

# **Far-from-equilibrium dynamics of molecules in $^4\text{He}$ nanodroplets: a quasiparticle perspective**

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Institute of Science and Technology Austria

Universidade Federal do Rio Grande do Norte — Natal, April 16th, 2020

# Quantum impurities

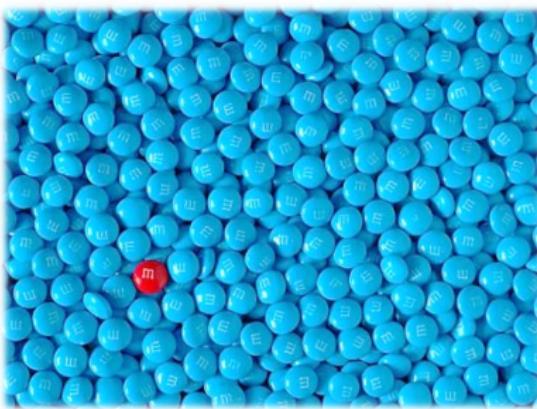
E

One particle (or a few particles)  
interacting with a many-body  
environment.

- Condensed matter
- Chemistry
- Ultracold atoms: tunable interaction with either bosons or fermions.

A prototype of a many-body system.

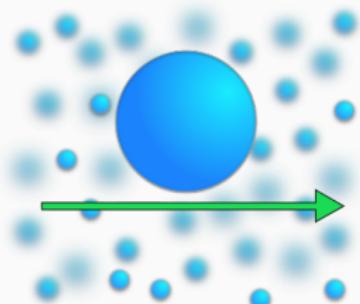
How are the properties of the  
particle modified by the interaction?



# Quantum impurities

**Structureless impurity:** translational degrees of freedom/linear momentum exchange with the bath.

Most common cases: electron in a solid, atomic impurities in a BEC.



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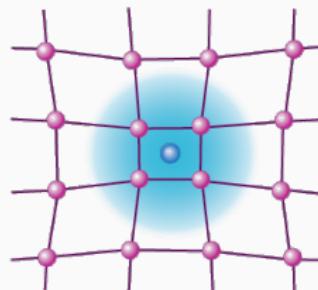


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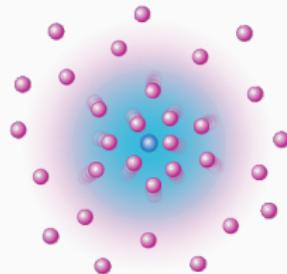


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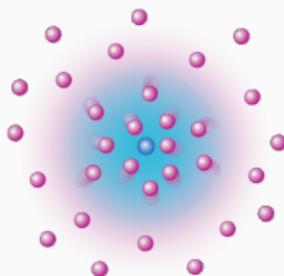
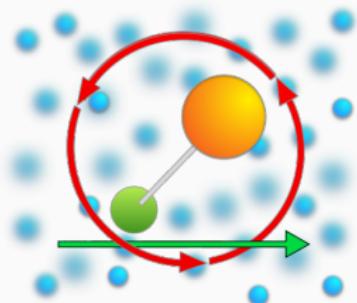


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**Composite impurity (e.g. a molecule):** translational *and* rotational degrees of freedom/linear and angular momentum exchange.

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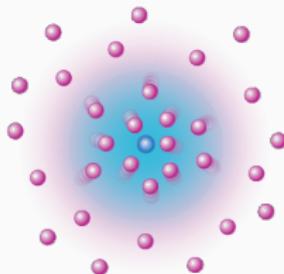
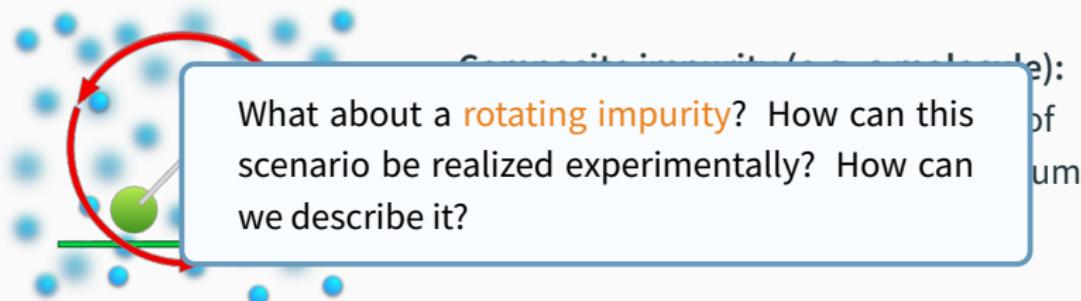


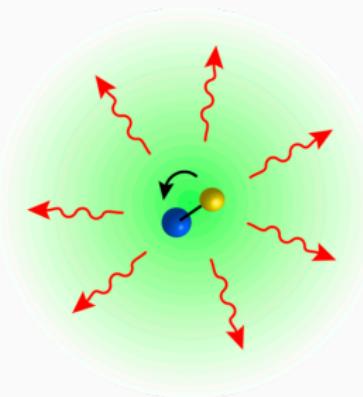
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# Composite impurities: where to find them

Strong motivation for the study of composite impurities comes from many different fields. Composite impurities can be realized as:

- Ultracold molecules and ions.



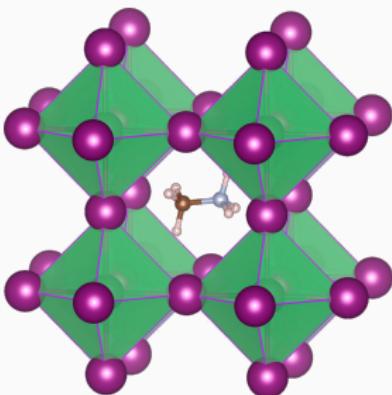
B. Midya, M. Tomza, R. Schmidt, and M. Lemeshko, Phys. Rev. A 94, 041601(R) (2016).

