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

G. Bighin and M. Lemeshko
Institute of Science and Technology Austria

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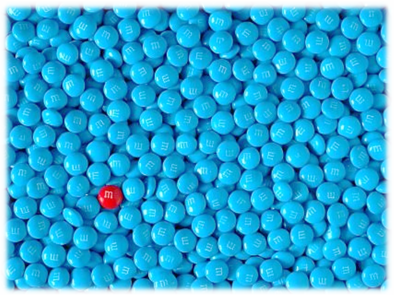
Impurity problems

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

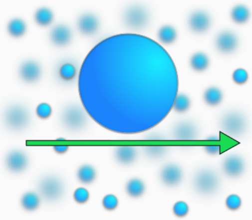
How are the properties of the particle modified by the interaction?

$\mathcal{O}(10^{23})$ 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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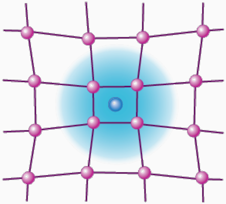

Image from: F. Chevy, Physics **9**, 86.

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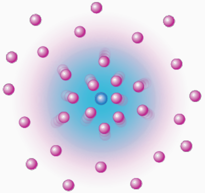

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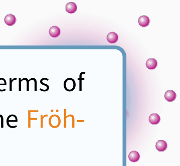
This scenario can be formalized in terms of quasiparticles using the polaron and the Fröhlich Hamiltonian.



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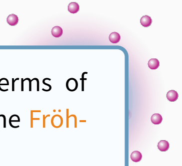

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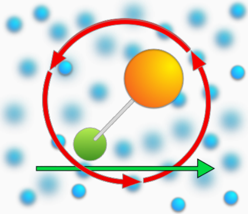

Composite impurity: translational *and internal* (i.e. rotational) degrees of freedom/linear and angular momentum exchange.

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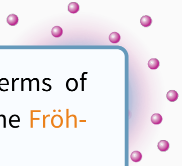

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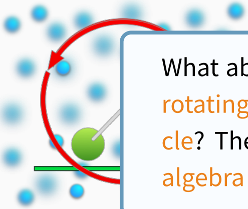


What about a rotating particle? Can there be a rotating counterpart of the polaron quasiparticle? The main difficulty: the non-Abelian SO(3) algebra describing rotations.

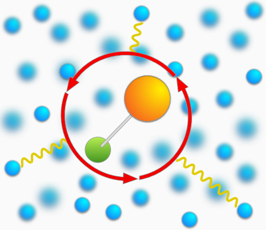
nd internal
near and

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].



[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.

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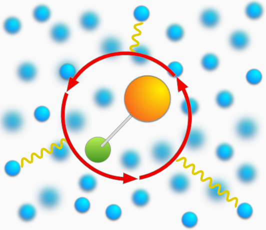

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Composite impurities: where to find them

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

- Molecules embedded into helium nanodroplets.

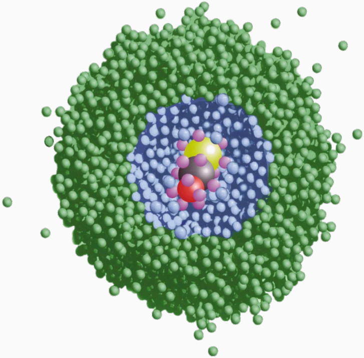

Image from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

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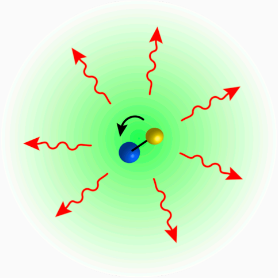

B. Midya, M. Tomza, R. Schmidt, and M. Lemeshko, Phys. Rev. A **94**, 041601(R) (2016).

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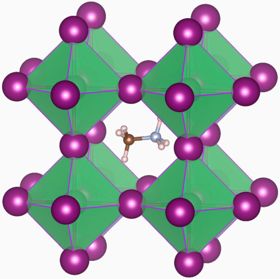

T. Chen et al., PNAS **114**, 7519 (2017).
J. Lahnsteiner et al., Phys. Rev. B **94**, 214114 (2016).
Image from: C. Eames et al, Nat. Comm. **6**, 7497 (2015).

