CFEL SCIENCE

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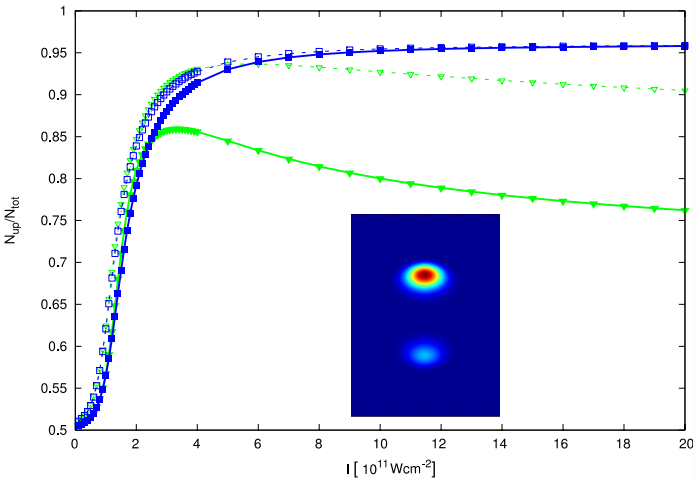
Juan J. Omiste Romero

Universidad de Granada, Spain

Theory of mixed-field orientation of linear molecules: Loss of adiabaticity

We present a theoretical study of mixed-field-orientation experiments of polar linear 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. Our results show that the adiabaticity of the mixed-field orientation depends on the avoided crossings that the states suffer, as well as, on the formation on the quasidegenate doublets that characterize the pendular regime. 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 measurements corresponding to the OCS molecule for different field configurations obtaining a good agreement [1]. This work complements our previous study for asymmetric top molecules exposed to tilted fields [2], where we have shown that an adiabatic description is not enough to explain the experimental results even for long laser pulses.



References

[1] Darko Dimitrovski, Mahmoud Abu-samha, Lars Bojer Madsen, Frank Filsinger, Gerard Meijer, Jochen Küpper, Lotte Holmegaard, Line Kalhøj, Jens H. Nielsen, and Henrik Stapelfeldt. Ionization of oriented carbonyl sulfide molecules by intense circularly polarized laser pulses. *Phys. Rev. A*, 83, 023405, 2011.
[2] J. J. Omiste, M. Gärttner, P. Schmelcher, R. González-Férez, L. Holmegaard, J. H. Nielsen, H. Stapelfeldt, and J. Küpper. Theoretical description of adiabatic laser alignment and mixed-field orientation: the need for a non-adiabatic model. *Phys. Chem. Chem. Phys.*, 13, 18815–18824, 2011.

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