Far-from-equilibrium dynamics of molecules in ^{4}He nanodroplets: a quasiparticle perspective

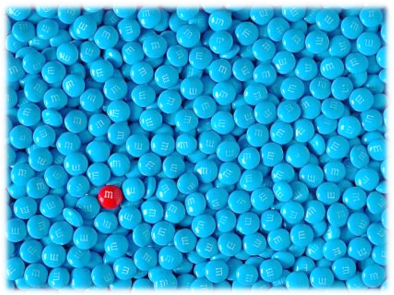
Giacomo Bighin
Institute of Science and Technology Austria

Universität Heidelberg, May 23th, 2019

Quantum impurities

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

- Condensed matter
- Chemistry
- Ultracold atoms



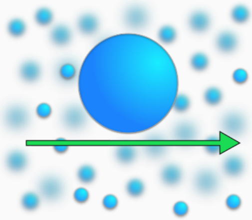
How are the properties of the particle modified by the interaction?

$\mathcal{O}(10^{23})$ degrees of freedom.

Impurities and quasiparticles

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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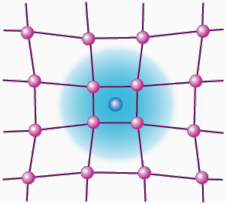

Image from: F. Chevy, Physics **9**, 86.

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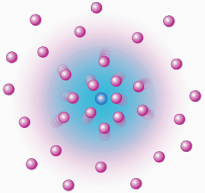

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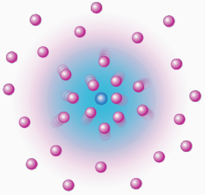

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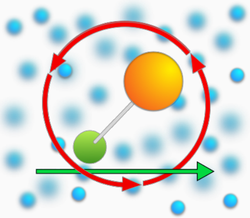

Composite impurity: translational *and internal* (i.e. rotational) degrees of freedom/linear and angular momentum exchange.

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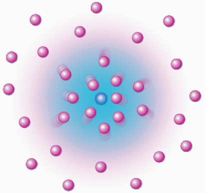

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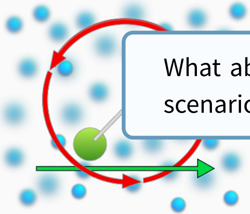


nd internal

near and

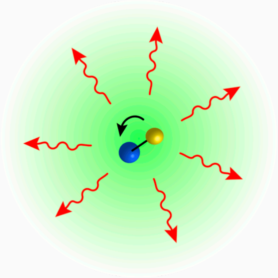
angular momentum exchange.

What about a rotating particle? How can this scenario be realized?

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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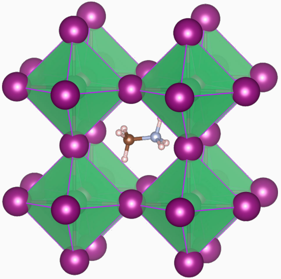

T. Chen et al., PNAS **114**, 7519 (2017).
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Image from: C. Eames et al, Nat. Comm. **6**, 7497 (2015).

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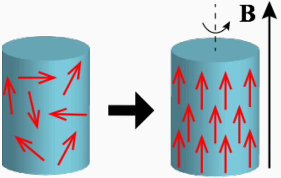

J.H. Mentink, M.I. Katsnelson, M. Lemeshko, *"Quantum many-body dynamics of the Einstein-de Haas effect"*, Phys. Rev. B **99**, 064428 (2019).

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- Ultracold molecules and ions.
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- Angular momentum transfer from the electrons to a crystal lattice.
- Molecules embedded into helium nanodroplets.