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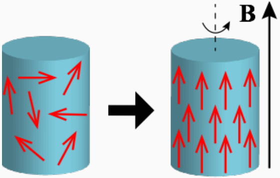

J.H. Mentink, M.I. Katsnelson, M. Lemeshko, *"Quantum many-body dynamics of the Einstein-de Haas effect"*, arXiv:1802.01638

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- Ultracold
- Rotating molecules inside a 'cage' in perovskites.
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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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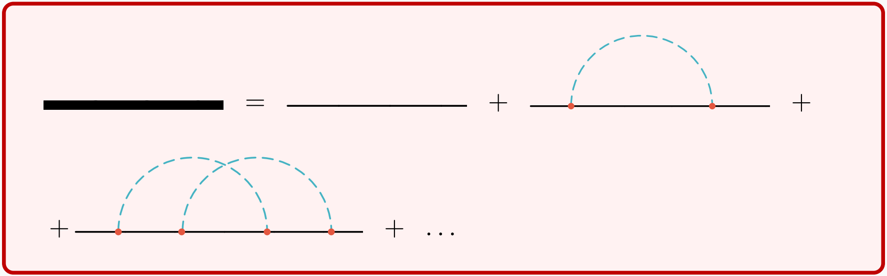
Perturbation theory/Feynman diagrams:



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

Perturbative approach and Feynman diagrams

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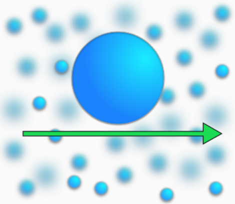
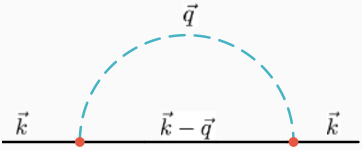
Perturbation theory/Feynman diagrams:

Fröhlich polaron





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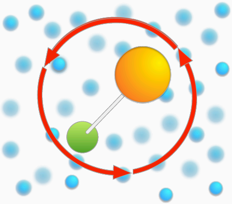

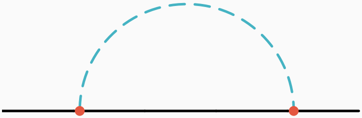

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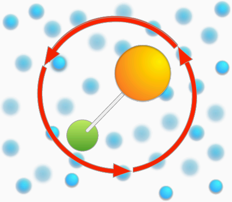
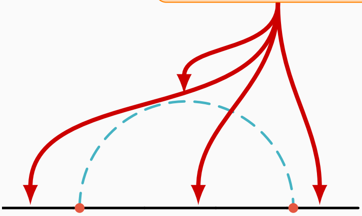
Angulon

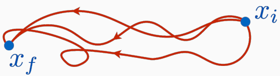




From path integral to Feynman rules

The path integral in QM describes the transition amplitude between two states with a weighted average over all trajectories, S is the classical action.

$$G(x_i, x_f; t_f - t_i) = \langle x_f, t_f | x_i, t_i \rangle = \int \mathcal{D}x \, e^{\mathrm{i}S[x(t)]}$$



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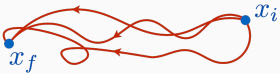

Linear molecule, two angles θ and ϕ.

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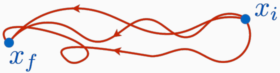

Linear molecule, two angles θ and ϕ.

$$\begin{aligned} G(\theta_i, \phi_i; \theta_f, \phi_f; T) &= \int \mathcal{D}\theta \mathcal{D}\phi \prod_{k\lambda\mu} \mathcal{D}b_{k\lambda\mu} \, e^{\mathrm{i}(S_{\text{mol}} + S_{\text{bos}} + S_{\text{mol-bos}})} \\ &= \int \mathcal{D}\theta \mathcal{D}\phi \, e^{\mathrm{i}S_{\text{mol}} + \mathrm{i}S_{\text{int}}} \\ &= \int \mathcal{D}\theta \mathcal{D}\phi \, e^{\mathrm{i}S_{\text{mol}}} \left(1 + \mathrm{i}S_{\text{int}} - \frac{1}{2}S_{\text{int}}^2 + \dots\right) = G^{(0)} + G^{(1)} + G^{(2)} + \dots \end{aligned}$$

The result can be interpreted as a diagrammatic expansion, from which one can derive the Feynman rules for angular momentum.

From path integral to Feynman rules

The path integral in QM describes the transition amplitude between two states with a weighted average over all trajectories. S is the classical action.