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Strong motivation for the study of composite impurities comes from many different fields. Composite impurities can be realized as:

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- Rotating molecules inside a 'cage' in **perovskites**.



T. Chen et al., PNAS **114**, 7519 (2017).

J. Lahnsteiner et al., Phys. Rev. B **94**, 214114 (2016).

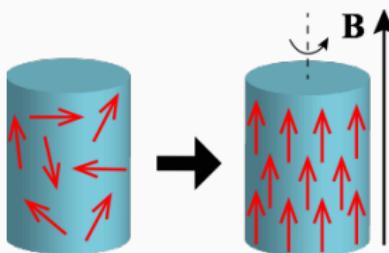
Image from: C. Eames et al, Nat. Comm. **6**, 7497 (2015).

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- Angular momentum transfer from the **electrons** to a **crystal lattice**.



J.H. Mentink, M.I. Katsnelson, M. Lemeshko, “*Quantum many-body dynamics of the Einstein-de Haas effect*”, Phys. Rev. B **99**, 064428 (2019).

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- Angular momentum transfer from the **electrons** to a **crystal lattice**.
- **Molecules** embedded into helium nanodroplets.

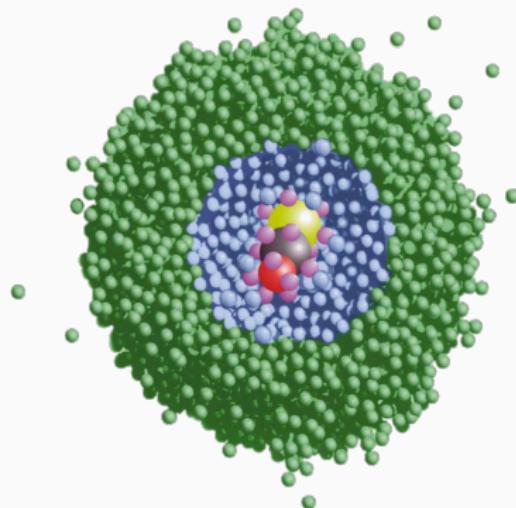
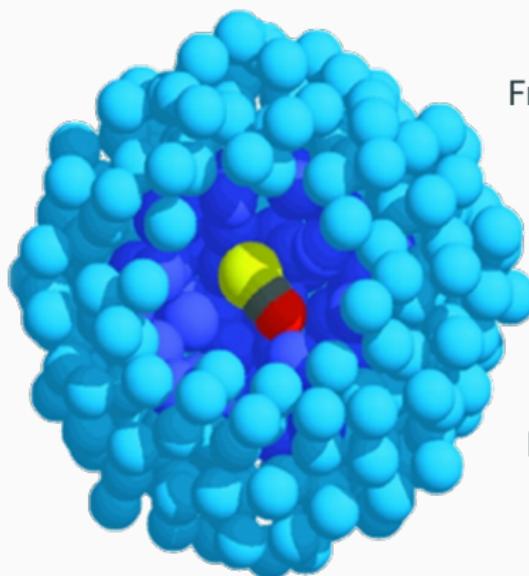


Image from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

# Molecules in helium nanodroplets

A molecular impurity embedded into a helium nanodroplet: a controllable system to explore angular momentum redistribution in a many-body environment.



Temperature  $\sim 0.4\text{K}$

Droplets are superfluid

Easy to produce

Free of perturbations

Only rotational degrees of freedom

Easy to manipulate by a laser

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

# Molecules in helium nanodroplets

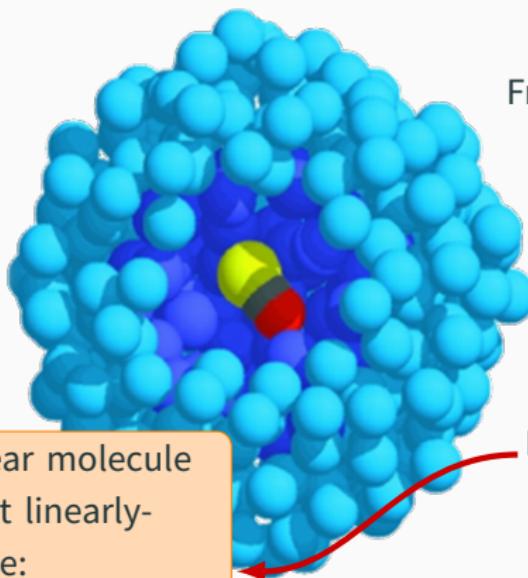
A molecular impurity embedded into a helium nanodroplet: a controllable system to explore angular momentum redistribution in a many-body environment.

Temperature  $\sim 0.4\text{K}$

Droplets are superfluid

Interaction of a linear molecule with an off-resonant linearly-polarized laser pulse:

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



Free of perturbations

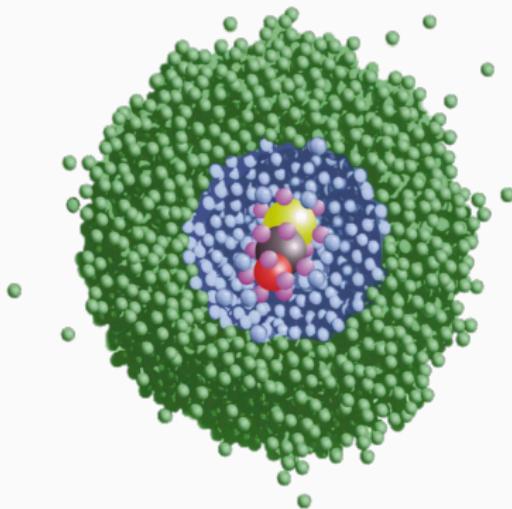
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# Rotational spectrum of molecules in He nanodroplets