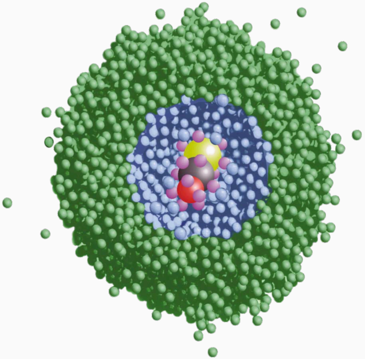


Image from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

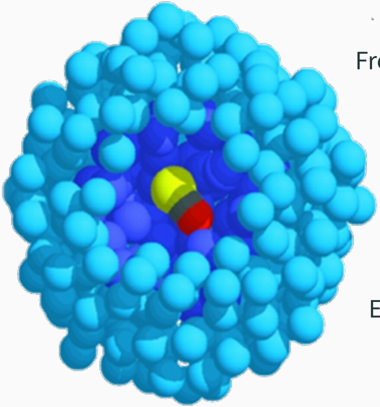
Molecules in helium nanodroplets

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

Temperature $\sim$ 0.4K

Droplets are superfluid

Easy to produce



Free of perturbations

Only rotational degrees of freedom

Easy to manipulate by a laser

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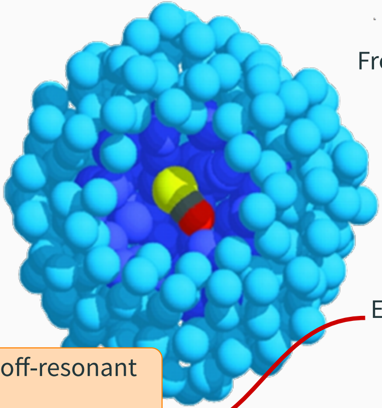

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Easy to manipulate by a laser

Interaction with an off-resonant laser pulse:

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

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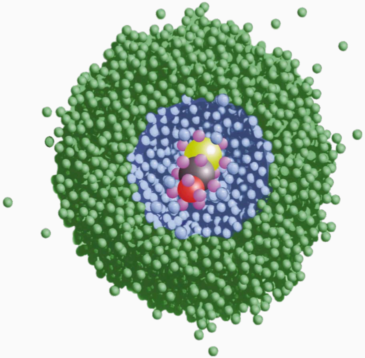
Rotational spectrum of molecules in He nanodroplets

Molecules embedded into helium nanodroplets: rotational spectrum



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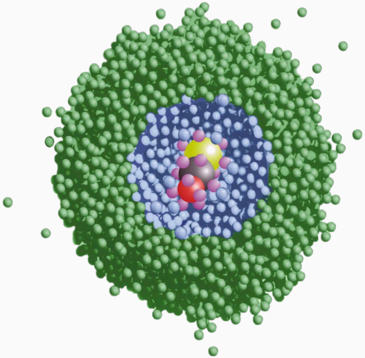

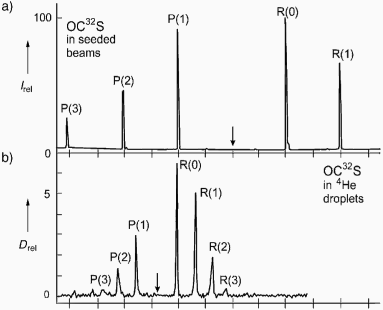

Gas phase (free)

in ^{4}He

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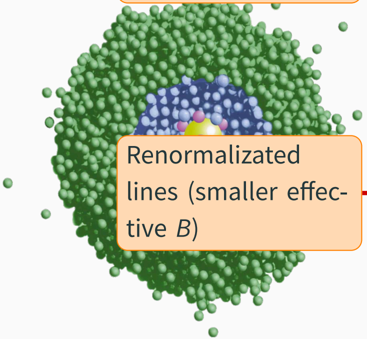
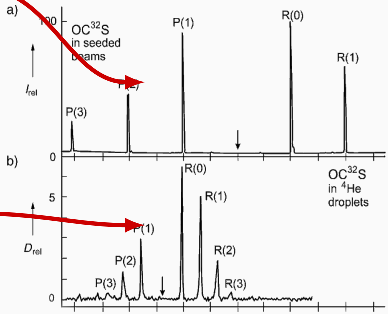
Rotational spectrum



Renormalizated lines (smaller effective B)



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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

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

with:

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

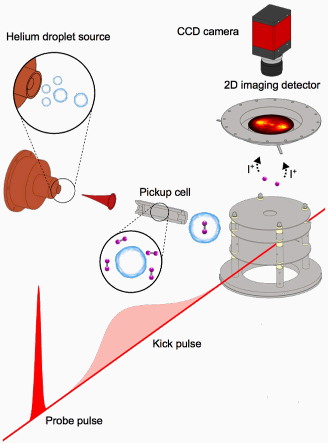


Image from: B. Shepperson et al., Phys. Rev. Lett. **118**, 203203 (2017).

