NON-MARKOVIAN AND NON SECULAR COLLISIONAL DYNAMICS IN THE DISSIPATION OF LASER-ALIGNED MOLECULES

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Studying molecular alignment at very short times enables to measure the influence of some mechanisms that affect the evolution of the open system [1]. Here we experimentally, based on molecular dynamics predictions, study the limits of the two widely used secular and Markov approximations. The first one neglects all the exchanges between the quantum coherences that oscillate at different frequencies [2] and the second considers that all collisions are complete in the time interval of observation. To test the limits of both these approximations, we study the influence of the pressure on the decay of the molecular alignment soon after the excitation by a femtosecond laser pulse. Four systems have been chosen, involving all the situations at the investigated time scale : Practically secular and Markovian (HCl diluted in Helium) or non-Markovian (pure HCl), nonsecular and Markovian (CO_2 diluted in Helium) or non-Markovian (pure CO_2) [3,4]. We show here that, depending the studied system, those approximations lead to important errors on the predicted relaxation at short times, before becoming valid after tens of picoseconds. Those nonsecular and non-Markovian effects might be observed in the future on more complex molecular systems than linear rotors investigated in the present study.

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