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

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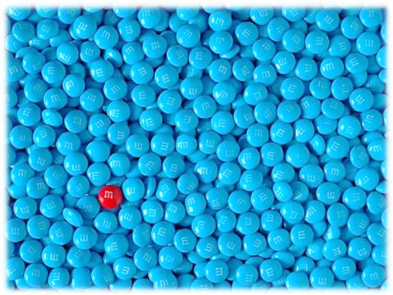
Universitat Politècnica de Catalunya — Barcelona, September 18th, 2019

# Quantum impurities

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

- Condensed matter
- Chemistry
- Ultracold atoms

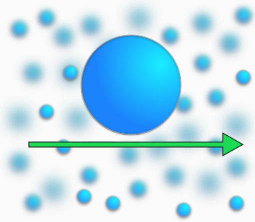


How are the properties of the particle modified by the interaction?

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

# Quantum impurities

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



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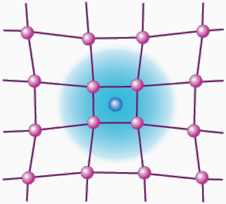

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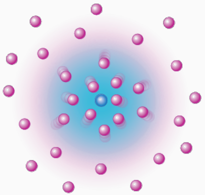

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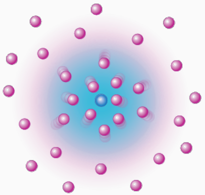

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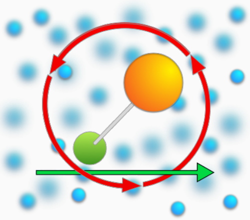

**Composite impurity (e.g. a molecule):** translational *and rotational* degrees of freedom/linear and angular momentum exchange.

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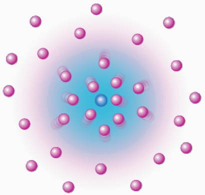

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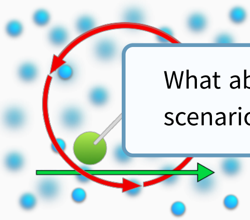

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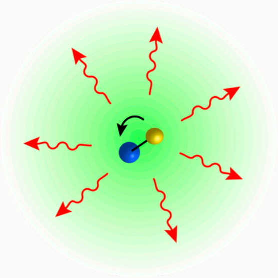
exchange.

What about a rotating impurity? How can this scenario be realized experimentally?

# 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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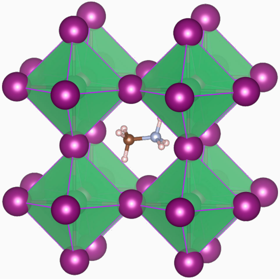

T. Chen et al., PNAS **114**, 7519 (2017).
J. Lahnsteiner et al., Phys. Rev. B **94**, 214114 (2016).
Image from: C. Eames et al, Nat. Comm. **6**, 7497 (2015).

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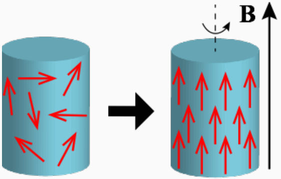


J.H. Mentink, M.I. Katsnelson, M. Lemeshko, *"Quantum many-body dynamics of the Einstein-de Haas effect"*, Phys. Rev. B **99**, 064428 (2019).

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- Rotating molecules inside a 'cage' in perovskites.
- Angular momentum transfer from the electrons to a crystal lattice.
- Molecules embedded into helium nanodroplets.

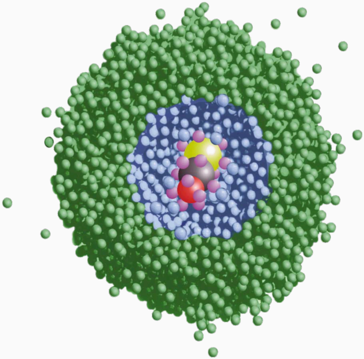


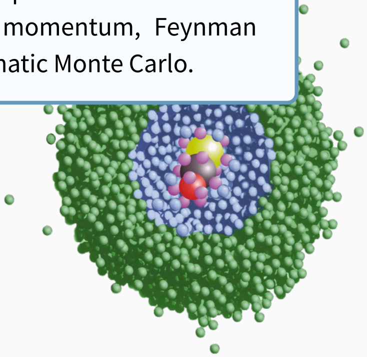
Image from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

# Composite impurities: where to find them

Strong motiv… many different fiel…

**First part:** out-of-equilibrium dynamics of molecules in He nanodroplets.
**Second part:** angular momentum, Feynman diagrams and Diagrammatic Monte Carlo.

- Ultracol…
- Rotating … 'cage' in perovskites.
- Angular momentum transfer from the electrons to a crystal lattice.
- Molecules embedded into helium nanodroplets.