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:

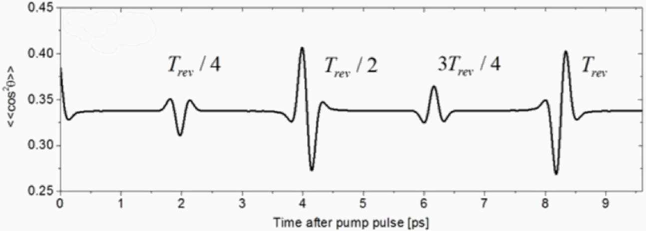
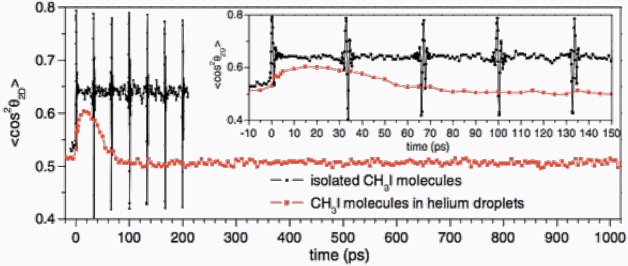


Image from: G. Kaya *et al.*, Appl. Phys. B **6**, 122 (2016).

Movie

Dynamical alignment of molecules in He nanodroplets

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

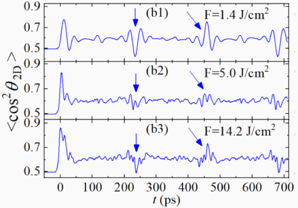


Stapelfeldt group, Phys. Rev. Lett. **110**, 093002 (2013).

Very noticeable differences in the timescales and in the approach to equilibrium.
An intriguing puzzle: not even a qualitative understanding. Monte Carlo? He-DFT?

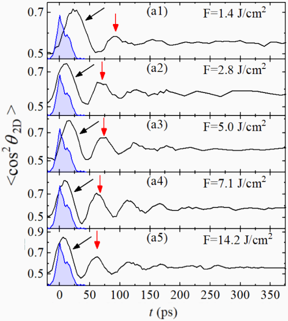
Dynamical alignment of molecules in He nanodroplets

Dynamics of isolated I_2 molecules



Experiment: Henrik Stapelfeldt, Lars Christiansen, Anders Vestergaard Jørgensen (Aarhus University)

Dynamics of I_2 molecules in helium

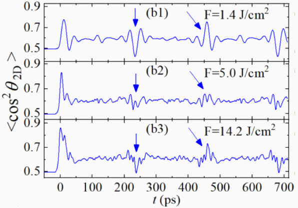


Striking differences between the two cases:

- 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.

Dynamical alignment of molecules in He nanodroplets

Dynamics of isolated I_2 molecules



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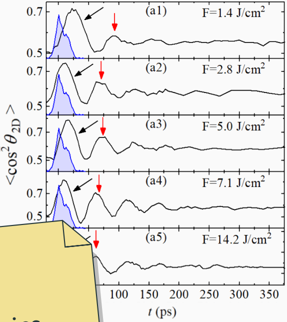

Experiment: Henri
Anders Vestergaa

— Strong coupling

— Out-of-equilibrium dynamics

— Finite temperature ($B \sim k_B T$)

Striking differ

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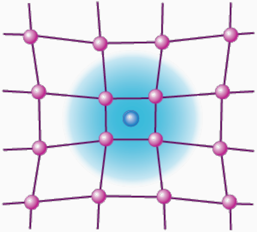
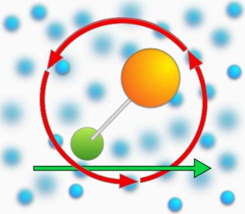
The quantum mechanical treatment of many-body systems is always challenging. How can one simplify the quantum impurity problem?

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

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





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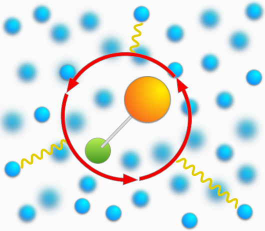
The angulon

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

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

- Linear molecule.
- Derived rigorously for a molecule in a weakly-interacting BEC[1].
- Phenomenological model for a molecule in any kind of bosonic bath[3].



[1]R. Schmidt and M. Lemeshko, Phys. Rev. Lett. **114**, 203001 (2015).
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$\lambda = 0$: spherically symmetric part.
$\lambda \geq 1$ anisotropic part.

