# A diagrammatic approach to composite, rotating impurities.

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

Padova, June 14th, 2017

# Summary

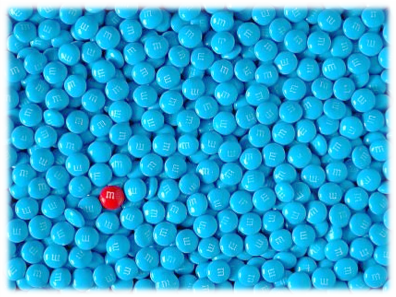
- Introduction: impurity problems
- The angulon quasiparticle
- A path integral/diagrammatic approach to the angulon
- The angulon spectrum
- Dynamics

# 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?

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

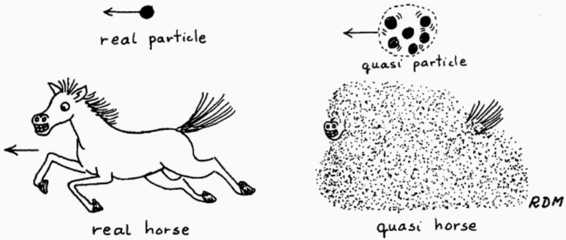


- Condensed matter (electrons in solids)
- Chemistry (molecules in a solution)
- Ultracold atoms (atomic impurities in a BEC)

# Quasiparticles

**Quasiparticles** provide a trick to understand what happens in a complex system.

Bare particle + field of many body excitations

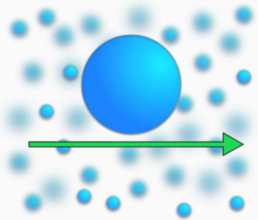


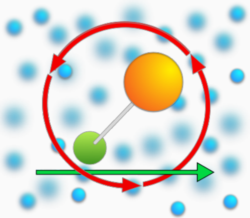

Picture from Richard D. Mattuck, *"A Guide to Feynman Diagrams in the Many-Body Problem"*.

# 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.





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

# 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.

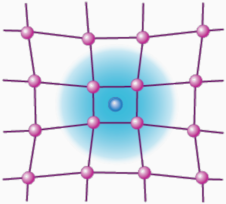
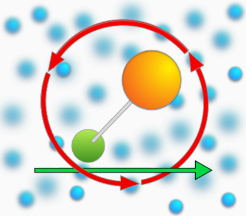


Image from: F. Chevy, Physics **9**, 86.



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

# 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.

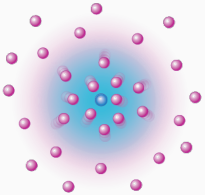
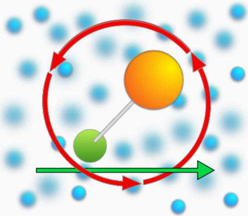


Image from: F. Chevy, Physics **9**, 86.



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

# From impurities to quasiparticles

**Structureless impurity:** translational
degrees of
momentu

> This scenario can be formalized using the polaron.

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

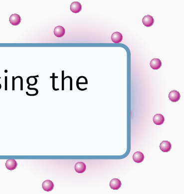
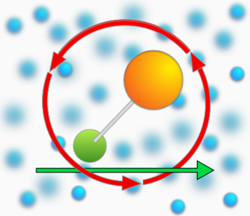


Image from: F. Chevy, Physics **9**, 86.



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

# From impurities to quasiparticles

**Structureless impurity:** translational
degrees of
momentum

This scenario can be formalized using the polaron.

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

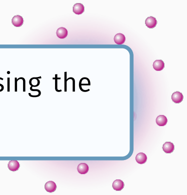
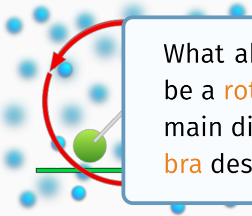


Image from: F. Chevy, Physics **9**, 86.



*and*
f
entum

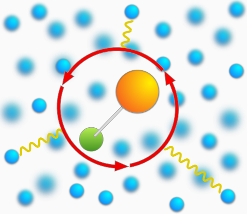
What about a rotating particle? Can there be a rotating analogue of the polaron? The main difficulty: the non-Abelian SO(3) algebra describing rotations.

# The angulon

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

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

- 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).
[3] M. Lemeshko, Phys. Rev. Lett. **118**, 095301 (2017).
[4] Y. Shchadilova, *"Viewpoint: A New Angle on Quantum Impurities"*, Physics **10**, 20 (2017).

# Composite impurities and where to find them

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

- Molecules embedded into helium nanodroplets (rotational spectra, rotational constant renormalization).