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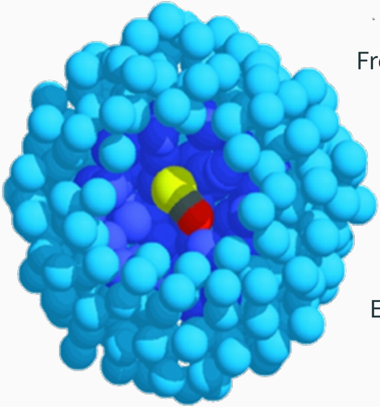
# Molecules in helium nanodroplets

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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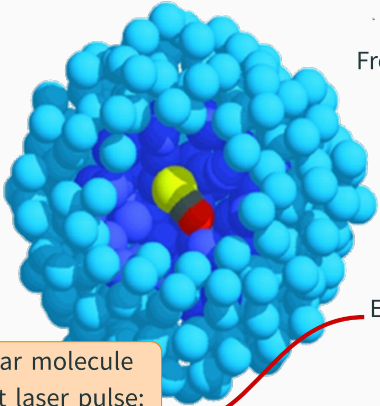

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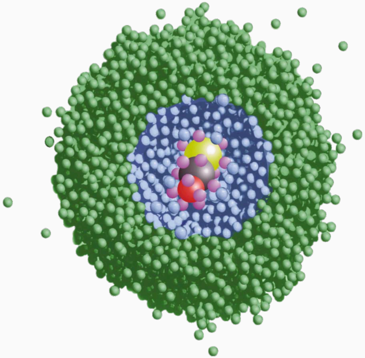
Interaction of a linear molecule with an off-resonant laser pulse:

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

# Rotational spectrum of molecules in He nanodroplets

Molecules embedded into helium nanodroplets: rotational spectrum



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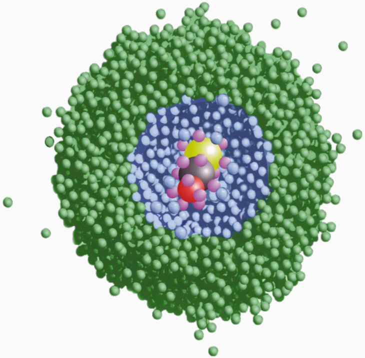

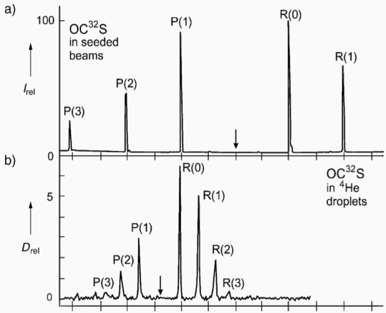


Gas phase (free)

in $^4$He

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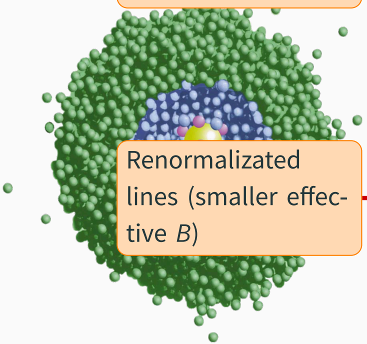
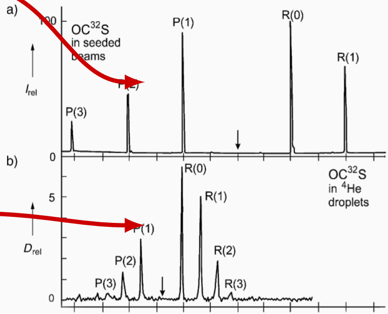
Molecules embedded into helium nanodroplets: rotational spectrum

Rotational spectrum



Renormalizated lines (smaller effective *B*)




Gas phase (free)

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**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:

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

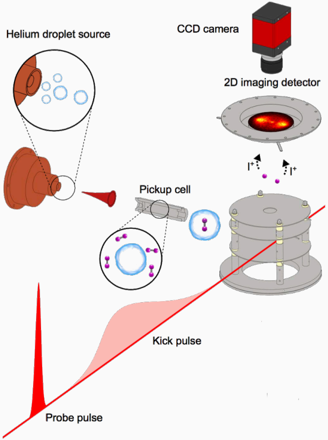



Image from: B. Shepperson *et al.*, Phys. Rev. Lett. **118**, 203203 (2017).

# 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:

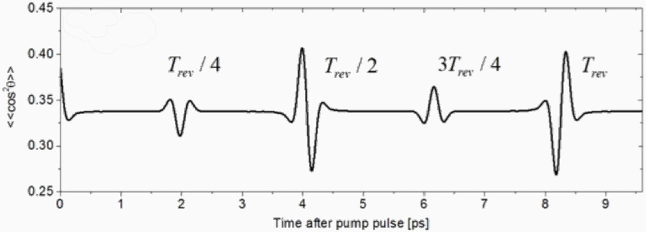
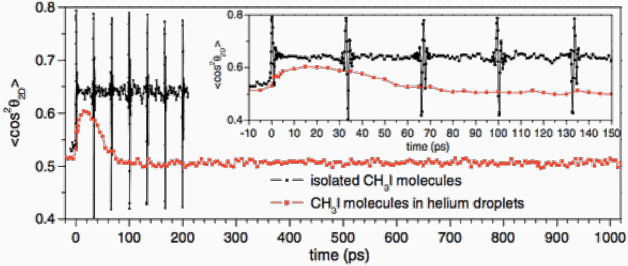



Image from: G. Kaya *et al.*, Appl. Phys. B **6**, 122 (2016).

