[1] J. J. Omiste and R. González-Férez, Phys. Rev. A **86**, 43437 (2012).

[2] J. J. Omiste and R. González-Férez, Phys. Rev. A **88**, 033416 (2013).

[3] J. Nielsen *et al.*, Phys. Rev. Lett. **108** (2012).

[4] J. J. Omiste *et al.*, Phys. Chem. Chem. Phys. **13**, 18815 (2011).

[5] S. Trippel *et al.*, arXiv:1401.6897.

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