An angulon quasiparticle perspective on impulsive molecular alignment in ^{4}He nanodroplets

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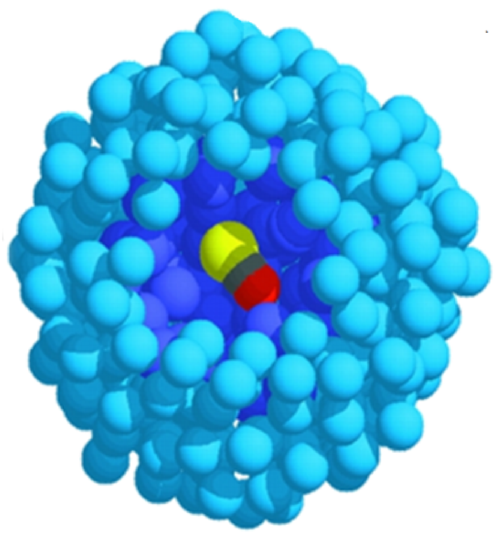
Molecular impurities

A molecular impurity embedded into a helium nanodroplet: a **fully controllable quantum system** to explore angular momentum dynamics **far from equilibrium**.

Temperature $\sim 0.4\,\mathrm{K}$

Droplets are superfluid

Easy to produce



Free of perturbations

Only rotational motion

Easy to manipulate by a laser

Image from: S. Grebenev *et al.*, Science **279**, 2083 (1998).

Interaction with an off-resonant laser pulse:

$$\hat{H}_{\mathrm{laser}} = -\frac{1}{4}\Delta\alpha E^2(t)\cos^2\hat{\theta}$$

Dynamical alignment of molecules

- Kick pulse, aligning the molecule.
- Probe pulse, destroying the molecule.
- Fragments are imaged, reconstructing alignment as a function of time.
- Averaging over multiple realizations, and varying the time between the two pulses, one gets

$$\left\langle \cos^2\hat{\theta}_{2\mathrm{D}} \right\rangle (t)$$

with:

$$\cos^2\hat{\theta}_{2\mathrm{D}} \equiv \frac{\cos^2\hat{\theta}}{\cos^2\hat{\theta}+\sin^2\hat{\theta}\sin^2\hat{\phi}}$$

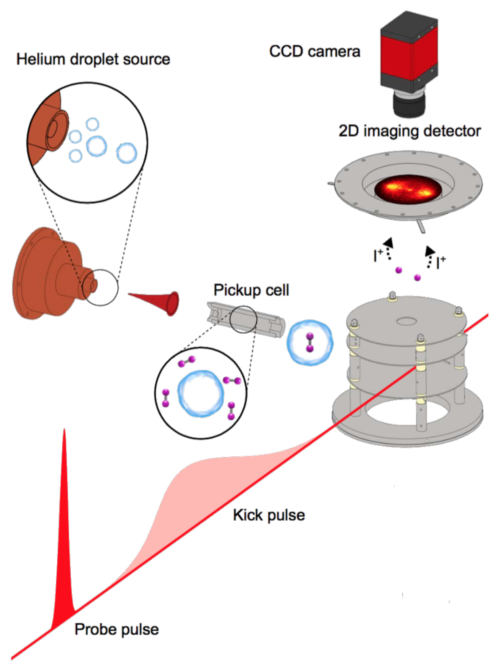


Image from: B. Shepperson et al., Phys. Rev. Lett. **118**, 203203 (2017).

The angulon

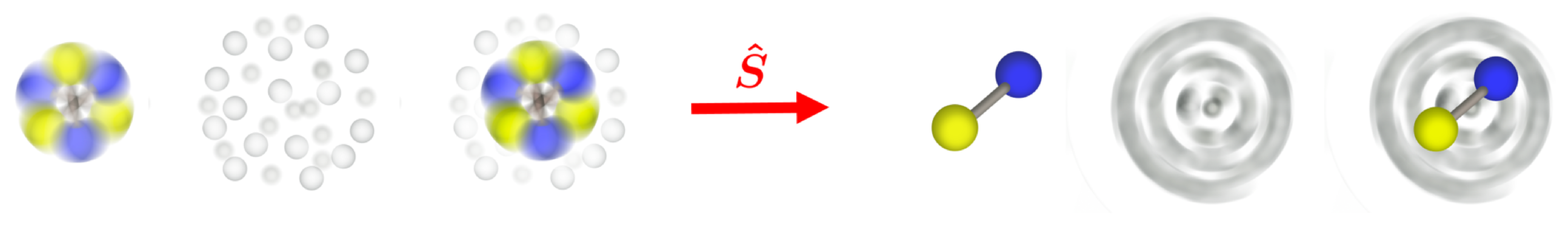
A slowly rotating linear molecule interacting with a bosonic bath can be described in the frame co-rotating with the molecule by the following Hamiltonian:

$$\hat{H} = B(\hat{\mathbf{L}}-\hat{\mathbf{\Lambda}})^2 + \sum_{k\lambda\mu}\omega_k \hat{b}^\dagger_{k\lambda\mu}\hat{b}_{k\lambda\mu} + \sum_{k\lambda} V_{k\lambda}(\hat{b}^\dagger_{k\lambda 0}+\hat{b}_{k\lambda 0}),$$