Movie

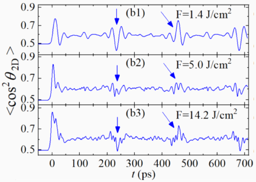
Effect of the environment is substantial: **free molecule** vs. **same molecule in He**.




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

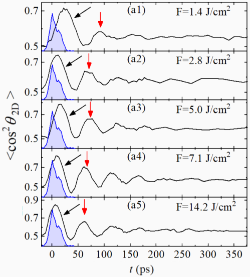
# Dynamical alignment of molecules in He nanodroplets

Dynamics of isolated $I_2$ molecules




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

Dynamics of $I_2$ 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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**Polaron**: an electron dressed by a field of many-body excitations.

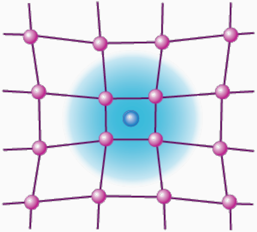
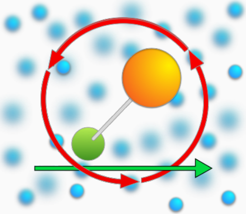


Image from: F. Chevy, Physics **9**, 86.

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

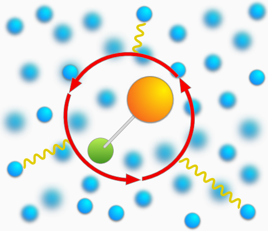


# The angulon

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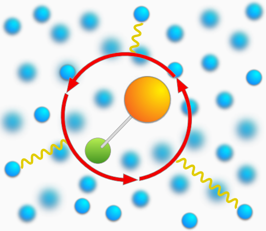

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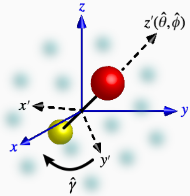
We apply a canonical transformation

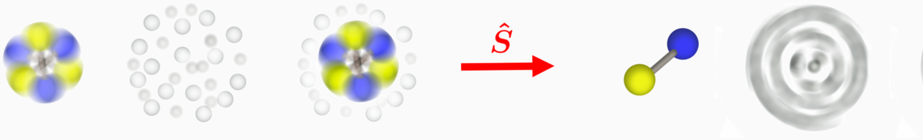
$$\hat{S} = e^{-i\hat{\phi}\otimes\hat{\Lambda}_z} e^{-i\hat{\theta}\otimes\hat{\Lambda}_y} e^{-i\hat{\gamma}\otimes\hat{\Lambda}_z}$$

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.

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

# 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:

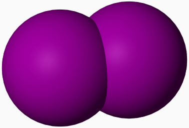
$$\frac{d}{dt}\frac{\partial\mathcal{L}}{\partial\dot{x}_i} - \frac{\partial\mathcal{L}}{\partial x_i} = 0$$

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

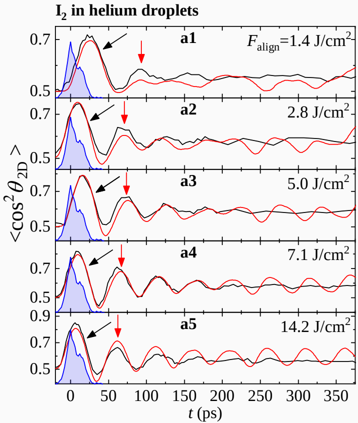
$$\begin{cases} \dot{g}_{LM}(t) = \dots \\ \dot{\alpha}^{LM}_{k\lambda n}(t) = \dots \end{cases}$$

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

## Theory vs. experiments: $I_2$



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

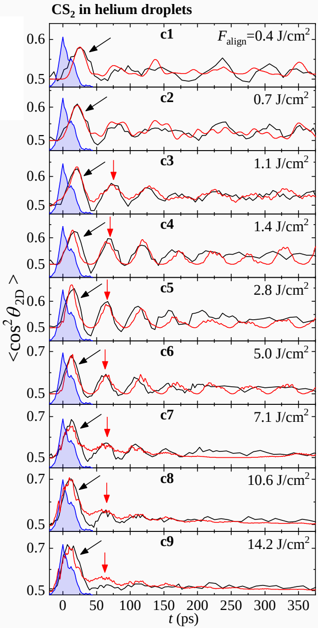



Generally good agreement for the main features in experimental data:

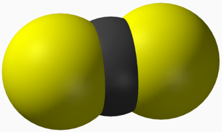
- Oscillations with a period of 50ps, growing in amplitude as the laser fluence is increased.
- Oscillations decay: at most 4 periods are visible.
- The width of the first peak does not change much with fluence.

