Composite, rotating impurities interacting with a many-body environment: analytical and numerical approaches

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Institute of Science and Technology Austria

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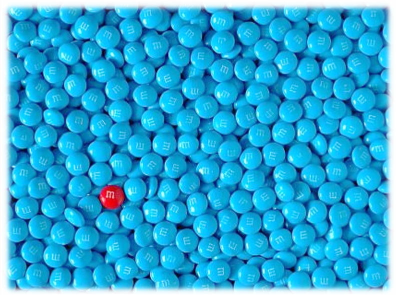
Quantum impurities

Definition: one (or a few particles) interacting with a many-body environment.

- Condensed matter
- Chemistry
- Ultracold atoms

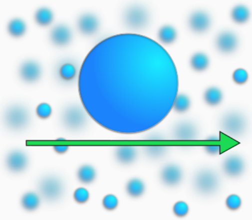


How are the properties of the particle modified by the interaction?

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

From impurities to 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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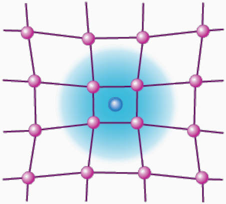

Image from: F. Chevy, Physics **9**, 86.

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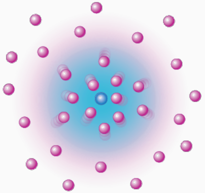

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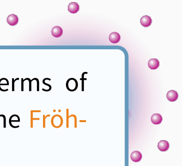
From impurities to quasiparticles

Structureless impurity: translational degrees of f exchange w

Most comm atomic impurities in a BEC.

This scenario can be formalized in terms of quasiparticles using the polaron and the Fröhlich Hamiltonian.



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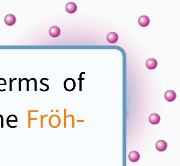

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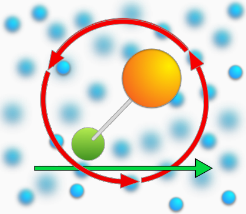

Composite impurity: translational *and internal* (i.e. rotational) degrees of freedom/linear and angular momentum exchange.

From impurities to quasiparticles

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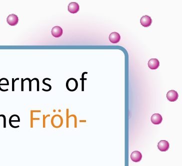

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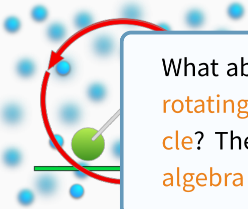


nd *internal*
near and

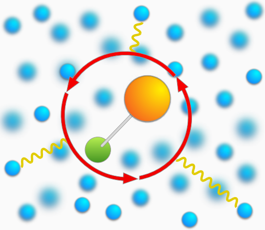
What about a rotating particle? Can there be a rotating counterpart of the polaron quasiparticle? The main difficulty: the non-Abelian SO(3) algebra describing rotations.

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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[4] Y. 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.

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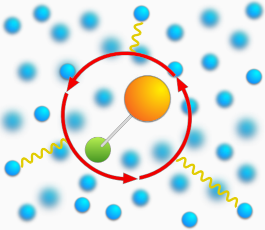

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Composite impurities: where to find them

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

- Molecules embedded into helium nanodroplets.

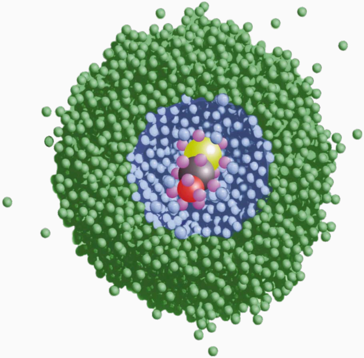

Image from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

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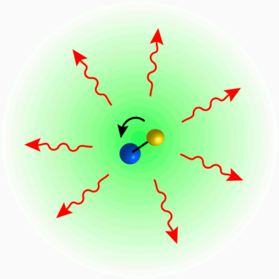
- Ultracold molecules and ions.



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

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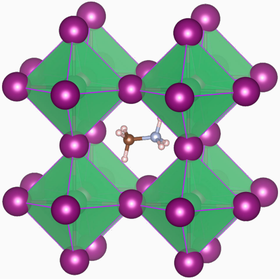

T. Chen et al., PNAS **114**, 7519 (2017).
J. Lahnsteiner et al., Phys. Rev. B **94**, 214114 (2016).
Image from: C. Eames et al, Nat. Comm. **6**, 7497 (2015).