> The **angulon quasiparticle**: a quantum rotor dressed by a field of many-body excitations.

We study dynamics via a time-dependent variational Ansatz:

$$|\psi_{LM,i}(t)\rangle = \hat{U}\left(g_{LM}(t)\,|LM0\rangle_{\mathrm{mol}} + \sum_{k\lambda n}\alpha_{k\lambda n}(t)\,|LMn\rangle_{\mathrm{mol}}\,\hat{b}^\dagger_{k\lambda n}\right)|i\rangle_{\mathrm{bos}}$$



laboratory frame **rotating frame**

References

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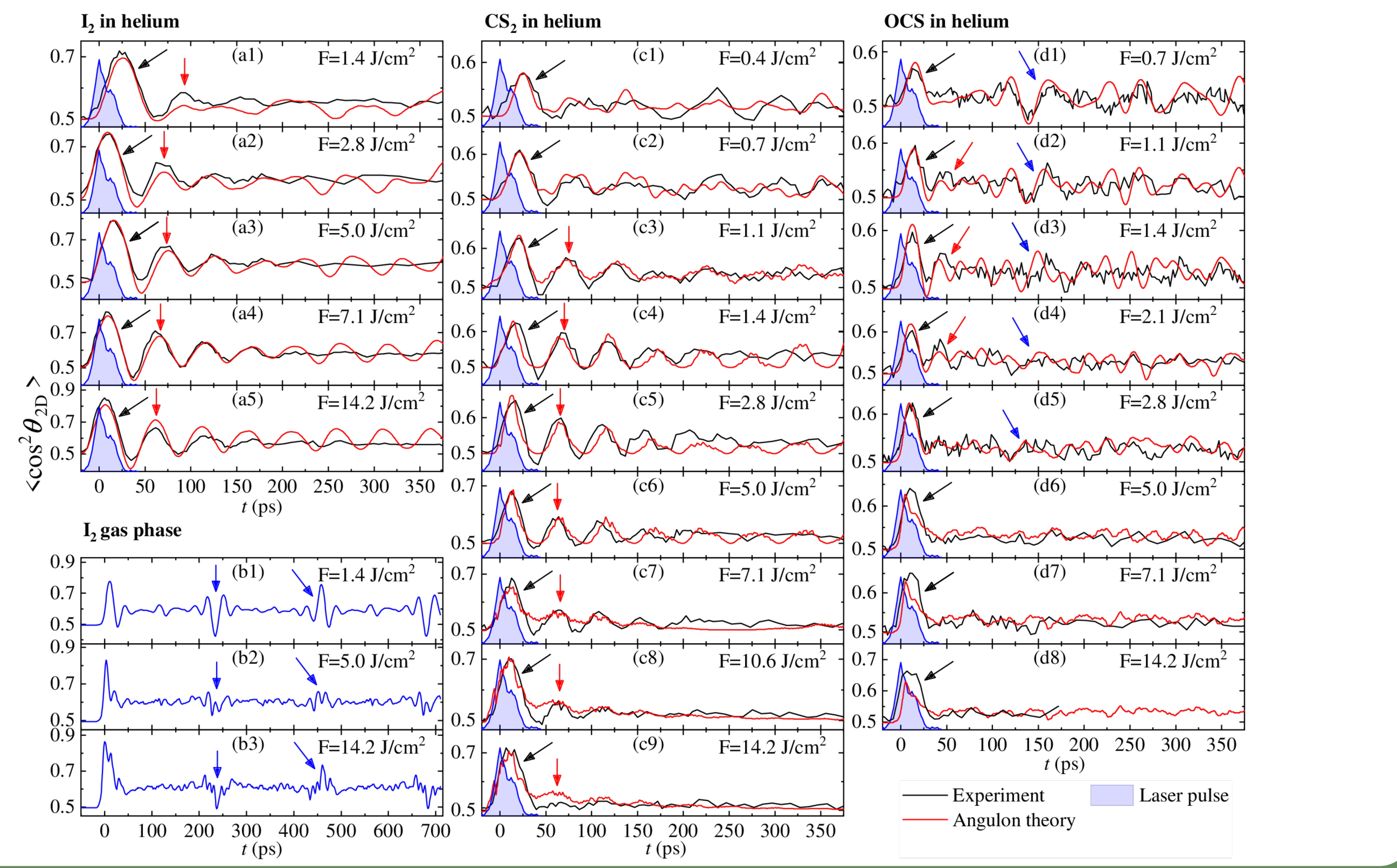
Experiments vs. theory: alignment