Experiment — Laser pulse

Angulon theory

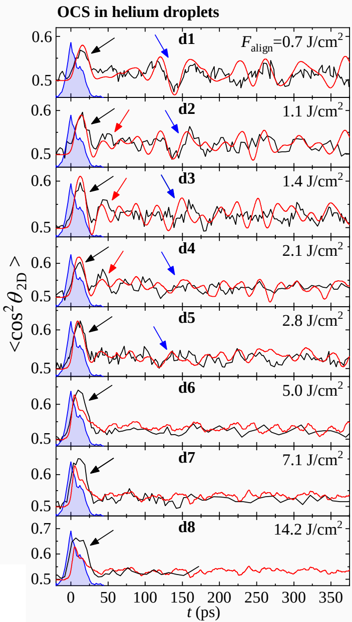



Comparison with experimental data from Stapelfeldt group, Aarhus University, for different molecules: $CS_2$.

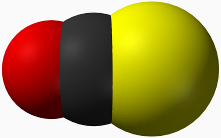


- Again, a persistent oscillatory pattern.
- For higher values of the fluence the oscillatory pattern disappears.

# 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.

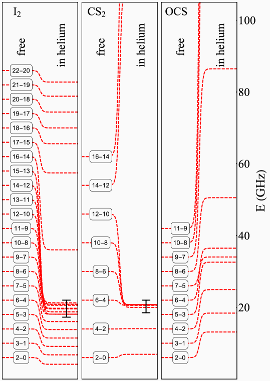
- Can we shed light on the origin of oscillations? Why the 50ps period? Why do they sometimes disappear? What about the decay?

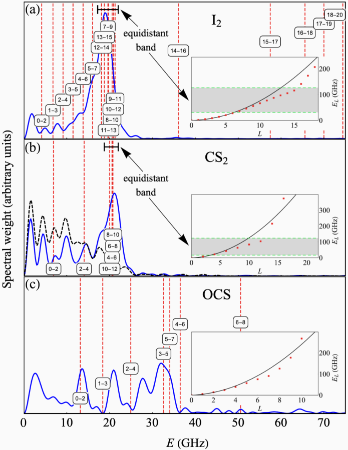
- Can we shed light on the origin of oscillations? Why the 50ps period? Why do they sometimes disappear? What about the decay?

- 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.

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?

$E_{L+2} - E_L$



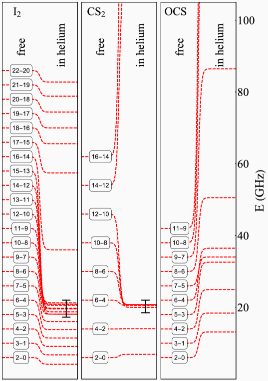



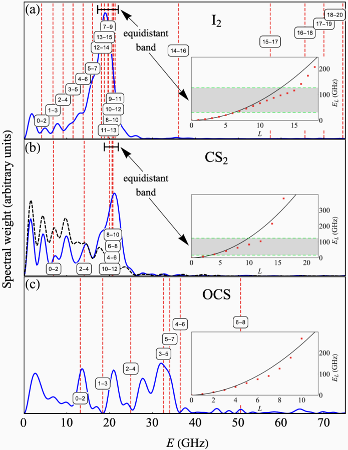
# Experiments vs. theory: spectrum

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

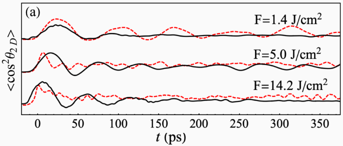
$E_{L+2} - E_L$



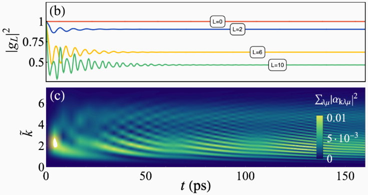
# Many-body dynamics of angular momentum

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 $\sum_k |\alpha_{k\lambda\mu}|^2$.

i) Is this the fu
dynamics be
renormalised

ii) How long c
equilibrate wi
and form an a
requires tens
timescale of t

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

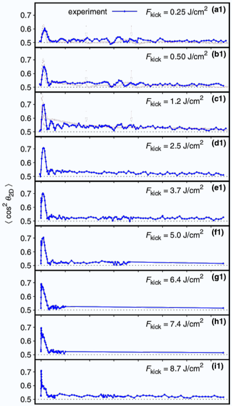
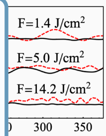



Image from: B. Shepperson *et al.*, Phys. Rev. Lett. **118**, 203203 (2017).




ed levels) vs.
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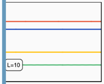


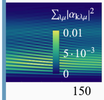


siparticle
opulations

# Summary of the first part

- 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

# Angular momentum and Feynman diagrams

# Perturbative approach and Feynman diagrams

Back to the angulon Hamiltonian:

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

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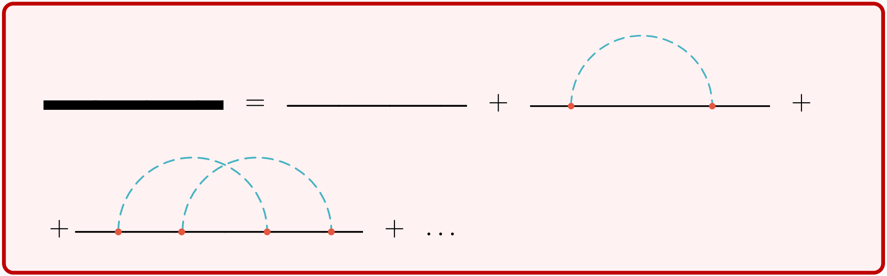
Perturbation theory/Feynman diagrams:



How does **angular momentum** enter this picture?