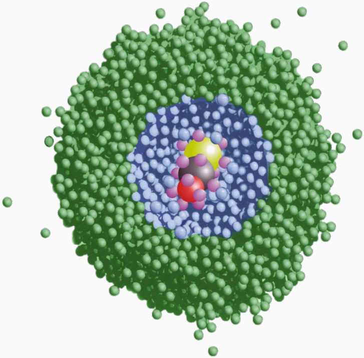
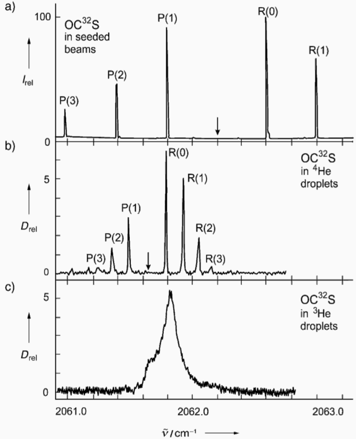


Image from: J. P. Toennies and A. F. Vilesov, Angew. Chem. Int. Ed. **43**, 2622 (2004).

# Composite impurities and where to find them

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

- Molecules embedded into helium nanodroplets (rotational spectra, rotational constant renormalization).




Gas phase

in $^4$He

in $^3$He

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

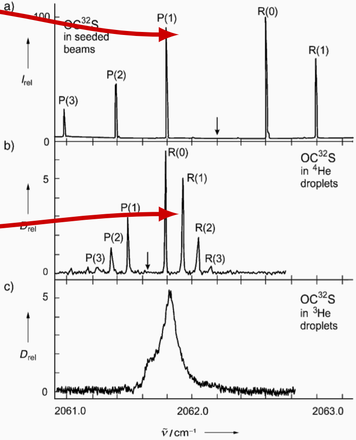
# Composite impurities and where to find them

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

- Mole… helium nanodroplets (rotational spectra, rotational constant renormalization).

Rotational spectrum

Renormalizated lines (smaller effective $B$)




Gas phase

in $^4$He

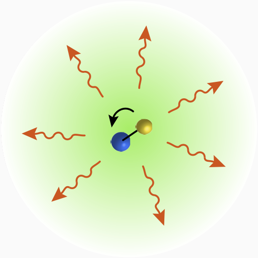
in $^3$He

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

# Composite impurities and where to find them

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

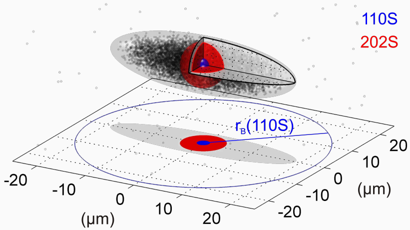
- Molecules embedded into helium nanodroplets (rotational spectra, rotational constant renormalization).
- Ultracold molecules and ions.



# Composite impurities and where to find them

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

- Molecules embedded into helium nanodroplets (rotational spectra, rotational constant renormalization).
- Ultracold molecules and ions.
- Electronic excitations in Rydberg atoms.



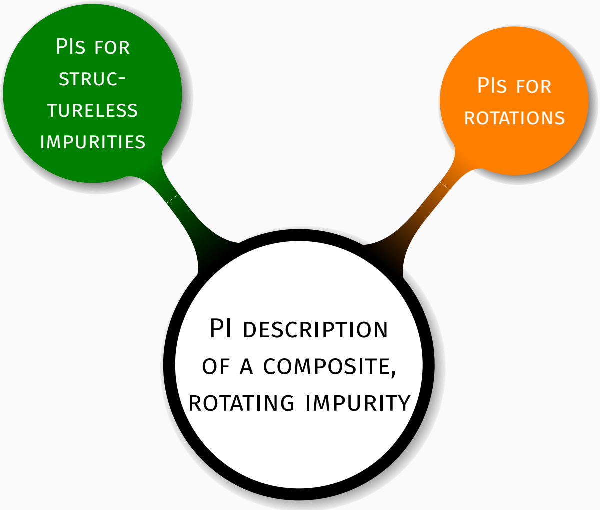

Pfau group, Nature **502**, 664 (2013).

# Composite impurities and where to find them

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

- Molecules embedded into helium nanodroplets (rotational spectra, rotational constant renormalization).
- Ultracold molecules and ions.
- Electronic excitations in Rydberg atoms.
- Angular momentum transfer from the electrons to a crystal lattice.