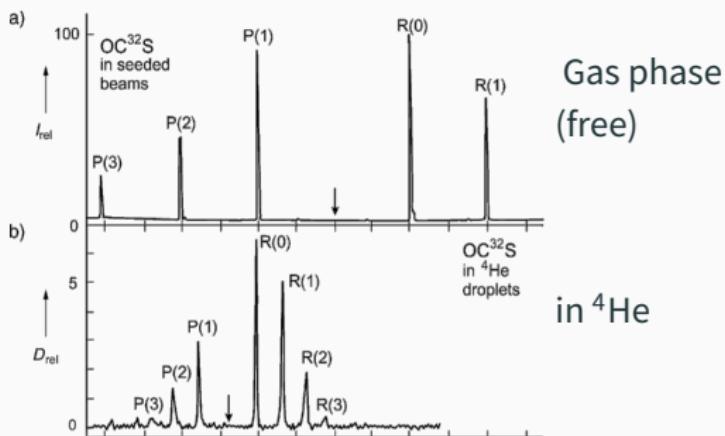
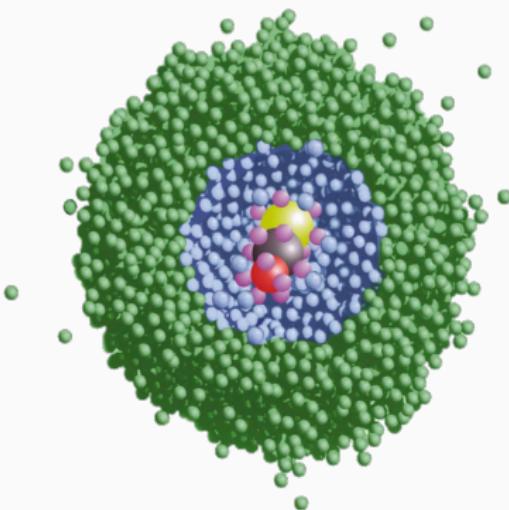
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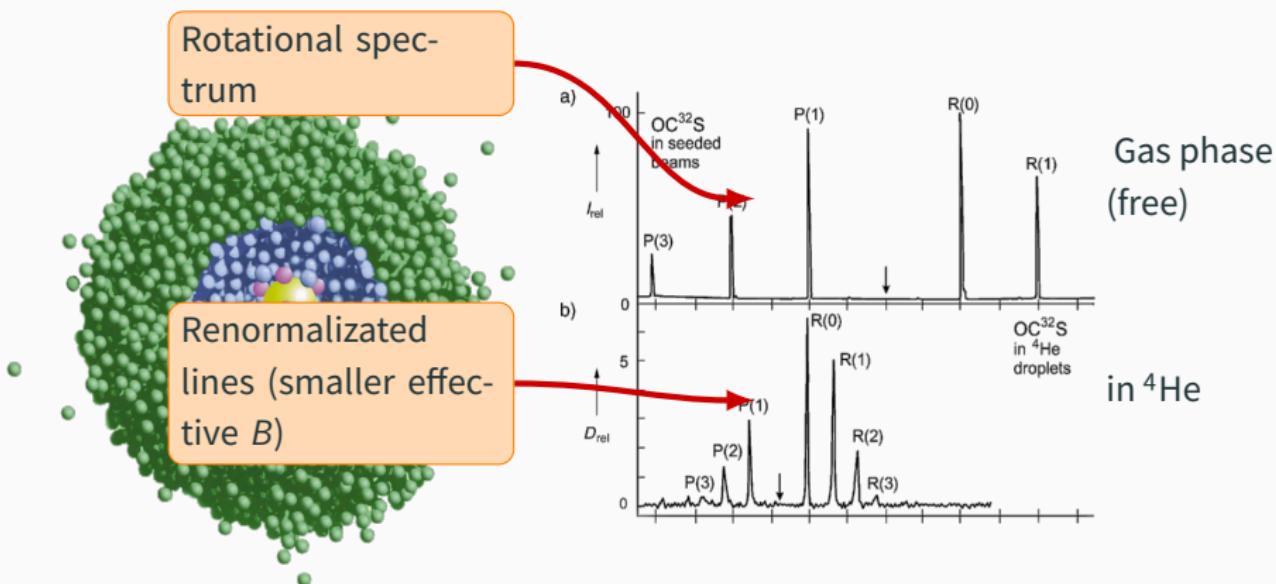
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Molecules embedded into helium nanodroplets: rotational spectrum



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# Dynamical alignment of molecules in He nanodroplets

## Dynamical alignment experiments

(Stapelfeldt group, Aarhus University):

- **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

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

with:

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

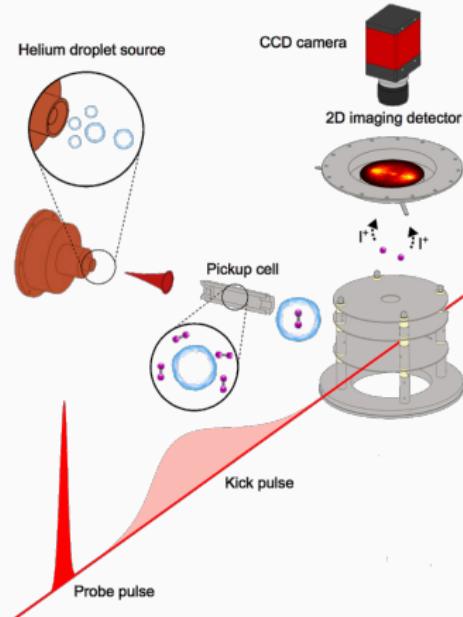


Image from: B. Shepperson *et al.*, Phys. Rev. Lett. 118, 203203 (2017).

