Femtosecond timed imaging of rotation of alkali dimers on the surface of helium nanodroplets

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Alkali dimers, Ak_2, residing on the surface of helium nanodroplets, are doubly ionized by an intense fs laser pulse leading to Coulomb explosion into two Ak^+ fragment ions. We show that the kinetic energy of these fragment ions can be used to identify if the dimers were initially populated in the X $^1\Sigma_g^+$ ground state or in the lowest-lying triplet state a $^3\Sigma_u^+$ [1, 2]. We use the quantum-state-sensitive detection to investigate rotational dynamics of dimers in either the X or in the a state.

The work presented here focuses on the time-dependent alignment of Na_2, K_2, and Rb_2 on He nanodroplet surfaces. The dimers are set into rotation by a 1 ps alignment pulse and their instantaneous spatial orientation measured by Coulomb explosion with an intense delayed 50 fs probe pulse. The measured alignment traces show distinct periodic features that differs qualitatively from that expected for freely rotating gas phase molecules. Instead, the observed alignment dynamics of Na_2 (see Fig. 1) and of K_2 in the a $^3\Sigma_u^+$ state agree with that obtained from a 2D rigid rotor model, strongly indicating that the rotation of each dimer occurs in a plane – defined by the He droplet surface [3, 4].

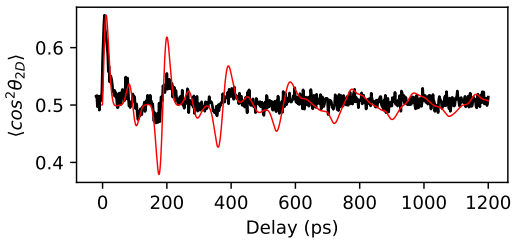

Figure 1: Time-dependent degree of alignment for Na_2 in the a $^3\Sigma_u^+$ state (black) and the expected alignment trace from a 2D rigid rotor model (red) with inhomogeneous broadening.

References:

[1] H. H. Kristensen, *et al.*, Phys. Rev. Lett. **128** (2022), 093201
[2] H. H. Kristensen, *et al.*, Phys. Rev. A **107** (2023), 023104
[3] L. Kranabetter, *et al.*, Phys. Rev. Lett. **131** (2023), 053201
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