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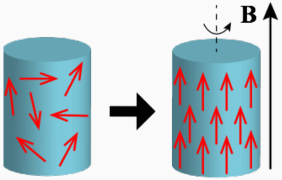

J.H. Mentink, M.I. Katsnelson, M. Lemeshko, *"Quantum many-body dynamics of the Einstein-de Haas effect"*, arXiv:1802.01638

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First part: angular momentum and Feynman diagrams.
Second part: out-of-equilibrium dynamics of molecules in He nanodroplets.

Angular momentum and Feynman diagrams

Perturbative approach and Feynman diagrams

Back to the angulon Hamiltonian:

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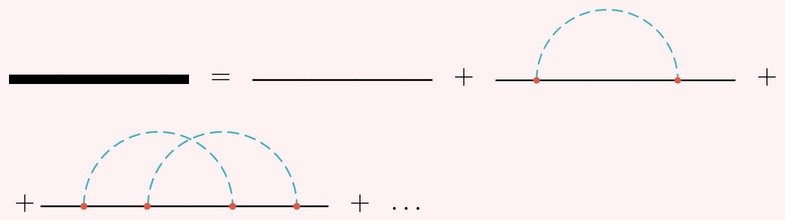
Perturbation theory/Feynman diagrams:



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

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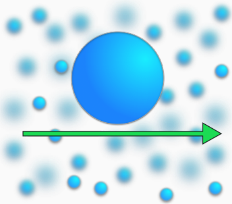
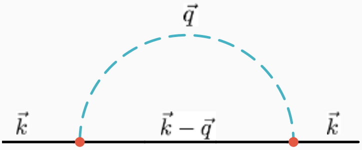
Perturbation theory/Feynman diagrams:

Fröhlich polaron





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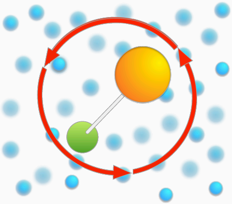
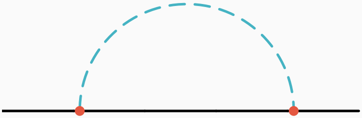
Perturbation theory/Feynman diagrams:

Angulon





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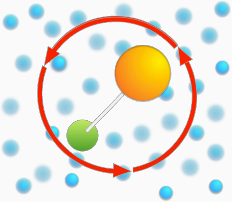
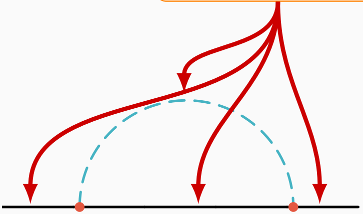


How does **angular momentum** enter here?



Feynman rules

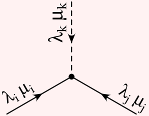
Each free propagator

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

Each phonon propagator

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

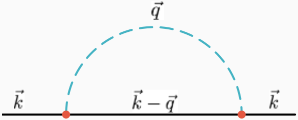
Each vertex



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

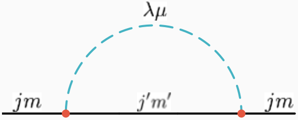
GB and M. Lemeshko, Phys. Rev. B **96**, 419 (2017).

Usually momentum conservation is enforced by an appropriate labeling.



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

$\sum_{j'm'}$



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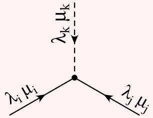

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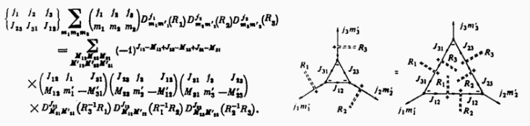
Diagrammatic theory of angular momentum (developed in the context of theoretical atomic spectroscopy)



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

Angulon spectral function

Let us use the Feynman diagrams! The plan is:

1. Self-energy (Σ)
2. Dyson equation to obtain the angulon Green's function (G)
3. Spectral function ($\mathcal{A}$)

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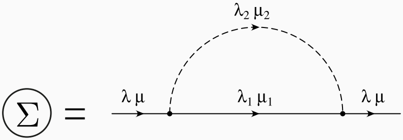

Equivalent to a simple, 1-phonon variational Ansatz (cf. Chevy Ansatz for the polaron)

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

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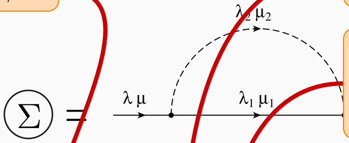
$|\text{bath}\rangle\,|\text{molecule}\rangle$

Quasiparticle weight

Variational coefficients

Clebsch-Gordan to couple angular momenta

First order:



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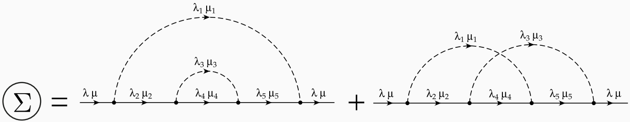