# Dynamical alignment of molecules in He nanodroplets

A simpler example: a **free** molecule interacting with an off-resonant laser pulse

$$\hat{H} = B\hat{\mathbf{J}}^2 - \frac{1}{4}\Delta\alpha E^2(t) \cos^2 \hat{\theta}$$

When acting on a **free molecule**, the laser excites in a short time many rotational states ( $L \leftrightarrow L + 2$ ), creating a **rotational wave packet**:

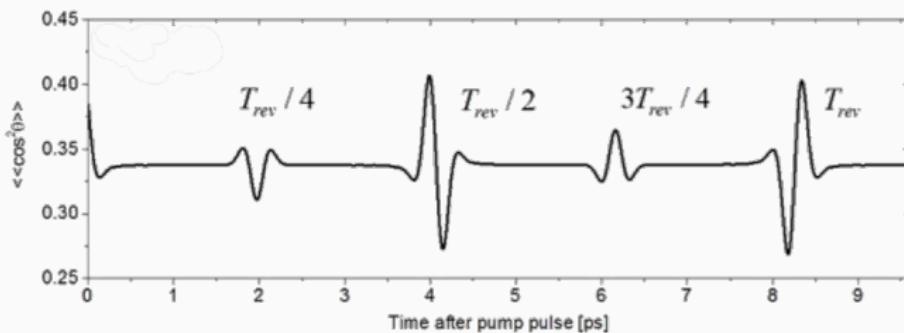
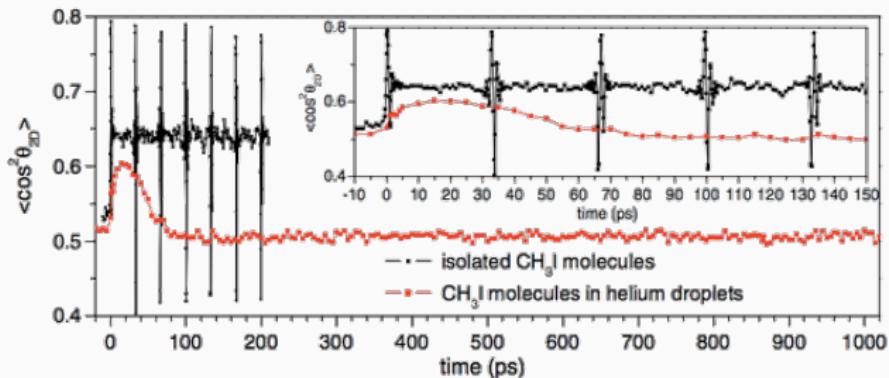


Image from: G. Kaya *et al.*, Appl. Phys. B 6, 122 (2016).

Movie

# Dynamical alignment of molecules in He nanodroplets

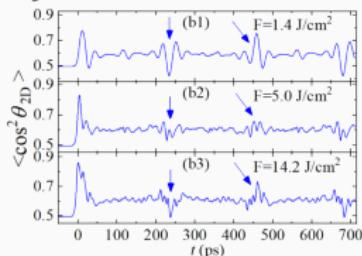
Effect of the environment is substantial: free molecule vs. **same molecule in He.**



Stapelfeldt group, Phys. Rev. Lett. 110, 093002 (2013).

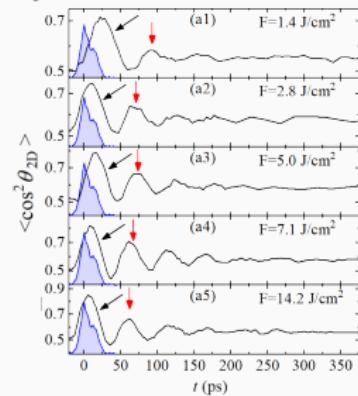
# Dynamical alignment of molecules in He nanodroplets

## Dynamics of isolated I<sub>2</sub> molecules



Experiment: Stapelfeldt group (Aarhus University).

## Dynamics of I<sub>2</sub> molecules in helium



Effect of the environment is substantial:

- The peak of **prompt alignment** doesn't change its shape as the fluence  $F = \int dt I(t)$  is changed.
- The revival structure differs from the gas-phase: revivals with a 50ps period of **unknown origin**.
- The oscillations appear weaker at **higher fluences**.
- An intriguing **puzzle**: not even a qualitative understanding. Monte Carlo? He-DFT?

## Quasiparticle approach

The quantum mechanical treatment of many-body systems is always challenging. How can one simplify the quantum impurity problem?

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The quantum mechanical treatment of many-body systems is always challenging. How can one simplify the **quantum impurity** problem?

**Polaron:** an electron dressed by a field of many-body excitations.

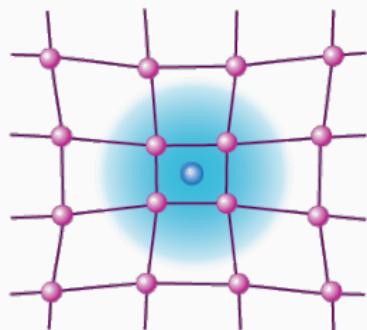
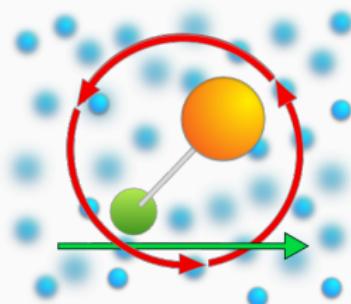


Image from: F. Chevy, Physics **9**, 86.

**Angulon:** a quantum rotor dressed by a field of many-body excitations.



R. Schmidt and M. Lemeshko, Phys. Rev. Lett. **114**, 203001 (2015).

R. Schmidt and M. Lemeshko, Phys. Rev. X **6**, 011012 (2016).

Yu. Shchadilova, "Viewpoint: A New Angle on Quantum Impurities", Physics **10**, 20 (2017).

