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Laser-induced rotation of molecules in He-nanodroplets: Revivals and breaking-loose

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High resolution infrared and microwave spectra of molecules dissolved in liquid helium nanodroplets display discrete rotational structure – a unique feature explained as the result of frictionless rotation of molecules adiabatically followed by a local solvation shell of He atoms [1]. The frictionless behavior did, however, not manifest itself in recent time-resolved experiments, based on femtosecond laser-induced molecular alignment techniques. In particular, the transient recurrences of alignment characteristic of freely rotating gas phase molecules were absent [2,3]. In this talk we present experiments on femtosecond laser-induced alignment of iodine molecules embedded in helium nanodroplets showing striking new phenomena [4]: 1) At low to moderate fluences the alignment pulse sets the molecule and a non-superfluid fraction of the He droplet into coherent motion. The coherence, although decaying, persists for many hundreds of picoseconds – long enough to allow the composite molecule-He-shell system to exhibit rotational revivals. Our experimental observations are rationalized by classical considerations and quantum theory based on the angulon quasiparticle [5]. 2) With increasing fluence the revivals disappear – instead, rotational dynamics as rapid as for an isolated molecule is observed during the first few picoseconds. Classical calculations trace this phenomenon to transient decoupling of the molecule from its He shell. **References** [1] M. Y. Choi, *et al.*, Int. Rev. Phys. Chem. **25**, 15 (2006). [2] D. Pentlehner *et al.*, *Phys. Rev. Lett.* **110,** 093002 (2013). [3] L. Christiansen *et al.*, *Phys. Rev. A.* **92,** 053415 (2015). [4] B. Shepperson *et al., submitted* (2017). [5] R. Schmidt and M. Lemeshko, Phys. Rev. Lett. **114** 203001 (2015).