Open quantum systems: a quantum rotor with memory.

$$S = \underbrace{\int_0^T \mathrm{d}t\; B\mathbf{J}^2}_{S_{\text{mol}}} + \underbrace{\frac{\mathrm{i}}{2}\int_0^T \mathrm{d}t \int_0^T \mathrm{d}s \sum_\lambda P_\lambda(\cos\gamma(t,s))\mathcal{M}_\lambda(|t-s|)}_{S_{\text{int}}}$$

$$G(x_i, x_f; t_f - t_i) = \langle x_f, t_f$$

angles θ and ϕ.

$$G(\theta_i, \phi_i; \theta_f, \phi_f; T) = \int \mathcal{D}\theta\mathcal{D}\phi \prod_{k\lambda\mu} \mathcal{D}b_{k\lambda\mu}\, e^{\mathrm{i}(S_{\text{mol}} + S_{\text{bos}} + S_{\text{mol-bos}})}$$

$$= \int \mathcal{D}\theta\mathcal{D}\phi\, e^{\mathrm{i}S_{\text{mol}} + \mathrm{i}S_{\text{int}}}$$

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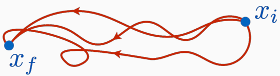

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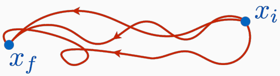
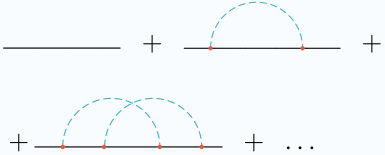


$$G^{(0)} + G^{(1)} + G^{(2)} + \ldots =$$



$G(\theta_i, \phi_i; \theta_f, \phi_$

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The result can be interpreted as a diagrammatic expansion, from which one can derive the Feynman rules for angular momentum.

Feynman rules

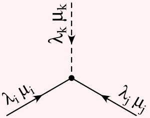
Each free propagator

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

Each phonon propagator

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

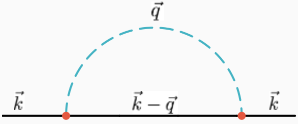
Each vertex



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

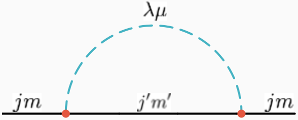
GB and M. Lemeshko, Phys. Rev. B **96**, 419 (2017).

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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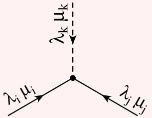
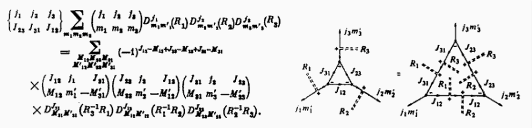
Each vertex



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

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

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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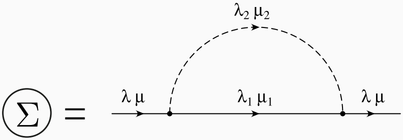
First order:



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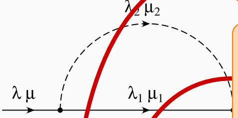
$|\text{bath}\rangle\,|\text{molecule}\rangle$

Quasiparticle weight

Variational coefficients

Clebsch-Gordan to couple angular momenta

First order: $\Sigma =$



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Angulon spectral function

Let us use the Feynman diagrams! The plan is:

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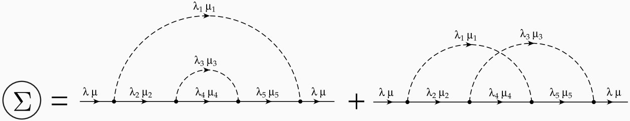
2. Dyson equation to obtain the angulon Green's function (G)
3. Spectral function ($\mathcal{A}$)

Second order:

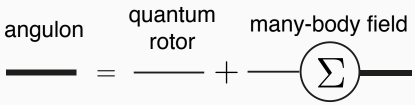


Angulon spectral function

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 ($\mathcal{A}$)

Dyson equation



Angulon spectral function

Let us use the Feynman diagrams! The plan is:

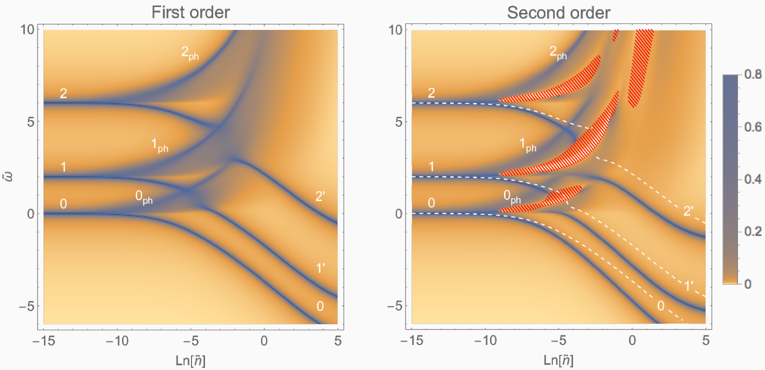
1. Self-energy (Σ)
2. Dyson equation to obtain the angulon Green's function (G)
3. Spectral function ($\mathcal{A}$)