# The Hamiltonian

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

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

Notation:

- $\hat{\mathbf{L}}$  the total angular-momentum operator of the combined system, consisting of a molecule and helium excitations.
- $\hat{\mathbf{\Lambda}}$  is the angular-momentum operator for the bosonic helium bath, whose excitations are described by  $\hat{b}_{k\lambda\mu}/\hat{b}_{k\lambda\mu}^\dagger$  operators.
- $k\lambda\mu$ : angular momentum basis.  $k$  the magnitude of linear momentum of the boson,  $\lambda$  its angular momentum, and  $\mu$  the z-axis angular momentum projection.
- $\omega_k$  gives the dispersion relation of superfluid helium.
- $V_{k\lambda}$  encodes the details of the molecule-helium interactions.

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- $k\lambda\mu$ : angular momentum basis.  $k$  the magnitude of linear momentum of

Compare with the Lee-Low-Pines Hamiltonian

$$\hat{H}_{LLP} = \frac{(\mathbf{P} - \sum_{\mathbf{k}} \mathbf{k} \hat{b}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}})^2}{2m_I} + \sum_{\mathbf{k}} \omega_{\mathbf{k}} \hat{b}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \frac{g}{V} \sum_{\mathbf{k}, \mathbf{k}'} \hat{b}_{\mathbf{k}'}^\dagger \hat{b}_{\mathbf{k}'}$$

## Dynamics: time-dependent variational Ansatz

We describe dynamics using a **time-dependent variational** Ansatz, including excitations up to one phonon:

$$|\psi_{LM}(t)\rangle = \hat{U}(\mathbf{g}_{LM}(t) |0\rangle_{\text{bos}} |LM0\rangle + \sum_{k\lambda n} \alpha_{k\lambda n}^{LM}(t) b_{k\lambda n}^\dagger |0\rangle_{\text{bos}} |LMn\rangle)$$

**Lagrangian** on the variational manifold defined by  $|\psi_{LM}\rangle$ :

$$\mathcal{L} = \langle \psi_{LM} | i\partial_t - \hat{\mathcal{H}} | \psi_{LM} \rangle$$

Euler-Lagrange **equations of motion**:

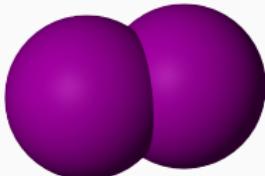
$$\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{x}_i} - \frac{\partial \mathcal{L}}{\partial x_i} = 0$$

where  $x_i = \{g_{LM}, \alpha_{k\lambda n}^{LM}\}$ . We obtain a **differential system**

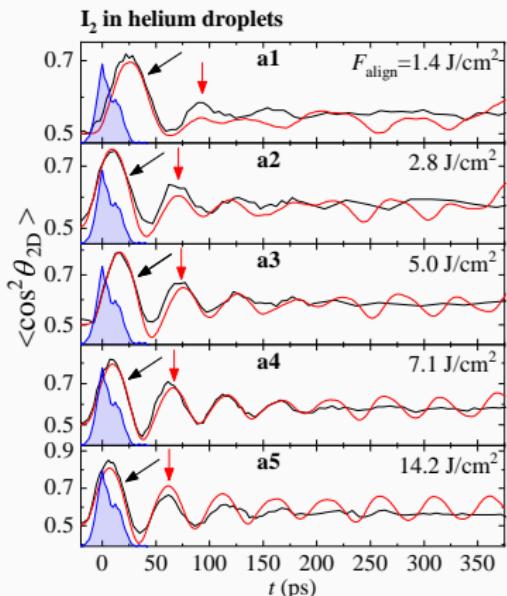
$$\begin{cases} \dot{g}_{LM}(t) = \dots \\ \dot{\alpha}_{k\lambda n}^{LM}(t) = \dots \end{cases}$$

to be solved numerically; in  $\alpha_{k\lambda n}$  the momentum  $k$  needs to be discretized.

## Theory vs. experiments: I<sub>2</sub>



Comparison with experimental data from Stapelfeldt group, Aarhus University, for different molecules: I<sub>2</sub>.

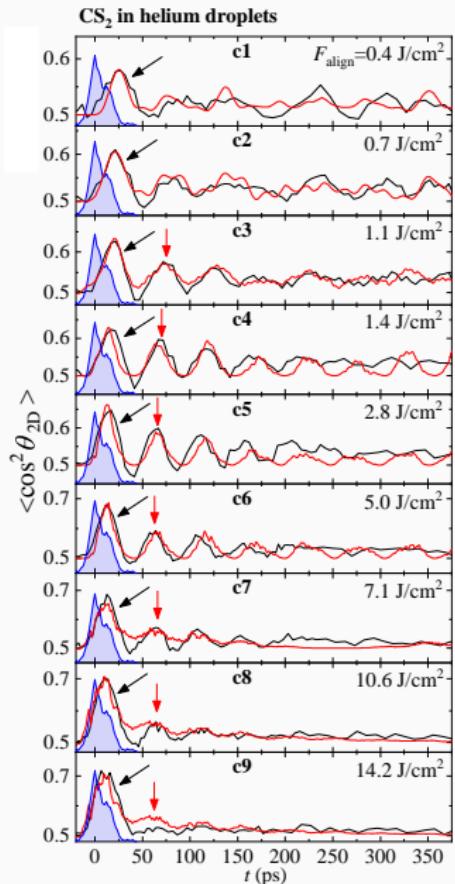


Generally good agreement for the main features in experimental data:

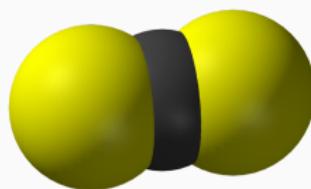
- Oscillations with a period of 50ps, growing in amplitude as the laser fluence is increased.
- Oscillations decay: at most 4 periods are visible.
- The width of the first peak does not change much with fluence.