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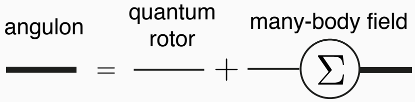
Dyson equation



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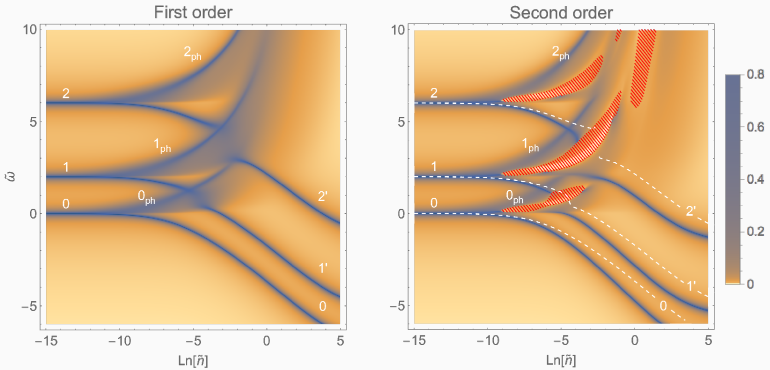
3. Spectral function ($\mathcal{A}$)

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

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

Angulon quasiparticle spectrum as a function of the bath density:



Angulon quasiparticle spectrum

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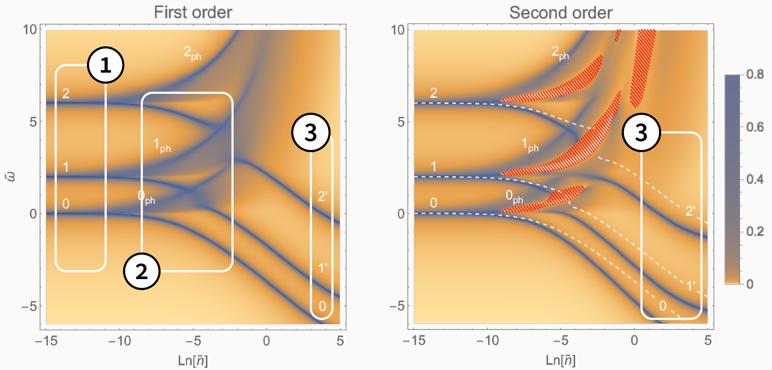
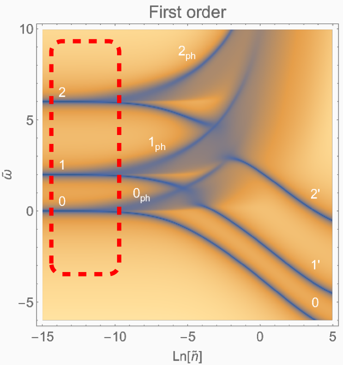


Angulon spectral function: low density

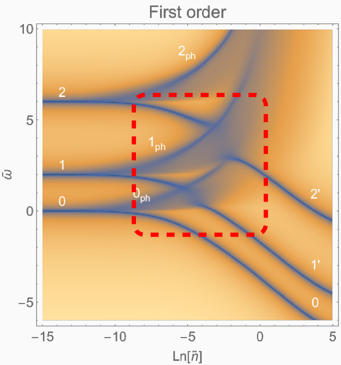


Low density: free rotor spectrum, $E = BL(L+1)$.

Many-body-induced fine structure[1]: upper phonon wing (one phonon with $\lambda = 0$, isotropic interaction).

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

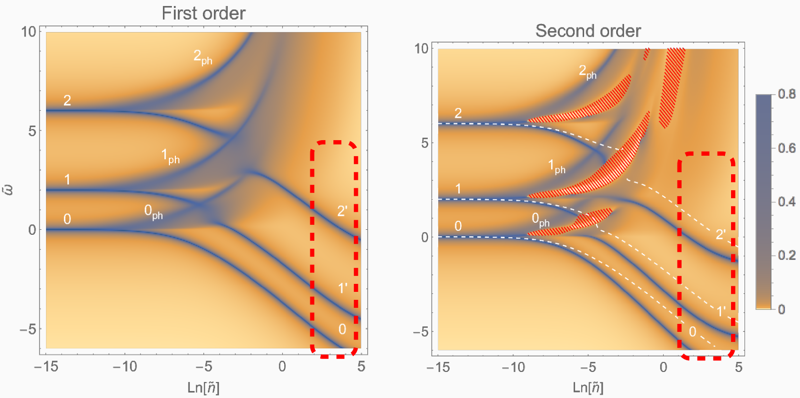
Angulon spectral function: instability



Intermediate region: angulon instability. Many-body resonance, corresponding to the emission of a phonon with $\lambda = 1$ (due to anisotropic interaction).