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

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

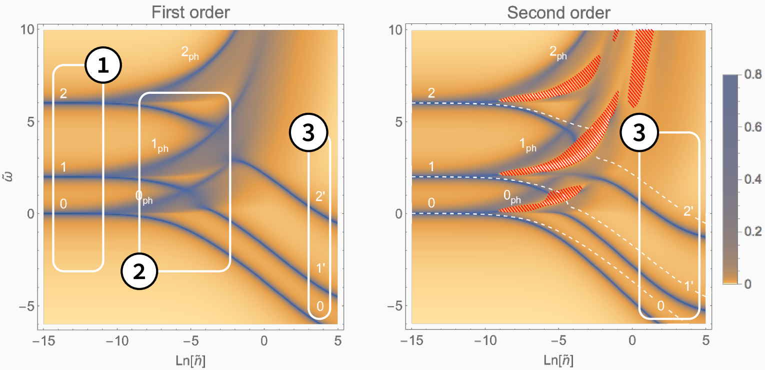
Angulon quasiparticle spectrum

Angulon quasiparticle spectrum as a function of the bath density:

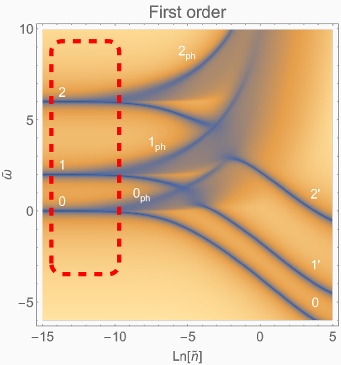


Angulon quasiparticle spectrum

Angulon quasiparticle spectrum as a function of the bath density:



Angulon quasiparticle spectrum: low density

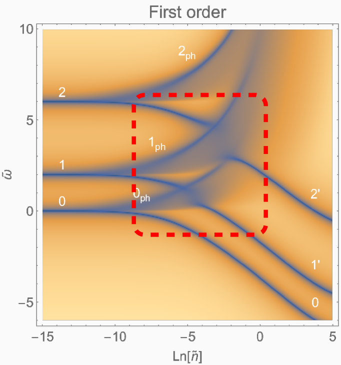


Low density: free rotor spectrum, $E = BL(L+1)$.

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).

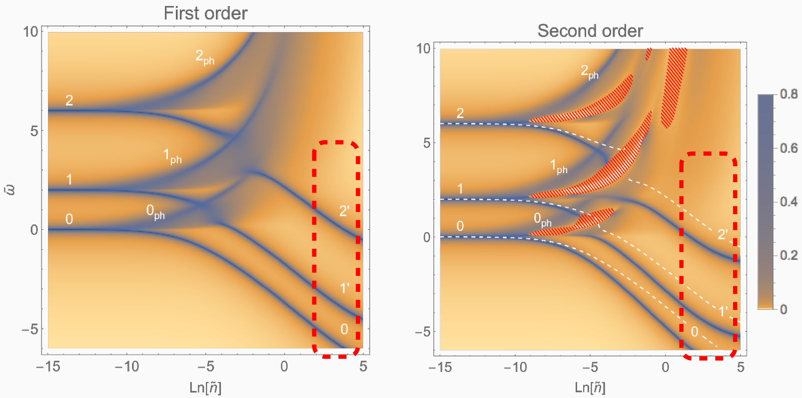
Angulon quasiparticle spectrum: instability



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

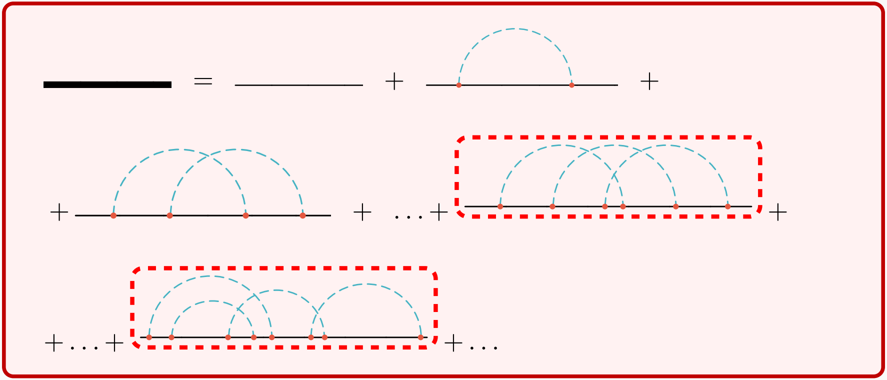
Experimental observation: I. N. Cherepanov, M. Lemeshko, *"Fingerprints of angulon instabilities in the spectra of matrix-isolated molecules"*, Phys. Rev. Materials **1**, 035602 (2017.