— Experiment      ■ Laser pulse  
— Angulon theory

# Theory vs. experiments: $CS_2$



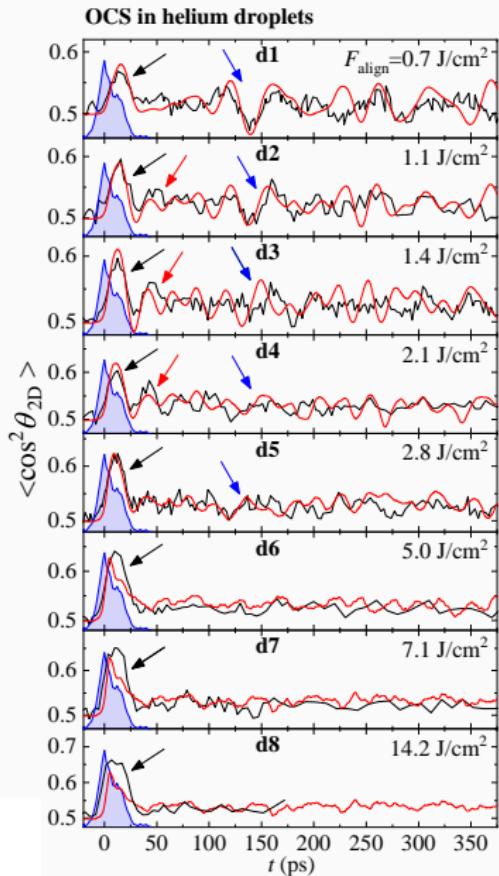
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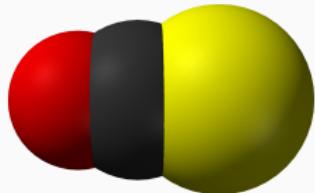
- Again, a persistent oscillatory pattern.
- For higher values of the fluence the oscillatory pattern disappears.

— Experiment      — Laser pulse  
— Angulon theory

# Theory vs. experiments: OCS



Comparison with experimental data from Stapelfeldt group, Aarhus University, for different molecules: OCS.



- Unfortunately the data is noisier.
- Oscillatory pattern not present, except in a couple of cases where one weak oscillation might be identified.

— Experiment      ■ Laser pulse  
— Angulon theory

- Can we shed light on the origin of oscillations? Why the 50ps period? Why do they sometimes disappear? What about the decay?



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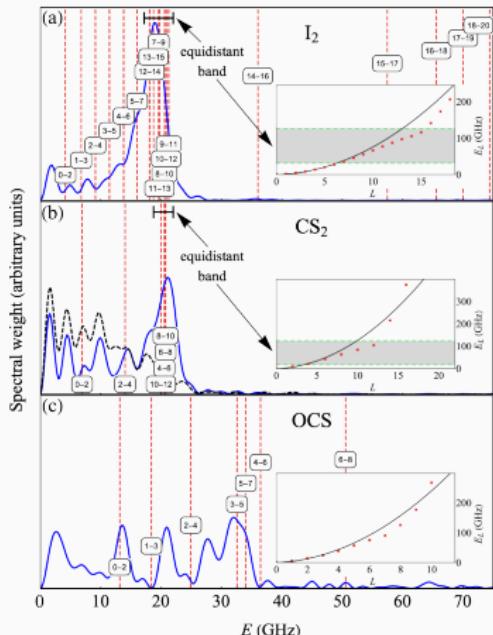
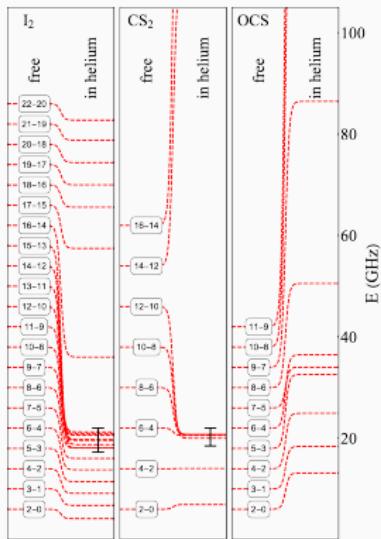


- A microscopical theory allows us to reconstruct the pathways of angular momentum redistribution: **microscopical insight** on the problem!
  - We can fully characterize the helium excitations dressing by the molecule.
  - At the same we can also analyze how molecular properties (populations, energy levels) are affected by the many-body environment.

# Experiments vs. theory: spectrum

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

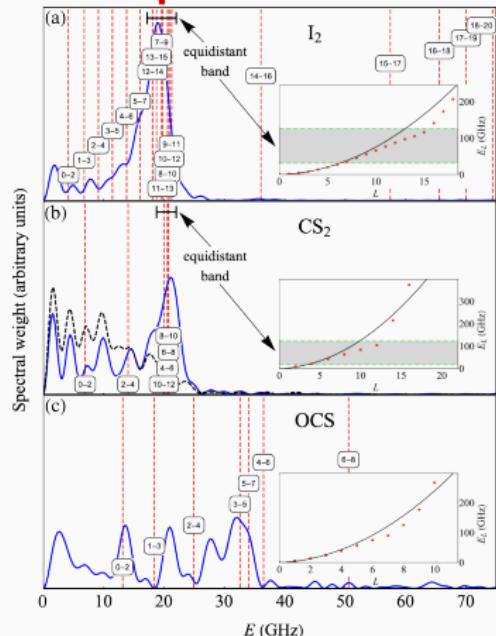
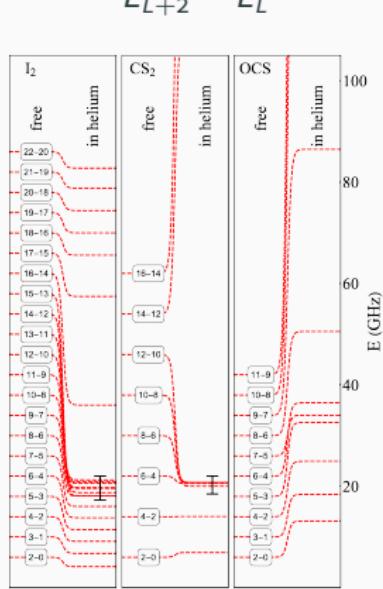
$$E_{L+2} - E_L$$