# Perturbative approach and Feynman diagrams

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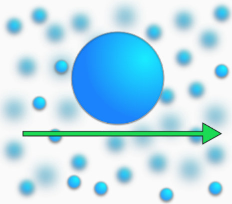
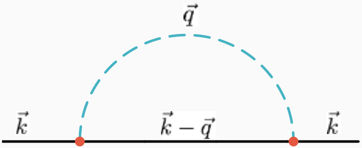
Perturbation theory/Feynman diagrams:

**Fröhlich polaron**





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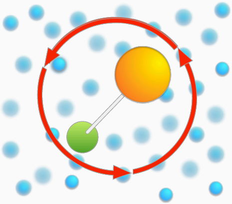
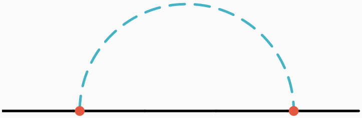
**Angulon**





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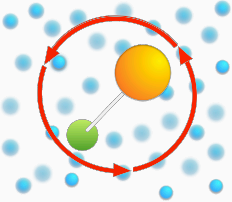
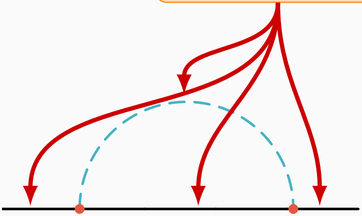
Perturbation theory/Feynman diagrams:

How does **angular momentum** enter here?

**Angulon**





## Feynman rules

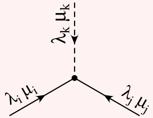
Each free propagator


$$\sum_{\lambda_i \mu_i} (-1)^{\mu_i} G_{0,\lambda_i}$$

Each phonon propagator


$$\sum_{\lambda_i \mu_i} (-1)^{\mu_i} D_{\lambda_i}$$

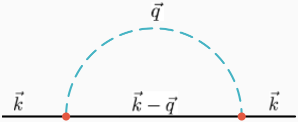
Each vertex




$$(-1)^{\lambda_i} \langle \lambda_i | | Y^{(\lambda_j)} | | \lambda_k \rangle \begin{pmatrix} \lambda_i & \lambda_j & \lambda_k \\ \mu_i & \mu_j & \mu_k \end{pmatrix}$$

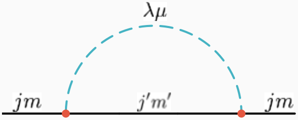
GB and M. Lemeshko, Phys. Rev. B **96**, 419 (2017).

Usually momentum conservation is enforced by an appropriate labeling.




Not the same for angular momentum, $j$ and $\lambda$ couple to $|j-\lambda|, \ldots, j+\lambda$.

$\sum_{j'm'}$



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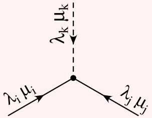
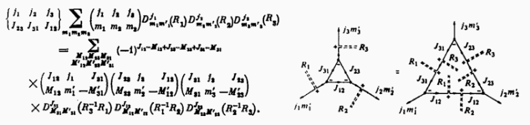



$$(-1)^{\lambda_i} \langle \lambda_i || Y^{(\lambda_j)} || \lambda_k \rangle \begin{pmatrix} \lambda_i & \lambda_j & \lambda_k \\ \mu_i & \mu_j & \mu_k \end{pmatrix}$$

GB and M. Lemeshko, Phys. Rev. B **96**, 419 (2017).

Diagrammatic theory of angular momentum (developed in the context of theoretical atomic spectroscopy)

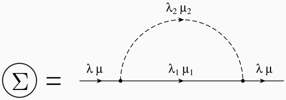



from D. A. Varshalovich, A. N. Moskalev, V. K. Khersonskii, *"Quantum Theory of Angular Momentum"*.

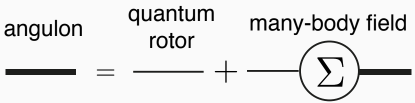
# Angulon spectral function

Let us use the Feynman diagrams!

First order self-energy:




Dyson equation :




Finally the spectral function allows for a study the whole excitation spectrum of the system:

$$\mathcal{A}_\lambda(E) = -\frac{1}{\pi} \operatorname{Im} G_\lambda(E + \mathrm{i}0^+)$$

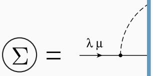
Equivalent to a simple, 1-phonon variational Ansatz (cf. Chevy Ansatz for the polaron)

$$|\psi\rangle = Z_{LM}^{1/2} |0\rangle \; |LM\rangle + \sum_{\substack{k\lambda\mu \\ jm}} \beta_{k\lambda j} C_{jm,\lambda\mu}^{LM} b_{k\lambda\mu}^\dagger |0\rangle \; |jm\rangle$$