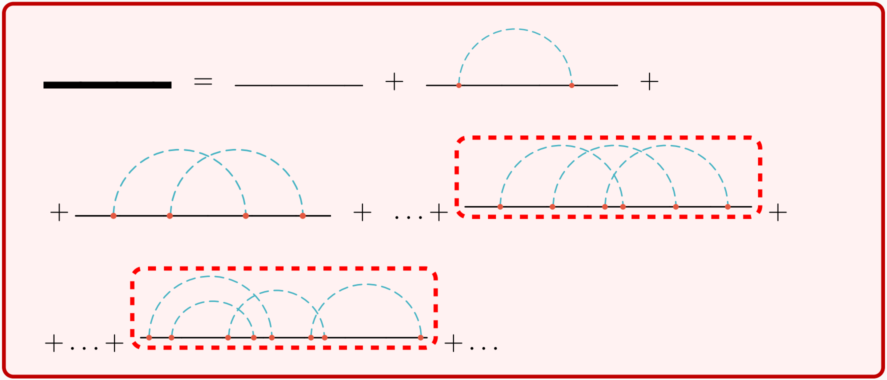
Experimental observation: I. N. Cherepanov, M. Lemeshko, *"Fingerprints of angulon instabilities in the spectra of matrix-isolated molecules"*, Phys. Rev. Materials **1**, 035602 (2017).

Angulon spectral function: high density



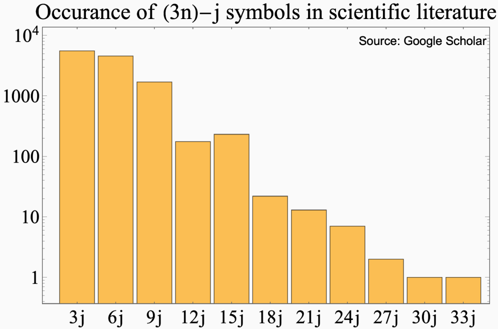
High density: the two-loop corrections start to be relevant.

What about higher orders?



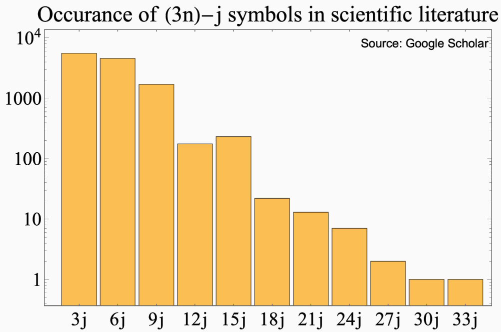
At order n: n integrals, and higher angular momentum couplings ($3n$-j symbols).

A feasible plan?



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

A feasible plan?



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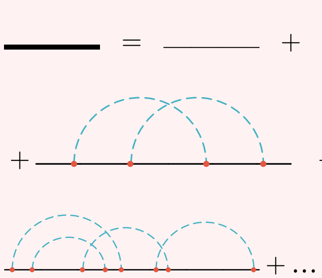
For monster stuff, like a 303-j symbol taking 2.3 years to compute, see: C. Brouder and G. Brinkmann, Journal of Electron Spectroscopy and Related Phenomena **86**, 127 (1997).

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!

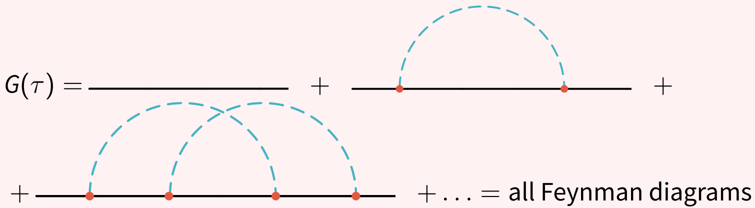
[1] N. V. Prokof'ev and B. V. Svistunov, Phys. Rev. Lett. **81**, 2514 (1998).

[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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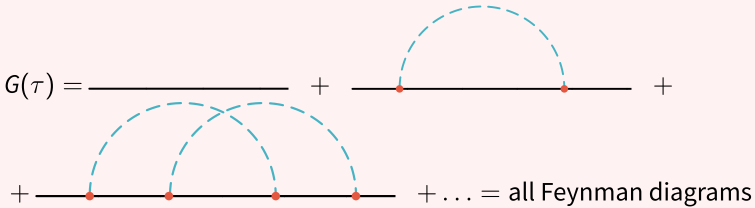

DiagMC idea: … ams[1].