Comparison with experimental data from Stapelfeldt group, Aarhus University, for different molecules (I_2, CS_2 and OCS) and different fluences of the aligning pulse:



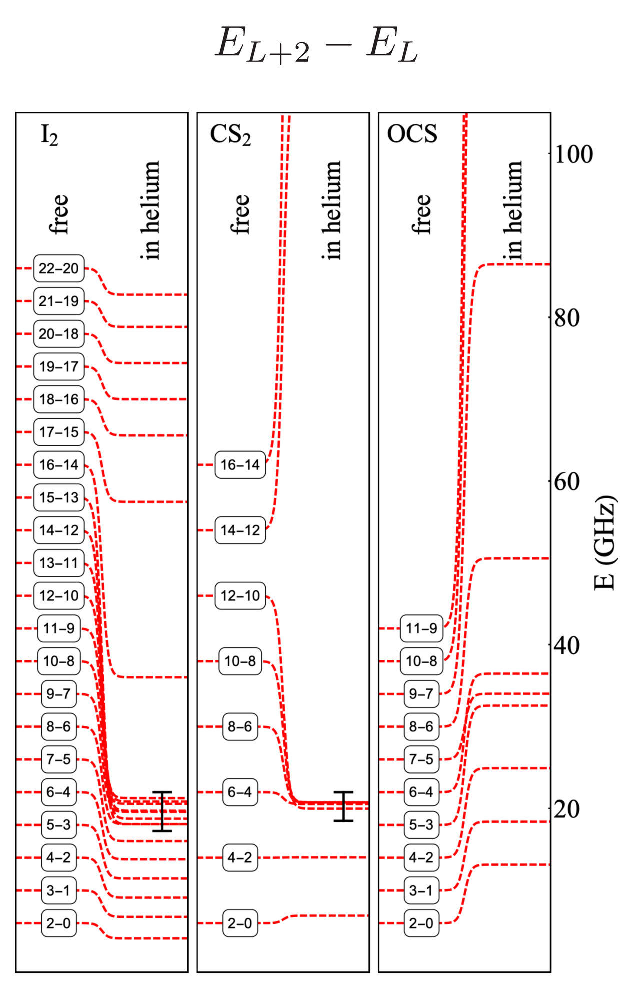
Experiments vs. theory: spectrum

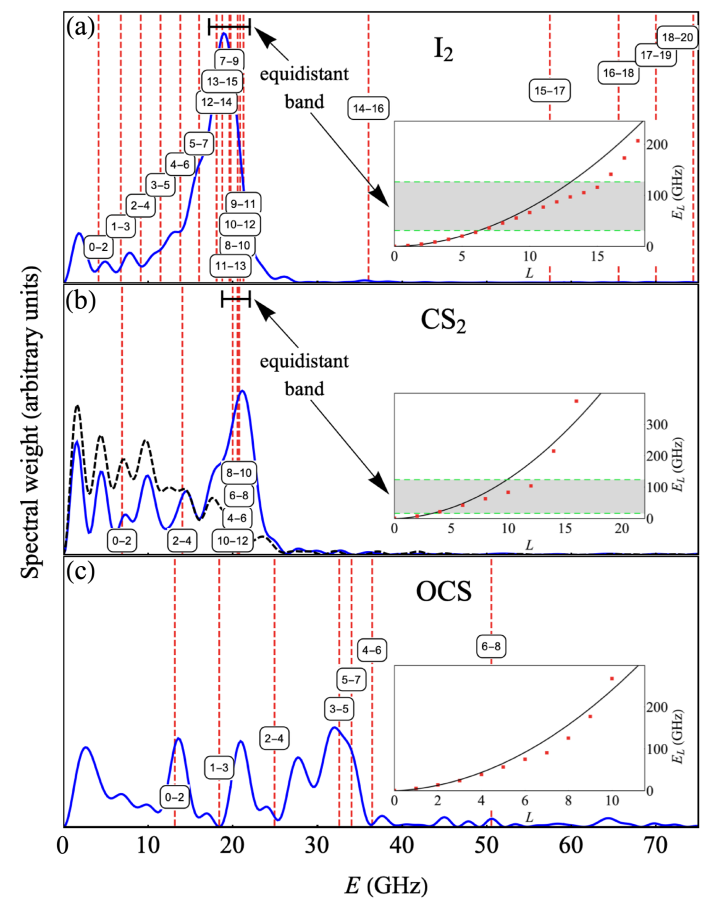
The rotational level structure is modified by the helium medium: one gets **rotational constant renormalisation** ($B \to B^*$) and **centrifugal distortion** (D^*):