- L...
- D... a molecule in a weakly-interacting BEC[1].
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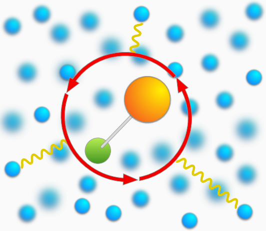

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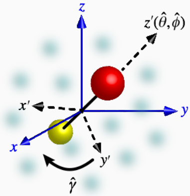
We apply a canonical transformation

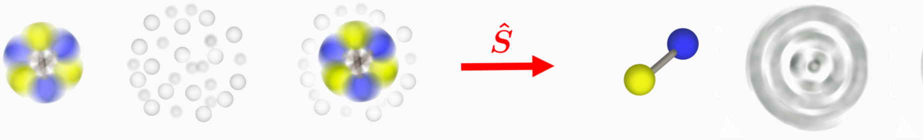
$$\hat{S} = e^{-i\hat{\phi}\otimes\hat{\Lambda}_z} e^{-i\hat{\theta}\otimes\hat{\Lambda}_y} e^{-i\hat{\gamma}\otimes\hat{\Lambda}_z}$$

where $\hat{\mathbf{\Lambda}} = \sum_{\mu\nu} b^{\dagger}_{k\lambda\mu}\vec{\sigma}_{\mu\nu}b_{k\lambda\nu}$ 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 **rotating frame**

Canonical transformation

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

$$\hat{\mathcal{H}} = \hat{S}^{-1}\hat{H}\hat{S} = B(\hat{\mathbf{L}} - \hat{\boldsymbol{\Lambda}})^2 + \sum_{k\lambda\mu} \omega_k \hat{b}^\dagger_{k\lambda\mu}\hat{b}_{k\lambda\mu} + \sum_{k\lambda} V_{k\lambda}\left(\hat{b}^\dagger_{k\lambda 0} + \hat{b}_{k\lambda 0}\right),$$

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Compare with the Lee-Low-Pines Hamiltonian

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

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- Macroscopic deformation of the bath, exciting an infinite number of bosons.
- Simplifies angular momentum algebra.
- Hamiltonian diagonalizable through a coherent state transformation $\hat{U}$ in the $B \to 0$ limit. An expansion in bath excitations is a strong coupling expansion.

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✓ Strong coupling

— Out-of-equilibrium dynamics

— Finite temperature ($B \sim k_B T$)

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Dynamics: time-dependent variational Ansatz

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

$$|\psi_{LM}(t)\rangle = \hat{U}(g_{LM}(t)\,|0\rangle_{\text{bos}}\,|LM0\rangle + \sum_{k\lambda n} \alpha^{LM}_{k\lambda n}(t) b^{\dagger}_{k\lambda n}\,|0\rangle_{\text{bos}}\,|LMn\rangle)$$

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

$$\mathcal{L}_{T=0} = \langle \psi_{LM}|\mathrm{i}\partial_t - \hat{\mathcal{H}}|\psi_{LM}\rangle$$

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 $x_i = \{g_{LM}, \alpha^{LM}_{k\lambda n}\}$. We obtain a differential system

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

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

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Euler-Lagran

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— Finite temperature ($B \sim k_B T$)

where $x_i = \{g$ ... em

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

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

Finite-temperature dynamics

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

For the bath: the zero-temperature bosonic expectation values in $\mathcal{L}$ are converted to finite temperature ones[1,2].

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

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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. Gatoff, 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 ad...

- The laser cha... the system. An appropriate wavefunctio...
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✓ Strong coupling

✓ Out-of-equilibrium dynamics

✓ Finite temperature ($B \sim k_B T$)

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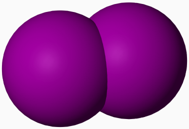
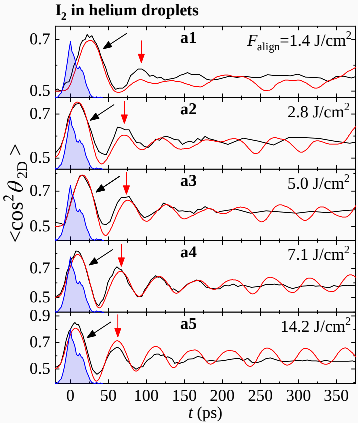
Theory vs. experiments: I_2

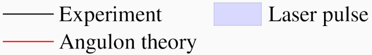


Comparison with experimental data from Stapelfeldt group, Aarhus University, for different molecules: I_2.