Angulon quasiparticle spectrum: high density



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

What about higher orders?



Diagrammatic Monte Carlo:[1] a stochastic process sampling among all diagrams.

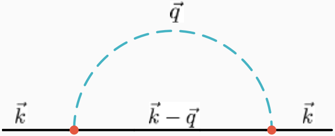
Up to now: structureless particles (Fröhlich polaron, Holstein polaron), or particles with a very simple internal structure (e.g. spin 1/2).

What about molecules[2]?

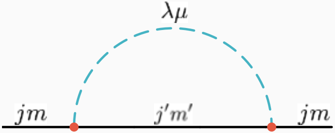
[1] N. V. Prokof'ev and B. V. Svistunov, Phys. Rev. Lett. **81**, 2514 (1998).
[2] GB, T.V. Tscherbul, M. Lemeshko, arXiv:1803:07990

Diagrammatics for a rotating impurity

Moving particle: linear momentum circulating on lines.



Rotating particle: angular momentum circulating on lines.



Diagrammatics for a rotating impurity

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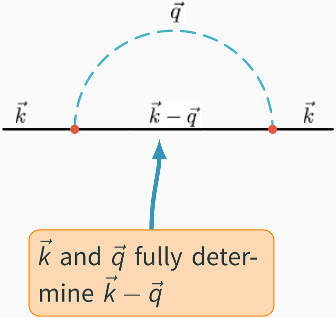
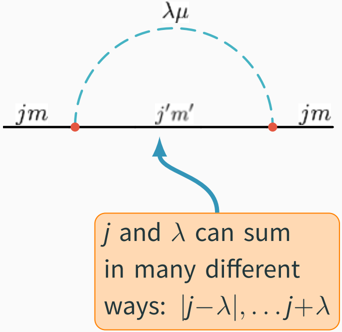


Rotating particle: angular momentum circulating on lines.



Diagrammatics for a rotating impurity

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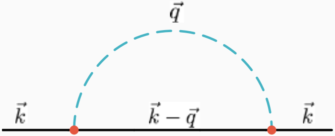

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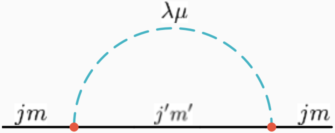
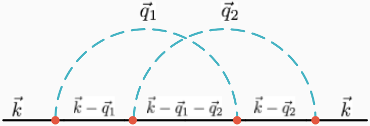
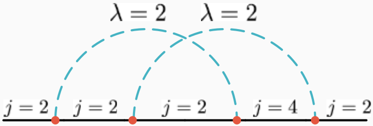


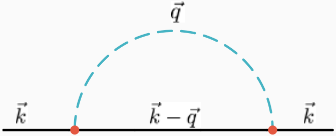
It gets weirder... Down the rabbit hole of angular momentum composition!



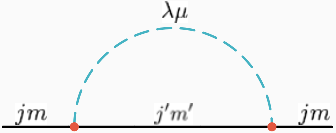


Diagrammatics for a rotating impurity

Moving particle: linear momentum circulating on lines.



Rotating particle: angular momentum circulating on lines.



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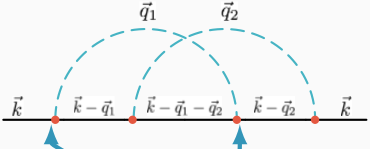
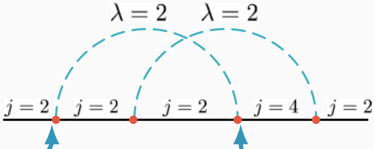




The phonon takes away $\vec{q}_1$ momentum...

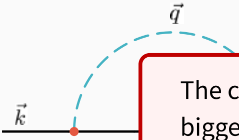
...and gives back $\vec{q}_1$ momentum

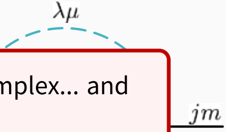
The phonon does not subtract angular momentum from the impurity...

...but gives back two quanta!

Moving particle: linear momentum circulating on lines.

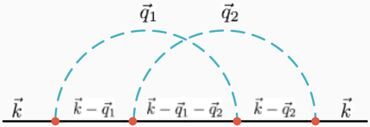
Rotating particle: angular momentum circulating on lines.