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

Configuratio… , times, etc... Numbe…

How: ergodic…

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

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Updates

We need updates spanning the whole configuration space:

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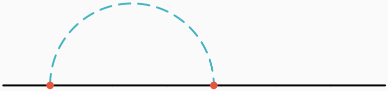

Add update: a new arc is added to a diagram.

Updates

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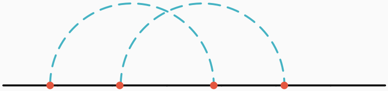

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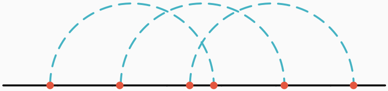

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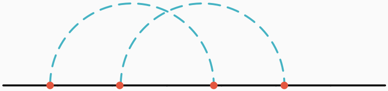

Add update: a new arc is added to a diagram.
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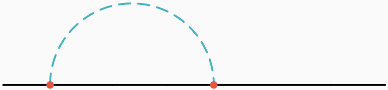

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Add update: a new arc is added to a diagram.
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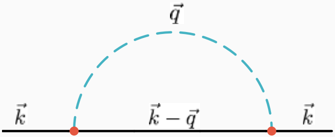
Change update: modifies the total length of the diagram.

Result: the time the stochastic process spends with diagrams of length τ 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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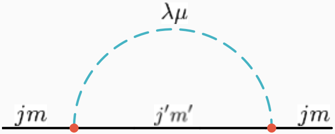

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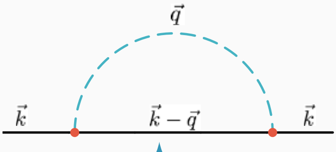
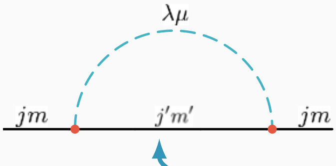


$\vec{k}$ and $\vec{q}$ fully determine $\vec{k}-\vec{q}$

Rotating particle: angular momentum circulating on lines.



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

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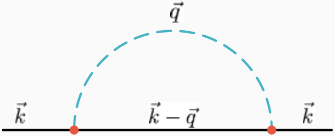

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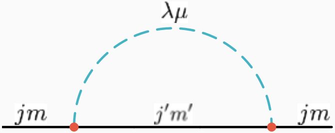
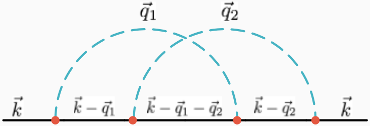
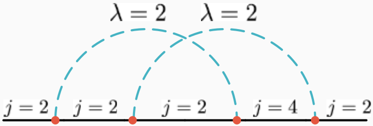


It gets weirder... Down the rabbit hole of angular momentum composition!





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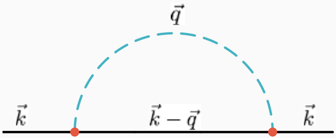

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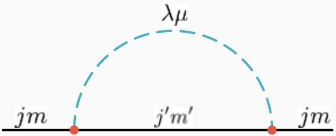

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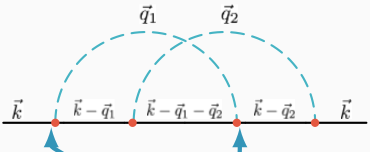
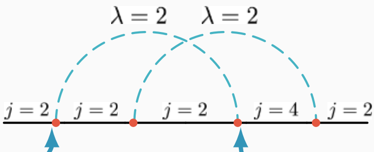




The phonon takes away $\vec{q}_1$ momentum...

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

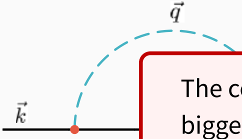
The phonon does not subtract angular momentum from the impurity...

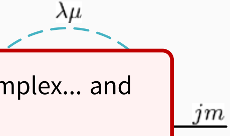
...but gives back two quanta!

Diagrammatics for a rotating impurity

Moving particle: linear momentum circulating on lines.

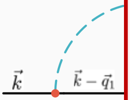
Rotating particle: angular momentum circulating on lines.

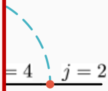




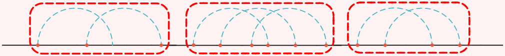
It gets weird

osition!





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



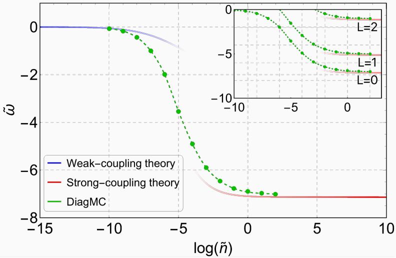
Shuffle update: select one 1-particle-irreducible component, shuffle the momenta couplings to another allowed configuration.