- Free molecule: $E_L = BL(L+1)$
- Molecule in helium: $E_L = B^*L(L+1) - D^*[L(L+1)]^2$

The Fourier transform of the measured alignment cosine $\langle\cos^2\hat{\theta}_{2\mathrm{D}}\rangle(t)$ is dominated by $(L)-(L+2)$ transitions. How is it affected when the level structure changes?

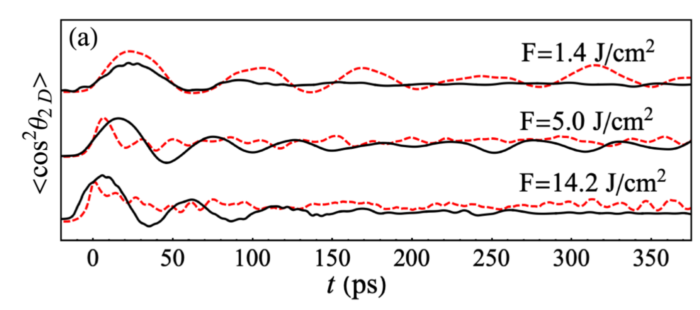
$$E_{L+2}-E_L$$



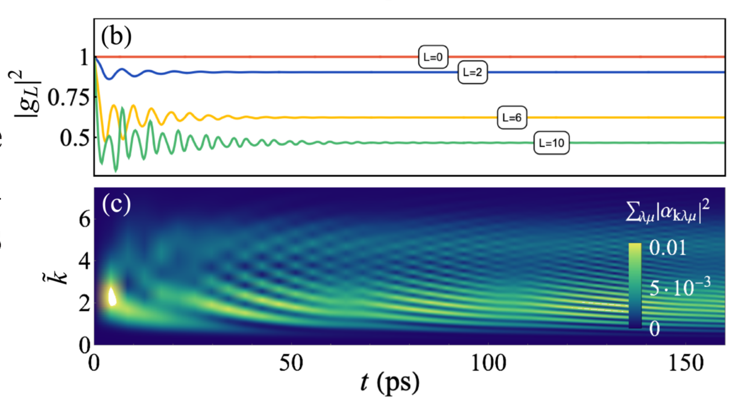


Many-body dynamics of angular momentum

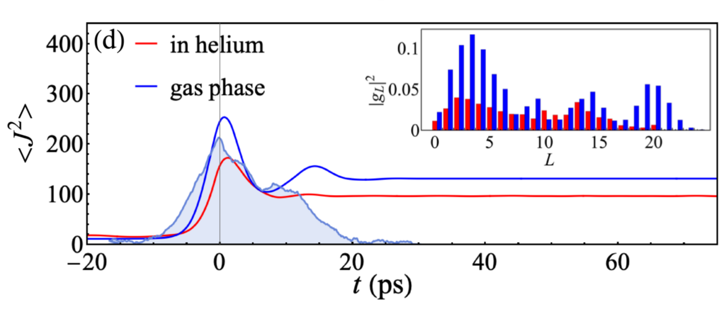
i) Can one explain the observed dynamics only by means of the **modified spectrum**? **No.** Red dashed lines (only renormalised levels) vs. solid black line (full many-body treatment).



ii) How long does it take for a molecule to equilibrate with the helium environment and form an angulon quasiparticle? This requires **tens of ps**; which is also the timescale of the laser!



iii) Effect of superfluid helium on angular momentum dynamics: it **prevents** the rotational angular momentum of the molecule from increasing as rapidly as it would in the gas phase.



Conclusions

- **A novel kind of pump-probe spectroscopy**, based on impulsive molecular alignment in the laboratory frame, providing access to the structure of highly excited rotational states.
- Superfluid bath leads to **formation of robust long-wavelength oscillations** in the molecular alignment.
- Our theoretical model allows us to interpret this behavior in terms of the dynamics of angulon quasiparticles, shedding light onto **many-particle dynamics** of angular momentum **at picosecond timescales**.