# Angulon spectral function

Let us use th

First order sel                                                                 :




field

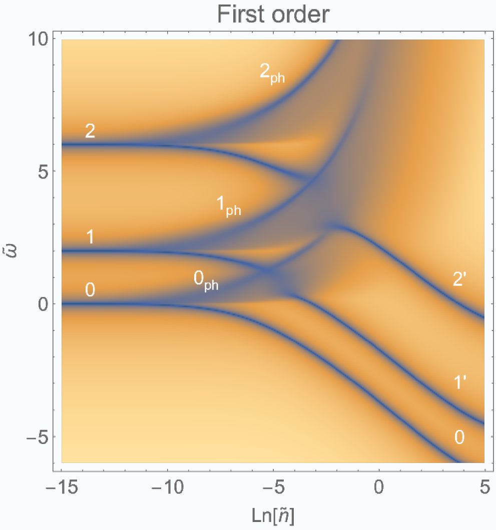
Finally the sp                                                                  ctrum of

the system:

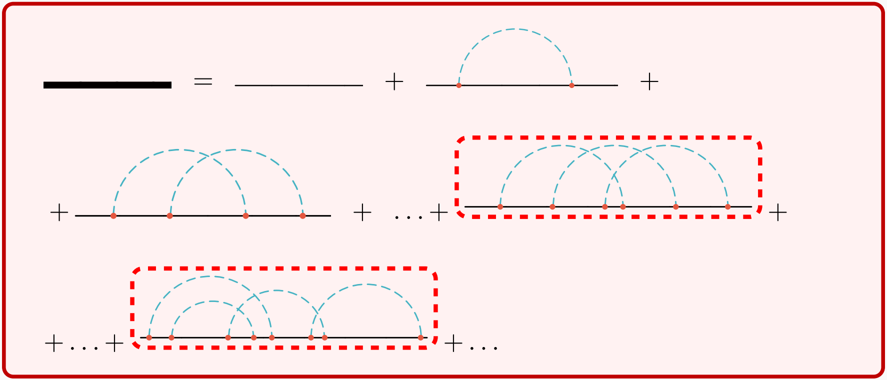
Equivalent to                                                                   r the

polaron)

Spectral function: $\mathcal{A}_\lambda(E)$

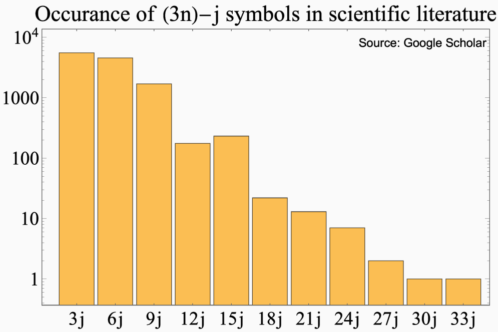


What about higher orders?



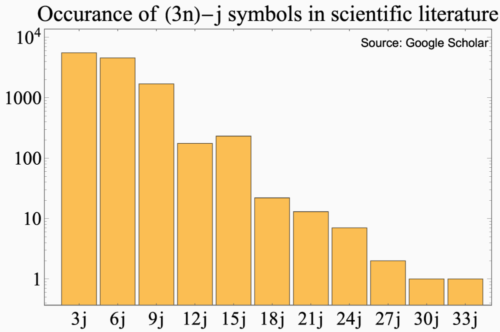
At order *n*: *n* integrals, and higher angular momentum couplings (3*n*-j symbols).

# A feasible plan?




Notice the logarithmic scale: exponentially rare, since they are exponentially more difficult to compute.

A feasible plan?

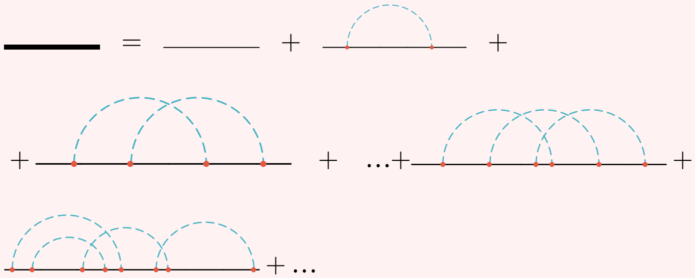



Notice the logarithmic scale: exponentially rare, since they are exponentially more difficult to compute.

For monster stuff, like a 303-j symbol taking 2.3 years to compute, see: C. Brouder and G. Brinkmann, Journal of Electron Spectroscopy and Related Phenomena **86**, 127 (1997).

# Diagrammatic Monte Carlo

Numerical technique for summing all Feynman diagrams[1]. More on this later...



Up to now: structureless particles (Fröhlich polaron, Holstein polaron), or particles with a very simple internal structure (e.g. spin 1/2).

Molecules[2]? Connecting DiagMC and molecular simulations!

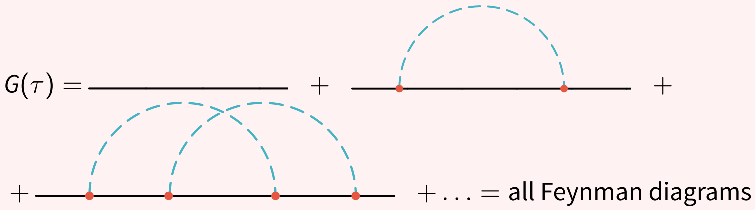
[1] N. V. Prokof'ev and B. V. Svistunov, Phys. Rev. Lett. **81**, 2514 (1998).