# Experiments vs. theory: spectrum

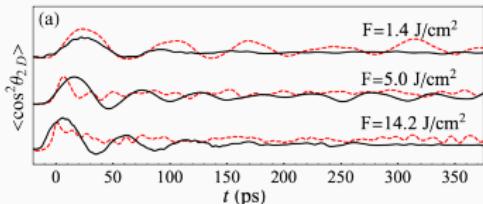
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20Ghz corresponds to 50ps



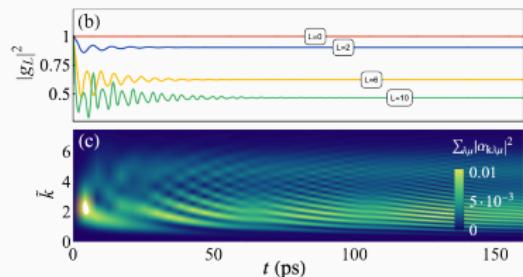
# Many-body dynamics of angular momentum

i) Is this the full story? Can the observed dynamics be explained **only by means of renormalised rotational levels?**



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!**

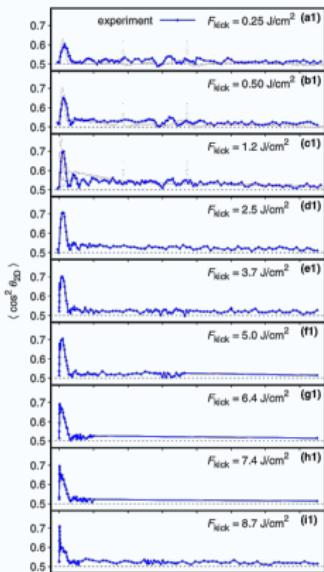


Approach to equilibrium of the quasiparticle weight  $|g_{LM}|^2$  and of the phonon populations  $\sum_k |\alpha_{k\lambda\mu}|^2$ .

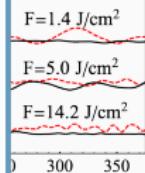
# Many-body dynamics of angular momentum

i) Is this the fundamental dynamics being renormalised

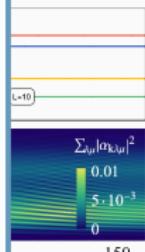
With a shorter 450 fs pulse, same molecule ( $I_2$ ), the strong oscillatory pattern is absent:



ii) How long does it take for the system to equilibrate with the environment and form an angular momentum state? This requires tens of picoseconds, which is a timescale of the order of the cavity lifetime.



Energy levels (in eV) vs. time (in picoseconds) after each treatment.



Biparticle populations

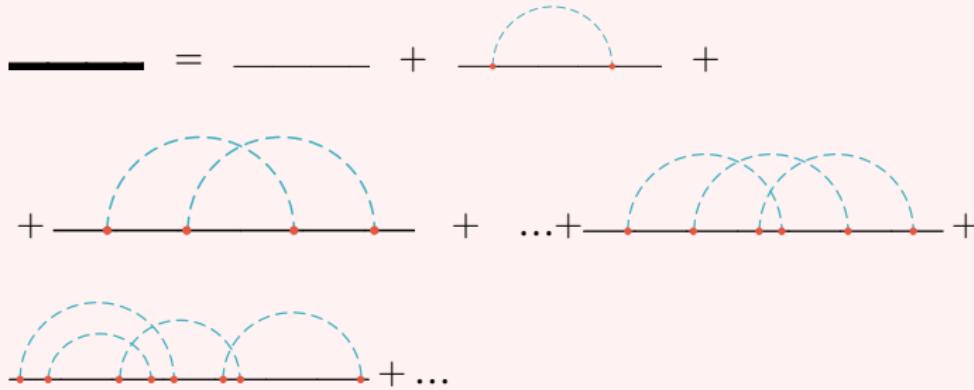
Image from: B. Shepperson *et al.*, Phys. Rev. Lett. **118**, 203203 (2017).

## 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; an explanation requires a **many-body theory** of angular momentum redistribution.
- 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 femtosecond timescales**.
- Future perspectives:
  - All molecular geometries (spherical tops, asymmetric tops).
  - Optical centrifuges and superrotors.
  - Can a rotating molecule create a vortex?
- For more details: arXiv:1906.12238

# Diagrammatic Monte Carlo

More numerical approach: **DiagMC**, sampling all diagrams in a stochastic way.



How do we describe angular momentum redistribution in terms of diagrams?  
How does the configuration space looks like?

Can we use DiagMC to describe a molecule?

# Lemeshko group @ IST Austria:



*Institute of Science and Technology*



Misha  
Lemeshko

Dynamics in He



Enderalp  
Yakaboylu



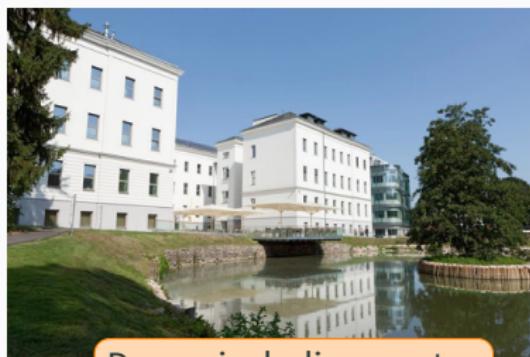
Xiang Li



Igor  
Cherepanov



Wojciech  
Rządkowski



Dynamical alignment  
experiments

## Collaborators:



Henrik  
Stapelfeldt  
(Aarhus)



Richard  
Schmidt  
(MPQ)

DiagMC



Timur  
Tscherbul  
(Reno) 22/23

# Thank you for your attention.



Der Wissenschaftsfonds.