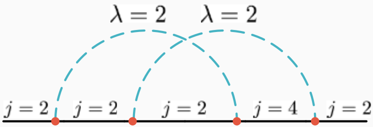




The configuration space is more complex... and bigger! We need different updates.

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



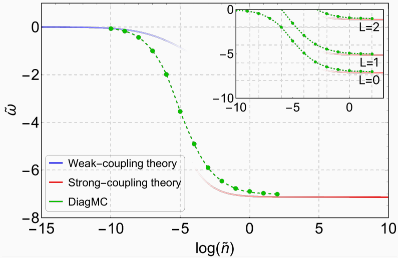


DiagMC: results

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.

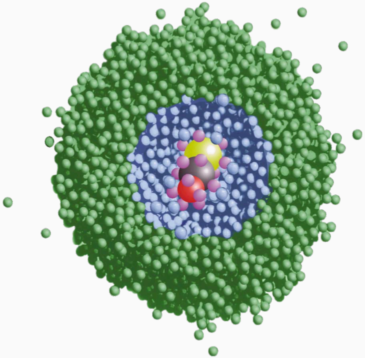


[1] GB, T.V. Tscherbul, M. Lemeshko, arXiv:1803:07990.
[2] R. Schmidt and M. Lemeshko, Phys. Rev. Lett. **114**, 203001 (2015).
[3] R. Schmidt and M. Lemeshko, Phys. Rev. X **6**, 011012 (2016).

Out-of-equilibrium dynamics of molecules in He nanodroplets

Dynamical alignment of molecules in He nanodroplets

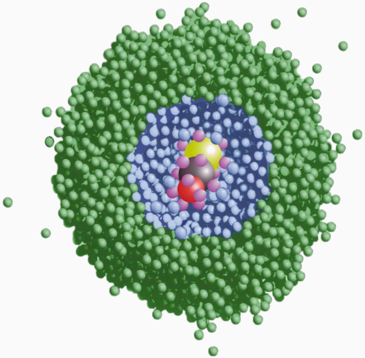
Molecules embedded into helium nanodroplets:

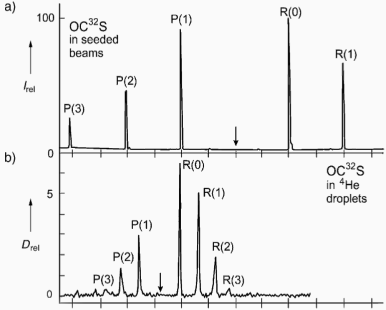


Images from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

Dynamical alignment of molecules in He nanodroplets

Molecules embedded into helium nanodroplets:





Gas phase (free)

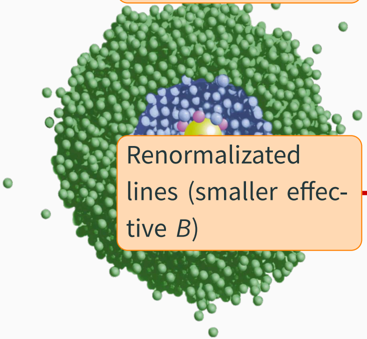
in ^{4}He

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

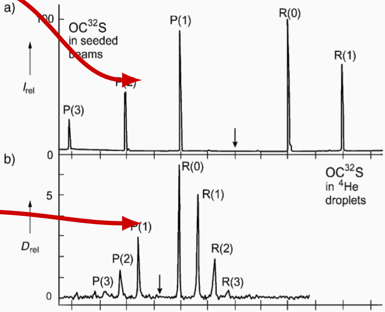
Dynamical alignment of molecules in He nanodroplets

Molecules embedded into helium nanodroplets:

Rotational spectrum



Renormalizated lines (smaller effective B)



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

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:

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

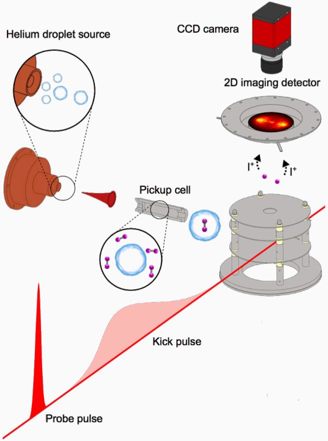


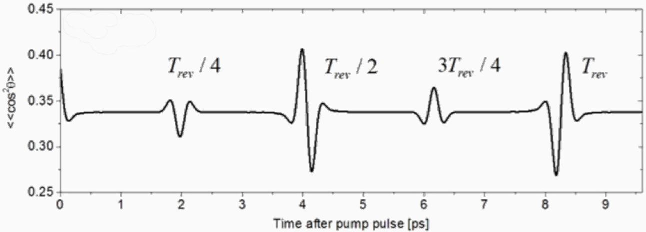
Image from B. Shepperson et al., Phys. Rev. Lett. **118**, 203203 (2017).