[2] GB, T.V. Tscherbul, M. Lemeshko, Phys. Rev. Lett. **121**, 165301 (2018).

# Diagrammatic Monte Carlo

Hamiltonian for an impurity problem: $\hat{H} = \hat{H}_{\text{imp}} + \hat{H}_{\text{bath}} + \hat{H}_{\text{int}}$

## Green's function




**DiagMC idea:** set up a stochastic process sampling among all diagrams[1].

**Configuration space:** diagram topology, phonons internal variables, times, etc... Number of variables varies with the topology!

**How:** ergodicity, detailed balance $w_1 p(1 \to 2) = w_2 p(2 \to 1)$

**Result:** each configuration is visited with probability $\propto$ its weight.

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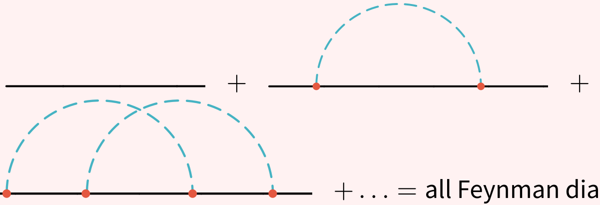
**Green's function**

$G(\tau) =$



$+ \ldots =$ all Feynman diagrams

**DiagMC idea:** set up a stochastic process sampling among all diagrams[1].

> Works in continuous time and in the thermodynamic limit: no finite-size effects or systematic errors.

**Configuratio**… , times, etc... Numbe…

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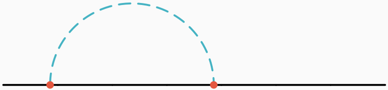

**Add** update: a new arc is added to a diagram.

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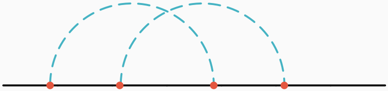

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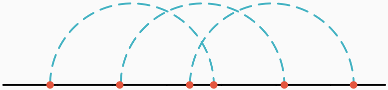

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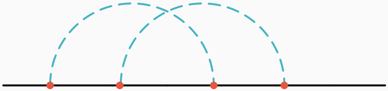

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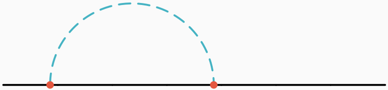

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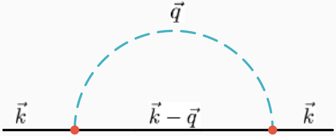
**Remove** update: an arc is removed from the diagram.
**Change** update: modifies the total length of the diagram.

**Result:** the time the stochastic process spends with diagrams of length $\tau$ will be proportional to $G(\tau)$. One can fill a histogram after each update and get the Green's function.

# Diagrammatics for a rotating impurity

Moving particle: linear momentum circulating on lines.

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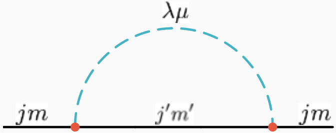

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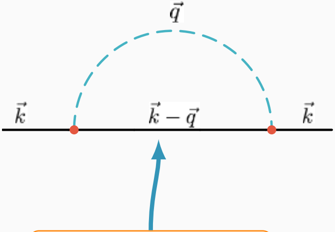
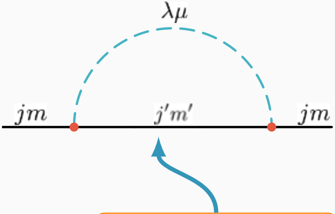


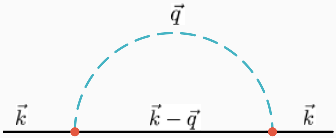

$\vec{k}$ and $\vec{q}$ fully determine $\vec{k}-\vec{q}$

Rotating particle: angular momentum circulating on lines.

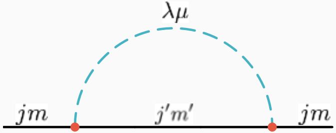



$j$ and $\lambda$ can sum in many different ways: $|j-\lambda|, \ldots j+\lambda$

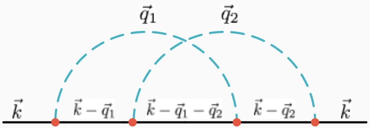
Moving particle: linear momentum circulating on lines.

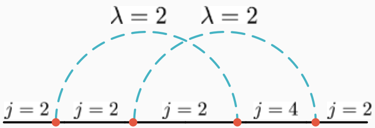



Rotating particle: angular momentum circulating on lines.



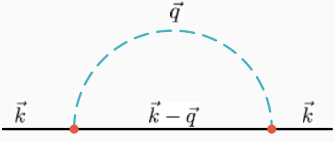

It gets weirder... Down the rabbit hole of angular momentum composition!



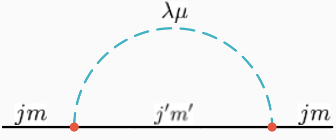



# Diagrammatics for a rotating impurity

Moving particle: linear momentum circulating on lines.




