Inhibited, Explosive and Anisotropic Relaxation in a Gas of Molecular Super-Rotors

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Recently, several femtosecond laser techniques have been developed [1–6] that are capable of bringing gas molecules to extremely fast rotation in a very short time, while keeping their translational motion intact and relatively slow. We investigate collisional equilibration dynamics of this new state of molecular gases, and find that it follows a remarkable generic scenario. The route to equilibrium starts with a durable metastable ‘gyroscopic stage’, in the course of which the molecules maintain their fast rotation and orientation of the angular momentum through many collisions. The inhibited rotational-translational relaxation is characterized by a persistent anisotropy in the molecular angular distribution, and is manifested in the long-lasting optical birefringence, and anisotropic diffusion in the gas. After a certain induction time, the ‘gyroscopic stage’ is abruptly terminated by a self-accelerating explosive rotational-translational (RT) energy exchange leading the gas towards the final thermal equilibrium. We illustrate our conclusions by direct Molecular Dynamics simulation of super-rotors in several gases consisting of common linear molecules (such as N_2, O_2 and CO_2).

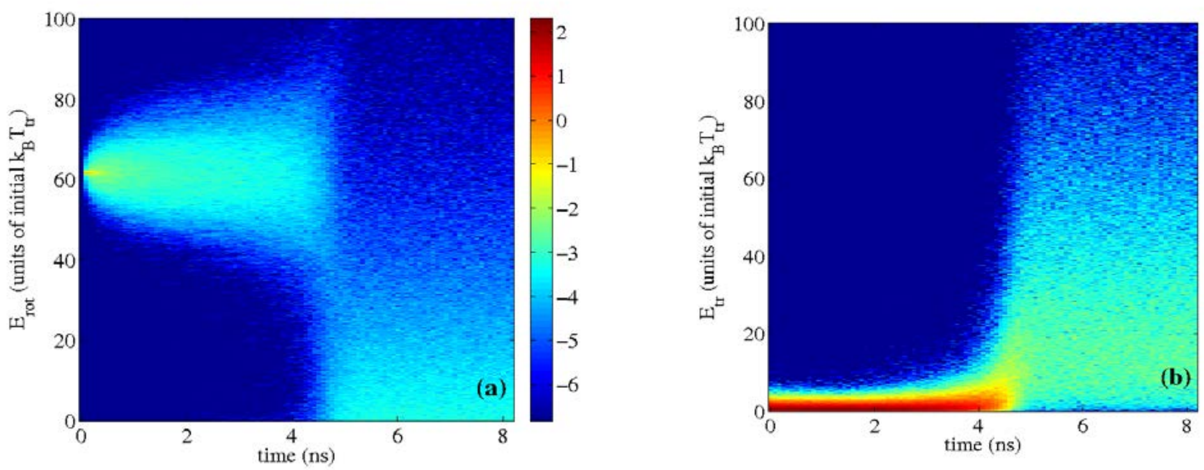

FIGURE: Density plots for time-dependent distributions of the rotational and translational energy of N_2 gas (panels (a) and (b), respectively). The molecules are initially at ambient conditions and are centrifuged into rotation with J = 80. An explosive RT transition towards the thermal Maxwell-Boltzmann distribution of the rotationally-heated gas is clearly seen near 5 ns.

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