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:

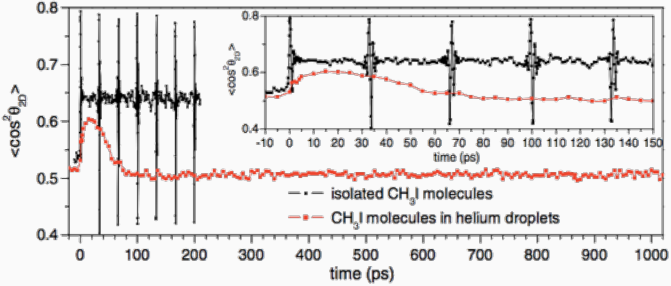


G. Kaya, Appl. Phys. B **6**, 122 (2016).

Movie

Dynamical alignment of molecules in He nanodroplets

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



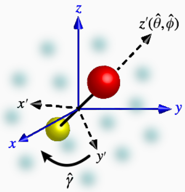
Stapelfeldt group, Phys. Rev. Lett. **110**, 093002 (2013).

Not even a qualitative understanding. Monte Carlo? Challenges:

- 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.

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

- Accounts for a macroscopic deformation of the bath, exciting an infinite number of bosons.
- Simplifies angular momentum algebra.
- An expansion in bath excitations after $\hat{S}$ is a strong-coupling expansion.

Time-dependent variational Ansatz

After the canonical transformation $\hat{S}$, we can use as time-dependent variational Ansatz an expansion in bath excitations:

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

Lagrangian:

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

Equations of motion:

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

where $x_i = \{g_{LM}, \alpha_{k\lambda n}\}$.

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

Finite-temperature dynamics

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

For the bath: defining the 'Chevy operator'

$$\hat{O} = g_{LM}(t)\,|LM0\rangle\,\mathbb{1} + \sum_{k\lambda n} \alpha^{LM}_{k\lambda n}(t)\,|LMn\rangle\,\hat{b}^{\dagger}_{k\lambda n}$$

at $T = 0$ the Lagrangian is

$$\mathcal{L}_{T=0} = \langle 0|\hat{O}^{\dagger}(\mathrm{i}\partial_t - \hat{H})\hat{O}|0\rangle_{\text{bos}} \;,$$

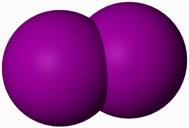
suggesting that at finite temperature

$$\mathcal{L}_T = \mathrm{Tr}\left[\rho_0\,\hat{O}^{\dagger}(\mathrm{i}\partial_t - \hat{H})\hat{O}\right]$$

where ρ_0 is the density matrix for the medium.

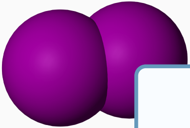
[1] A. R. DeAngelis and G. Gatoff, Phys. Rev. C **43**, 2747 (1991).
[2] W.E. Liu, J. Levinsen, M. M. Parish, *"Variational approach for impurity dynamics at finite temperature"*, arXiv:1805.10013

Theory vs. experiments: I_2

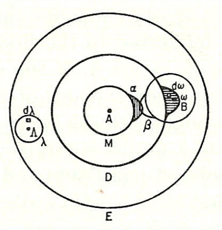


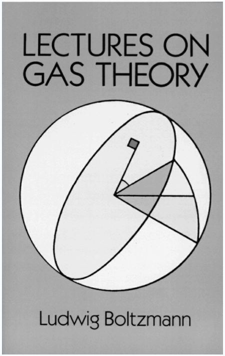
Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus University, for different molecules: I_2.

Theory vs. experiments: I_2

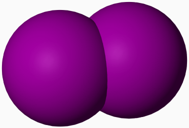


Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus



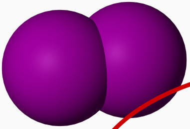


Theory vs. experiments: I_2



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

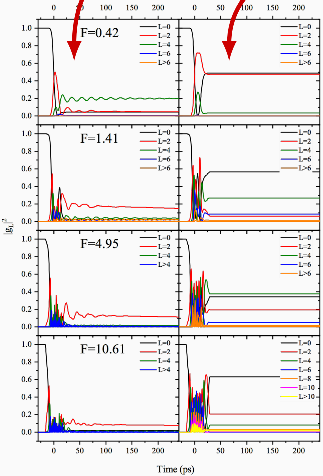
Which rotational states are populated as the laser is switched on, and after?