Rotating particle: angular momentum circulating on lines.

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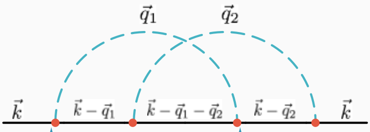
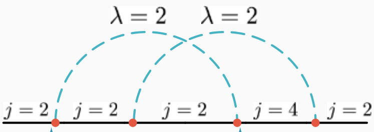



The phonon takes away $\vec{q}_1$ momentum...

...and gives back $\vec{q}_1$ momentum



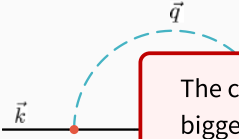

The phonon does not subtract angular momentum from the impurity...

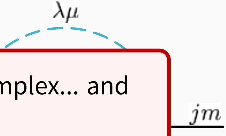
...but gives back two quanta!

# Diagrammatics for a rotating impurity

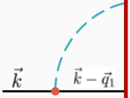
Moving particle: linear momentum circulating on lines.

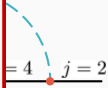
Rotating particle: angular momentum circulating on lines.



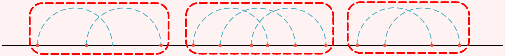




It gets weirde … osition!




The configuration space is more complex... and bigger! We need different updates.



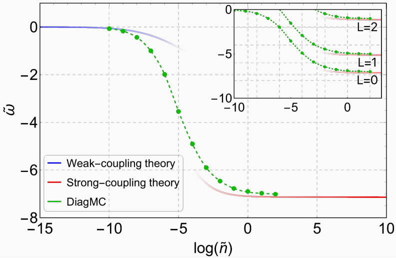
**Shuffle** update: select one 1-particle-irreducible component, shuffle the momenta couplings to another allowed configuration.

# DiagMC: results

The ground-state energy of the angulon Hamiltonian obtained using DiagMC[1] as a function of the dimensionless bath density, $\tilde{n}$, in comparison with the weak-coupling theory[2] and the strong-coupling theory[3].

The energy is obtained by fitting the long-imaginary-time behaviour of $G_j$ with $G_j(\tau) = Z_j \exp(-E_j\,\tau)$.

Inset: energy of the $L = 0, 1, 2$ states.




[1] GB, T.V. Tscherbul, M. Lemeshko, Phys. Rev. Lett. **121**, 165301 (2018).
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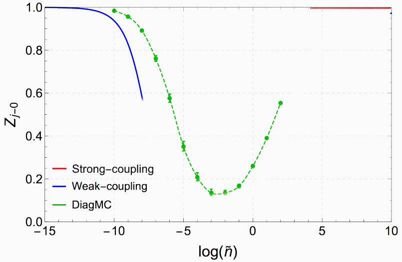


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# Conclusions

- A numerically-exact approach to quantum many-body systems involving coupled angular momenta.
- Works in continuous time and in the thermodynamic limit: no finite-size effects or systematic errors.
- Future perspectives:
  - More advanced schemes (e.g. $\Sigma$, bold).
  - Hybridisation of translational and rotational motion.
  - Real-time dynamics?
- More details: GB, T.V. Tscherbul, M. Lemeshko, Phys. Rev. Lett. **121**, 165301 (2018).

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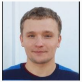
*Institute of Science and Technology*

## Lemeshko group @ IST Austria:

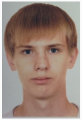


Misha Lemeshko

Dynamics in He

Enderalp Yakaboylu

Xiang Li



Igor Cherepanov

Wojciech Rządkowski

## Collaborators:

DiagMC

Henrik Stapelfeldt (Aarhus)

Richard Schmidt (MPI Garching)

Timur Tscherbul (Reno)

Dynamical alignment experiments

# Thank you for your attention.

Der Wissenschaftsfonds.

This work was supported by a Lise Meitner Fellowship of the Austrian Science Fund (FWF), project Nr. M2461-N27.

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

# Backup slide # 1: 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].

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

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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.
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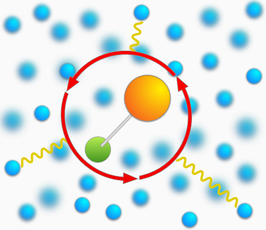
# Backup slide # 2: the angulon

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

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

- 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].



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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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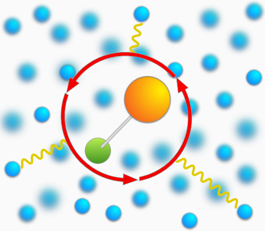

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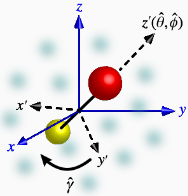
# Backup slide # 3: canonical transformation

We apply a canonical transformation

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

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**

# Finite-temperature dynamics

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

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✓ Strong coupling

✓ Out-of-equilibrium dynamics

✓ Finite temperature ($B \sim k_B T$)


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Some additional considerations:

- $|\Psi\rangle = \sum_{LM} |\psi_{LM}\rangle$
- Averages of the laser intensitiy.
- States with odd/even angular momenta may have different relative abundances, due to the nuclear spin.