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These slides at <http://bigh.in>

## Backup slide # 1: finite-temperature dynamics

For the **impurity**: average over a statistical ensemble, weights  $\propto \exp(-\beta E_L)$ .

For the **bath**: the zero-temperature bosonic expectation values in  $\mathcal{L}$  are converted to finite temperature ones<sup>1,2</sup>.

$$\mathcal{L}_{T=0} = \langle 0 | \hat{O}^\dagger (i\partial_t - \hat{\mathcal{H}}) \hat{O} | 0 \rangle_{bos} \longrightarrow \mathcal{L}_T = \text{Tr} \left[ \rho_0 \hat{O}^\dagger (i\partial_t - \hat{\mathcal{H}}) \hat{O} \right]$$

[1] A. R. DeAngelis and G. Gantoff, Phys. Rev. C 43, 2747 (1991).

[2] W.E. Liu, J. Levinsen, M. M. Parish, "Variational approach for impurity dynamics at finite temperature", arXiv:1805.10013

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A couple of additional details:

- The laser changes the total angular momentum of the system. An appropriate **wavefunction** is then  $|\Psi\rangle = \sum_{LM} |\psi_{LM}\rangle$
- **Focal averaging**, accounting for the fact that the laser is not always perfectly focused.
- States with odd/even angular momenta may have **different abundances**, due to the nuclear spin.

[1] A. R. DeAngelis and G. Gaitoff, Phys. Rev. C 43, 2747 (1991).

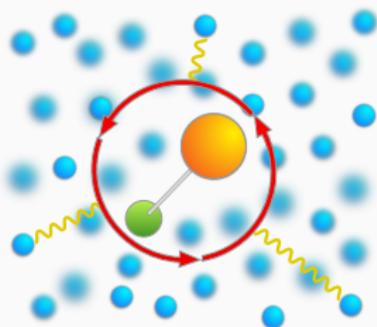
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## Backup slide # 2: the angulon

A composite impurity in a bosonic environment can be described by the angulon Hamiltonian<sup>1,2,3,4</sup> (angular momentum basis:  $\mathbf{k} \rightarrow \{k, \lambda, \mu\}$ ):

$$\hat{H} = \underbrace{B\hat{\mathbf{j}}^2}_{\text{molecule}} + \underbrace{\sum_{k\lambda\mu} \omega_k \hat{b}_{k\lambda\mu}^\dagger \hat{b}_{k\lambda\mu}}_{\text{phonons}} + \underbrace{\sum_{k\lambda\mu} U_\lambda(k) \left[ Y_{\lambda\mu}^*(\hat{\theta}, \hat{\phi}) \hat{b}_{k\lambda\mu}^\dagger + Y_{\lambda\mu}(\hat{\theta}, \hat{\phi}) \hat{b}_{k\lambda\mu} \right]}_{\text{molecule-phonon interaction}}$$

- Linear molecule.
- Derived rigorously for a molecule in a weakly-interacting BEC<sup>1</sup>.
- Phenomenological model for a molecule in any kind of bosonic bath<sup>3</sup>.



<sup>1</sup>R. Schmidt and M. Lemeshko, Phys. Rev. Lett. **114**, 203001 (2015).

<sup>2</sup>R. Schmidt and M. Lemeshko, Phys. Rev. X **6**, 011012 (2016).

<sup>3</sup>M. Lemeshko, Phys. Rev. Lett. **118**, 095301 (2017).

<sup>4</sup>Yu. Shchadilova, "Viewpoint: A New Angle on Quantum Impurities", Physics **10**, 20 (2017).

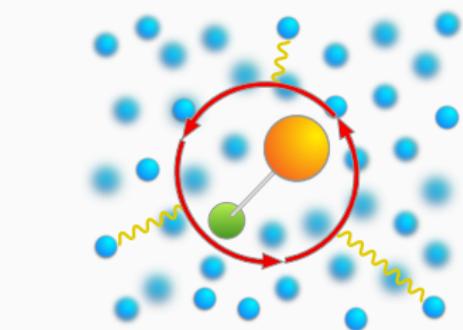
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$\lambda = 0$ : spherically symmetric part.

- $\lambda \geq 1$  anisotropic part.
- A molecule in a weakly-interacting BEC<sup>1</sup>.
- Phenomenological model for a molecule in any kind of bosonic bath<sup>3</sup>.



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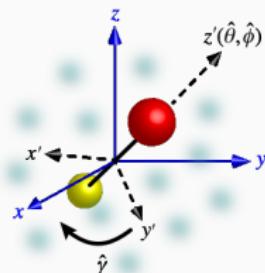
## Backup slide # 3: canonical transformation

We apply a canonical transformation

$$\hat{S} = e^{-i\hat{\phi}\otimes\hat{\Lambda}_z} e^{-i\hat{\theta}\otimes\hat{\Lambda}_y} e^{-i\hat{\gamma}\otimes\hat{\Lambda}_z}$$

where  $\hat{\Lambda} = \sum_{\mu\nu} b_{k\lambda\mu}^\dagger \vec{\sigma}_{\mu\nu} b_{k\lambda\nu}$  is the angular momentum of the bosons.

Cfr. the Lee-Low-Pines transformation for the polaron.

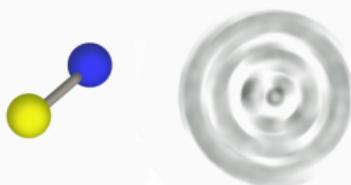


Bosons: laboratory frame ( $x, y, z$ )  
Molecule: rotating frame ( $x', y', z'$ )  
defined by the Euler angles  $(\hat{\phi}, \hat{\theta}, \hat{\gamma})$ .



laboratory frame

$$\hat{S} \rightarrow$$



rotating frame