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).
[2] R. Schmidt and M. Lemeshko, Phys. Rev. Lett. **114**, 203001 (2015).
[3] R. Schmidt and M. Lemeshko, Phys. Rev. X **6**, 011012 (2016).

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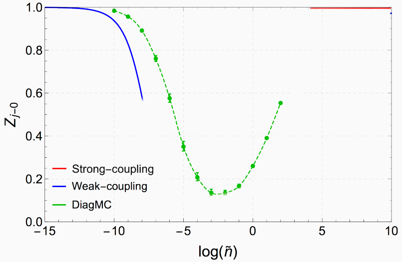

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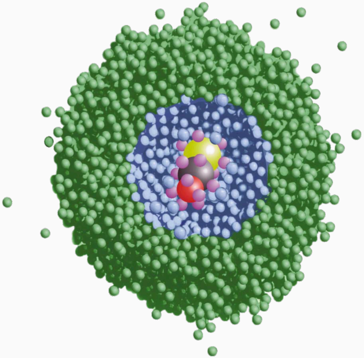
Out-of-equilibrium dynamics of molecules in He nanodroplets

Dynamical alignment of molecules in He nanodroplets

Molecules embedded into helium nanodroplets:



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

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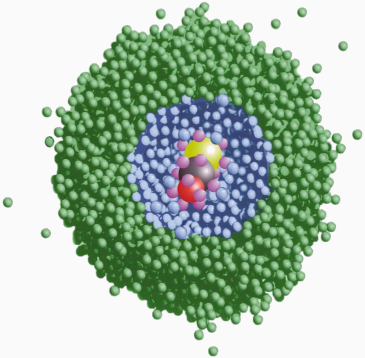

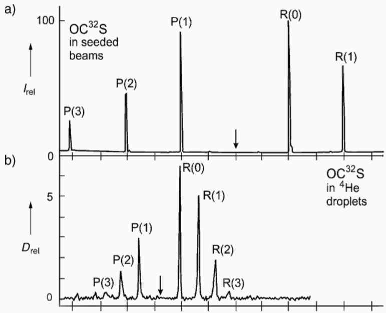

Gas phase (free)

in ^{4}He

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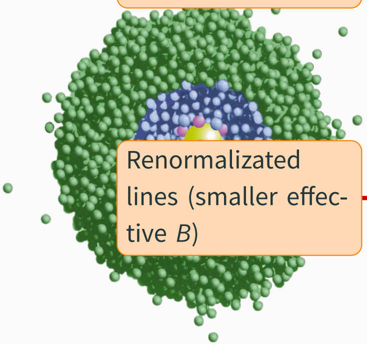
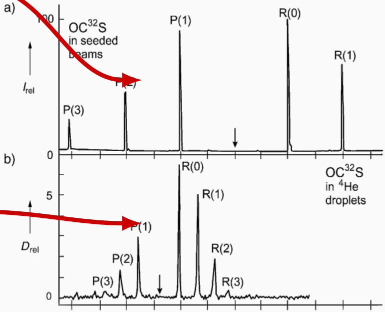
Molecules embedded into helium nanodroplets:

Rotational spectrum



Renormalizated lines (smaller effective B)



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Dynamical alignment of molecules in He nanodroplets

Dynamical alignment experiments:

- 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:

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

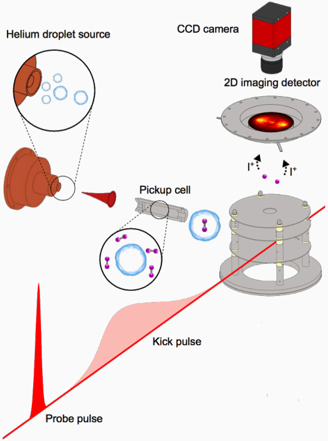


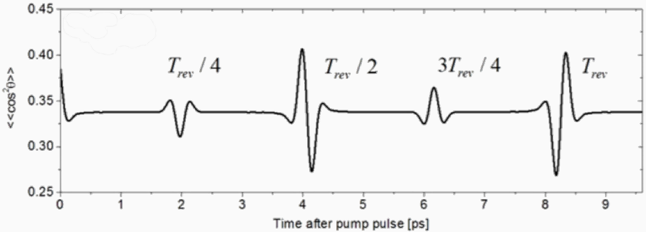
Image from B. Shepperson et al., Phys. Rev. Lett. **118**, 203203 (2017).