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

In Helium

Free molecule

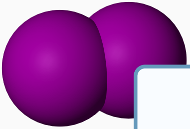


Which rotational states are populated as the laser is switched on, and after?

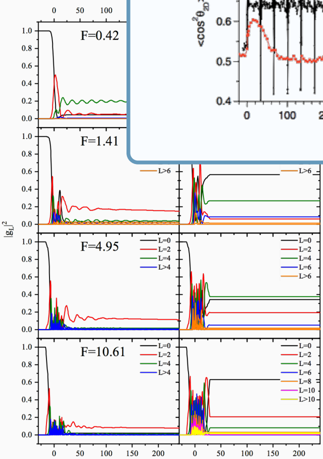
Free case: the angular momentum goes to the molecule.

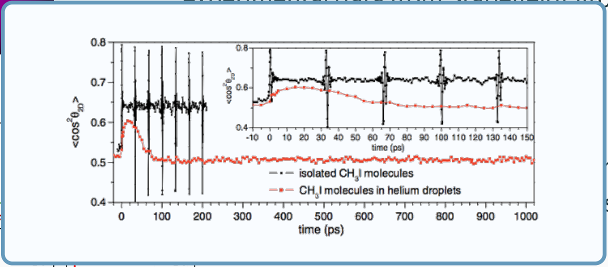
In a Helium droplet: the angular momentum goes to the molecule *and* to the bath.

Theory vs. experiments: I_2



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





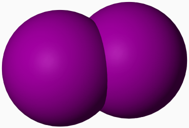
re

switched

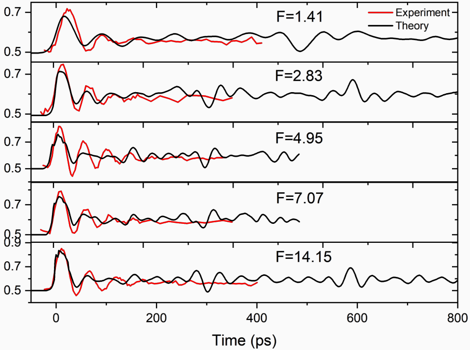
Free case: the angular momentum goes to the molecule.

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

Theory vs. experiments: I_2

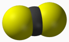


Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus University, for different molecules: I_2.

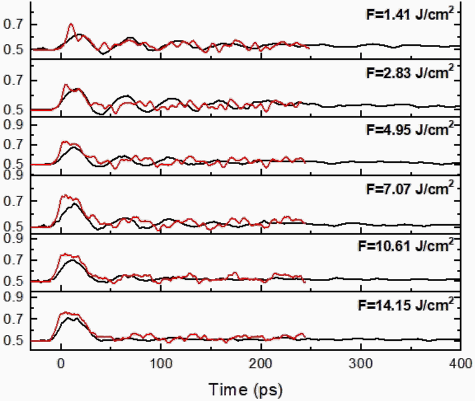


$$\left\langle \cos^2 \hat{\theta}_{2D} \right\rangle (t)$$

Theory vs. experiments: CS_2



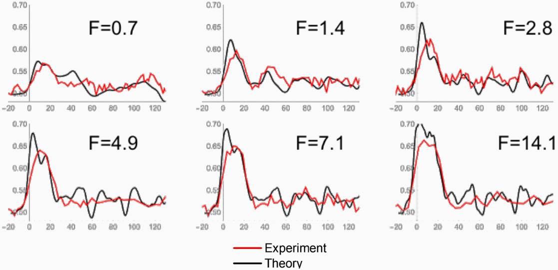
Comparison of the theory with preliminary experimental data from Stapelfeldt group, Aarhus University, for different molecules: CS_2.



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

Theory vs. experiments: *OCS*

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



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

Conclusions

- The angulon quasiparticle: a quantum rotor dressed by a field of many-body excitations.
- Angular momentum and Feynman diagrams.
- A technique for molecular simulations using the Diagrammatic Monte Carlo framework.
- Out-of-equilibrium dynamics of molecules in He nanodroplets can be interpreted in terms of angulons.

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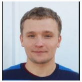
Institute of Science and Technology

Lemeshko group @ IST Austria:

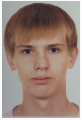


Misha Lemeshko

Dynamics in He

Enderalp Yakaboylu

Xiang Li



Igor Cherepanov

Wojciech Rządkowski

Collaborators:

Timur Tscherbul (Nevada Reno)

Diagrammatic MC

Thank you for your attention.

Der Wissenschaftsfonds.

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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