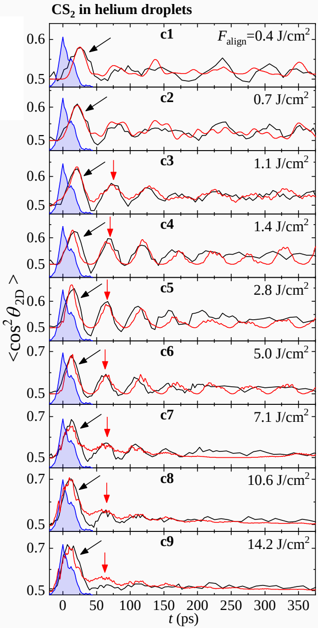


Generally good agreement for the main features in experimental data:

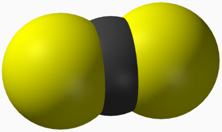
- Oscillations with a period of 50ps, growing in amplitude as the laser fluence is increased.
- Oscillations decay: at most 4 periods are visible.
- The width of the first peak does not change much with fluence.



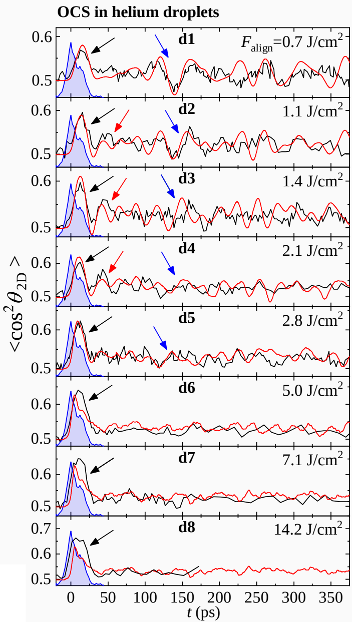
Theory vs. experiments: CS_2



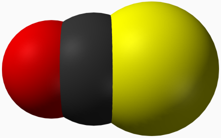
Comparison with experimental data from Stapelfeldt group, Aarhus University, for different molecules: CS_2.



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



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?

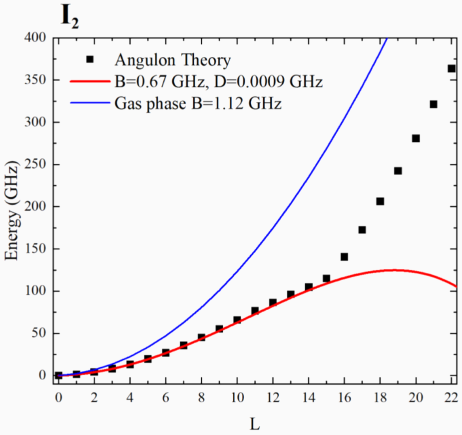
- Can we shed light on the origin of oscillations? Why the 50ps period? Why do they sometimes disappear? What about the decay?

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

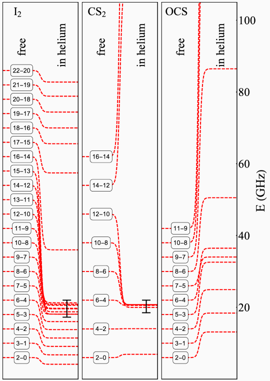
- Free molecule: $E_L = BL(L+1)$
- Molecule in helium: $E_L = B^* L(L+1) - D[L(L+1)]^2$

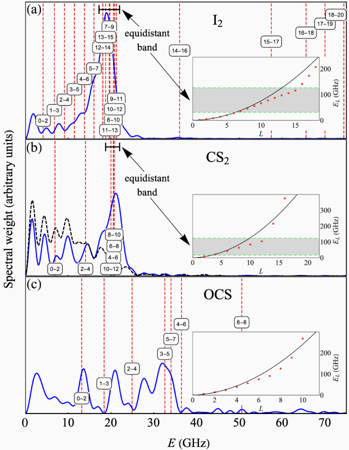


- For small values of L the rotational constant is renormalized $B \to B^*$.
- For intermediate values of L the centrifugal correction $D[L(L+1)]^2$ becomes relevant.
- For large L's one recovers a quadratic spectrum: detachment.

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?