Dynamical alignment of molecules in He nanodroplets

Interaction of a free molecule 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:

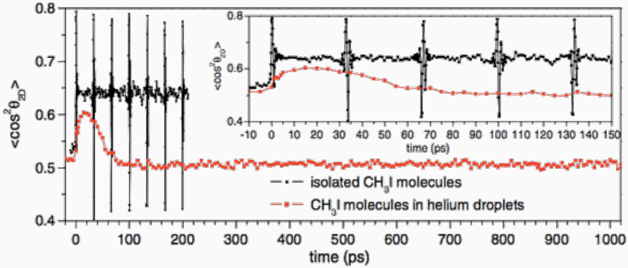


G. Kaya, Appl. Phys. B **6**, 122 (2016).

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).

Not even a qualitative understanding. Monte Carlo?

— Strong coupling

— Out-of-equilibrium dynamics

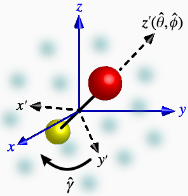
— Finite temperature ($B \sim k_B T$)

Canonical transformation

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

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

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



The $\hat{S}$ transformation takes us to the molecular frame.

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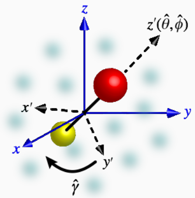

The $\hat{S}$ transformation takes us to the molecular frame.

- Macroscopic deformation of the bath, exciting an infinite number of bosons (cf. Lee-Low-Pines for the polaron).
- Simplifies angular momentum algebra.
- Hamiltonian diagonalizable through a coherent state transformation 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).

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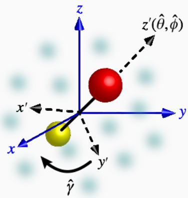

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Th… $\hat{S}$ transformation takes us to the …r frame.

✓ Strong coupling

— Out-of-equilibrium dynamics

— Finite temperature $(B \sim k_B T)$

- Macrosc… …an infinite number of bosons (cf. Lee-L…
- Simplifie…
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R. Schmidt and M. Lemeshko, Phys. Rev. X **6**, 011012 (2016).

Dynamics: time-dependent variational Ansatz

We use a time-dependent variational Ansatz:

$$|\psi\rangle = g_{LM}(t)\,|0\rangle_{\mathrm{bos}}\,|LM0\rangle + \sum_{k\lambda n} \alpha^{LM}_{k\lambda n}(t) b^\dagger_{k\lambda n}\,|0\rangle_{\mathrm{bos}}\,|LMn\rangle$$

Lagrangian on the variational manifold defined by $|\psi\rangle$:

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

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 $x_i = \{g_{LM}, \alpha^{LM}_{k\lambda n}\}$.

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

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

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 $x_i = \{g_{LM}, \alpha^{LM}_{k\lambda n}\}$.

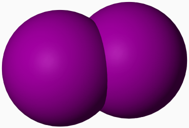
$$\begin{cases} \dot{g}_{LM}(t) = \dots \\ \dot{\alpha}^{LM}_{k\lambda n}(t) = \dots \end{cases}$$

✓ Strong coupling

✓ Out-of-equilibrium dynamics

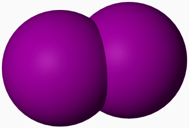
✓ Finite temperature ($B \sim k_B T$)

Theory vs. experiments: I_2



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

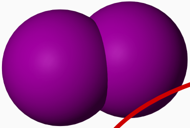
Theory vs. experiments: I_2



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

Which rotational states are populated as the laser is switched on, and after?

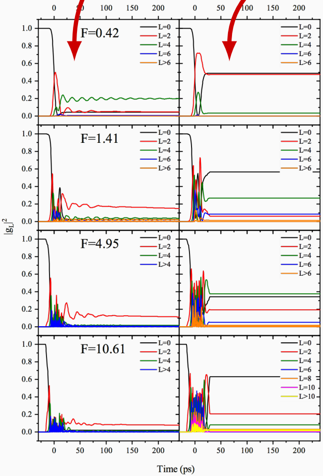
Theory vs. experiments: I_2



Comparison of the theory with preliminary experiment[...]dt group, Aarhus University,[...]s: I_2.

In Helium

Free molecule

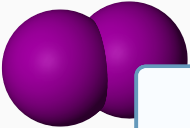


Which rotational states are populated as the laser is switched on, and after?

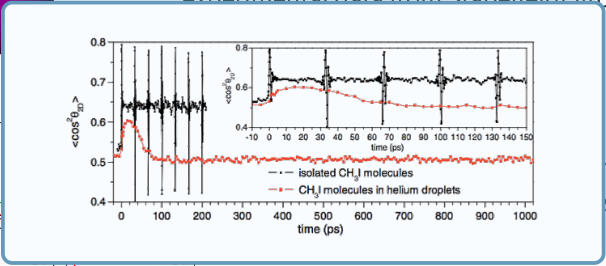
Free case: the angular momentum goes to the molecule.

