Laser-induced alignment of molecules in He-nanodroplets: Long-time coherence, revivals, and breaking-free

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Synopsis Laser-induced alignment of molecules inside helium droplets has been performed using femtosecond alignment experiments. The results indicate that at low fluence of the fs alignment pulse, the molecule and its solvation shell can be set into coherent collective rotation lasting long enough to form revivals. With increasing fluence, however, the revivals disappear instead, rotational dynamics as rapid as for an isolated molecule is observed during the first few picoseconds.

Rotation of molecules embedded inside He nanodroplets is explored by fs laser induced alignment experiments. We demonstrate that at low fluence of the fs alignment pulse, the molecule and its solvation shell can be set into coherent collective rotation lasting long enough to form revivals. With increasing fluence, however, 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. A cartoon depicting the laser-induced rotational dynamics for iodine molecules solvated inside helium droplets initiated by a weak and a strong kick pulse is shown in Figure 1. Our results open novel opportunities for studying non-equilibrium solute-solvent dynamics and quantum thermalization.

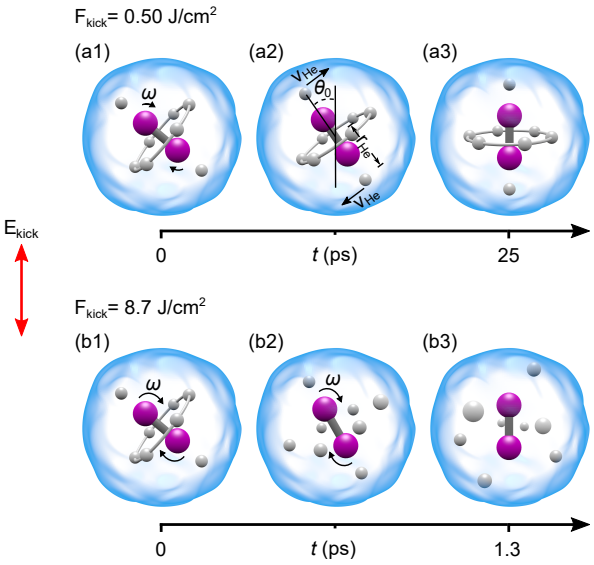

Figure 1. Schematic illustration of laser-induced rotation of iodine molecules inside He droplets for a weak [(a1)-(a3)] and a strong [(b1)- (b3)] kick pulse. (a2) illustrates parameters used in the classical model.

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