$E_{L+2} - E_L$



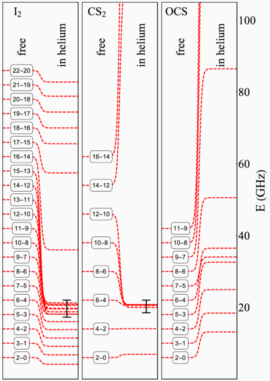


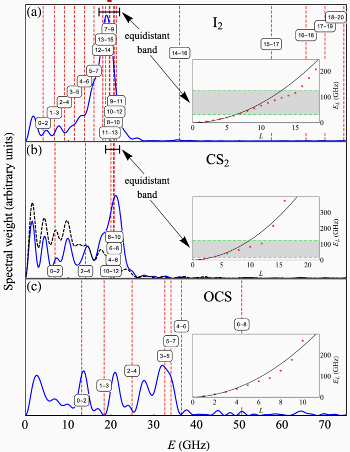
Experiments vs. theory: spectrum

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?

20Ghz corresponds to 50ps

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





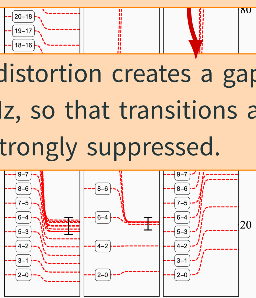
Experiments vs. theory: spectrum

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 str

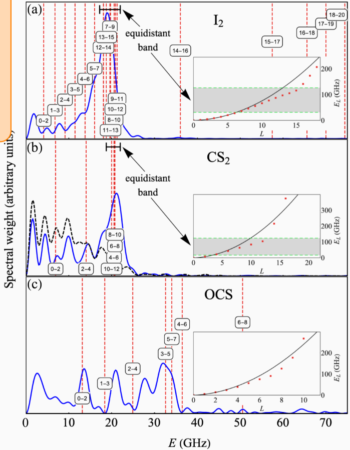
Transition probability under a Gaussian pulse

$$W_{fi} = \frac{|V_{fi}|^2}{\hbar^2} \exp\left(-\sigma^2 \omega_{fi}^2\right)$$

where $\omega_{fi} \equiv (E_f - E_i)/\hbar$ and σ is the pulse duration.

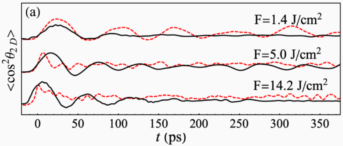


The distortion creates a gap after 20GHz, so that transitions after the gap are strongly suppressed.



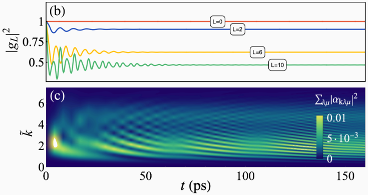
Many-body dynamics of angular momentum

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).

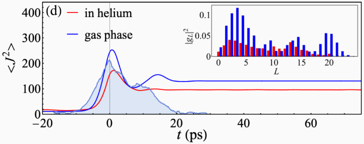
ii) How long does it take for a molecule to equilibrate with the helium environment and form an angulon quasiparticle? This requires tens of ps; which is also the timescale of the laser!



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

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



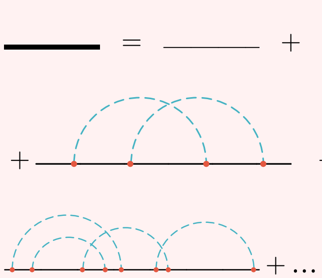
Time evolution of the molecular angular momentum, in helium (red) and in the gas phase (blue).

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).
  - Can a rotating molecule create a vortex?
- Soon to be on the arXiv!

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?

Connecting DiagMC and the theory of molecular simulations!

GB and M. Lemeshko, Phys. Rev. B **96**, 419 (2017).
GB, T.V. Tscherbul, M. Lemeshko, Phys. Rev. Lett. **121**, 165301 (2018).

Lemeshko group @ IST Austria:

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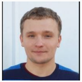
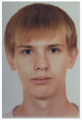


Misha Lemeshko

Dynamics in He

Enderalp Yakaboylu

Xiang Li



Igor Cherepanov

Wojciech Rządkowski

Dynamical alignment experiments

Collaborators:

Henrik Stapelfeldt (Aarhus)

Richard Schmidt (MPI Garching)

Thank you for your attention.

Der Wissenschaftsfonds.

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These slides at http://bigh.in/talks

Backup slide # 1

Backup slide # 2

Backup slide # 3