In a Helium droplet: the angular momentum goes to the molecule *and* to the bath.

Theory vs. experiments: I_2

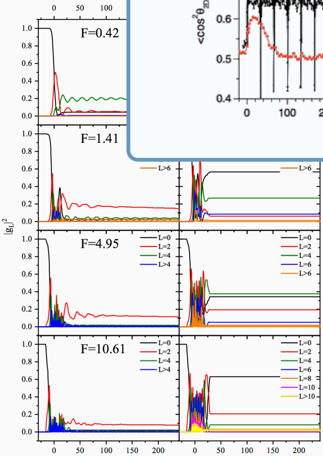


Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus



re

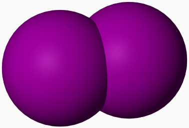
switched



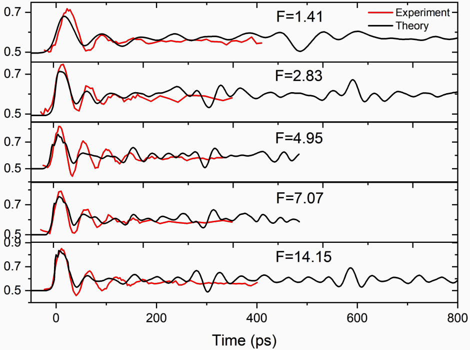
Free case: the angular momentum goes to the molecule.

In a Helium droplet: the angular momentum goes to the molecule *and* to the bath.

Theory vs. experiments: I_2



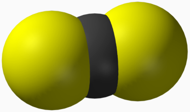
Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus University, for different molecules: I_2.



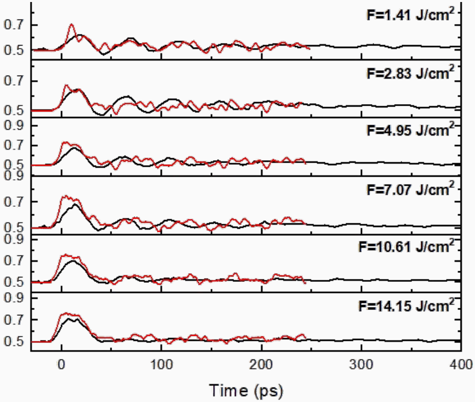
$\left\langle \cos^2 \hat{\theta}_{2D} \right\rangle (t)$

Laser fluence F measured in J/cm^2

Theory vs. experiments: CS_2

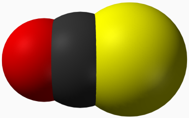


Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus University, for different molecules: CS_2.

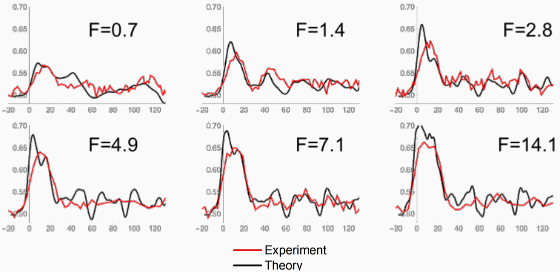


$$\left\langle \cos^2 \hat{\theta}_{2D} \right\rangle (t)$$

Theory vs. experiments: *OCS*



Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus University, for different molecules: *OCS*.



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

Laser fluence F measured in J/cm^2, time measured in *ps*.

Conclusions

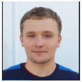
- The angulon quasiparticle: a quantum rotor dressed by a field of many-body excitations.
- Diagrammatic approach to angular momentum in a many-body context.
- Canonical transformation and finite-temperature variational Ansatz.
- Out-of-equilibrium dynamics of molecules in He nanodroplets can be interpreted in terms of angulons.

IST AUSTRIA

Institute of Science and Technology

Dynamical alignment experiments

Lemeshko group @ IST Austria:

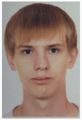


Misha Lemeshko

Dynamics in He

Enderalp Yakaboylu

Xiang Li



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Timur Tscherbul (Reno)

Thank you for your attention.

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Backup slide # 1

Free rotor propagator

$$G_{0,\lambda}(E) = \frac{1}{E - B\lambda(\lambda+1) + \mathrm{i}\delta}$$

Interaction propagator

$$\chi_\lambda(E) = \sum_k \frac{|U_\lambda(k)|^2}{E - \omega_k + \mathrm{i}\delta}$$

Backup slide # 2

Backup slide # 3