# Path integral description for the angulon

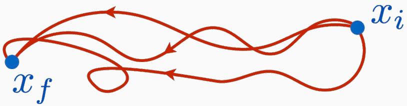



**Main reference**: GB and M. Lemeshko, arXiv:1704.02616

# Path integral description for the angulon

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

$$G(x_i, x_f; t_f - t_i) = \left\langle x_f, t_f \middle| x_i, t_i \right\rangle = \int \mathcal{D}x \, e^{iS[x(t)]}$$



# Path integral description for the angulon

The angulon's Green function is calculated in the same way. We need

- Molecular coordinates: two angles $(\theta, \phi)$ describing the orientation of the molecule.
- An infinite number of harmonic oscillators $b_{k\lambda\mu}$ to describe the bosonic bath.

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

# Path integral description for the angulon

The angulon's Green function is calculated in the same way. We need

- Molecular coordinates: two angles $(\theta, \phi)$ describing the orientation of the molecule.
- An infinite number of harmonic oscillators $b_{k\lambda\mu}$ t… bosonic bath.

Derived from the Hamiltonian

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

# Path integral description for the angulon

The angulon's Green function is calculated in the same way. We need

- Molecular coordinates: two angles $(\theta, \phi)$ describing the orientation of the molecule.
- An infinite number of harmonic oscillators $b_{k\lambda\mu}$ to describe the bosonic bath.

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

Critically the environment $(b_{k\lambda\mu})$ can be integrated out exactly

$$G(\theta_i, \phi_i \to \theta_f, \phi_f; T) = \int \mathcal{D}\theta \mathcal{D}\phi \, e^{\mathrm{i}S_{\text{eff}}[\theta(t), \phi(t)]}$$

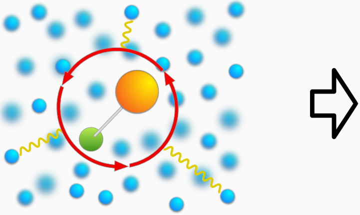
and included in an effective action $S_{\text{eff}}$.

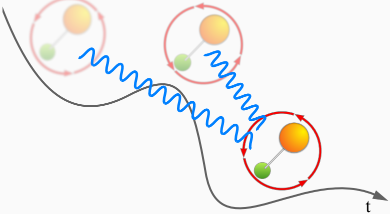
# Path integral description for the angulon

A closer look at the effective action:

$$S_{\text{eff}} = \underbrace{\int_0^T \mathrm{d}t\, B\mathbf{J}^2}_{S_0} + \underbrace{\frac{\mathrm{i}}{2}\int_0^T \mathrm{d}t \int_0^T \mathrm{d}s \sum_\lambda P_\lambda(\cos\gamma(t,s))\mathcal{M}_\lambda(|t-s|)}_{S_{\text{int}}}$$

- A term describing a free molecule $\sim B\mathbf{J}^2$.
- A memory term accounting for the many-body environment, a function of the angle $\gamma(t,s)$ between the angulon position at different times.





# Path integral description for the angulon

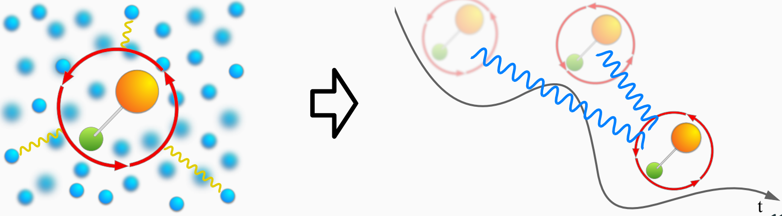
A closer look at the effective action:

Legendre polynomials → $P_\lambda$

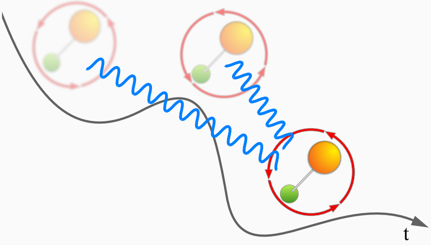
Memory kernel → $\mathcal{M}_\lambda$

$$S_{\text{eff}} = \underbrace{\int_0^T \mathrm{d}t\, B\mathbf{J}^2}_{S_0} + \underbrace{\frac{\mathrm{i}}{2}\int_0^T \mathrm{d}t \int_0^T \mathrm{d}s \sum_\lambda P_\lambda(\cos\gamma(t,s))\mathcal{M}_\lambda(|t-s|)}_{S_{\text{int}}}$$

- A term describing a free molecule $\sim B\mathbf{J}^2$.
- A memory term accounting for the many-body environment, a function of the angle $\gamma(t,s)$ between the angulon position at different times.



# Path integral description for the angulon



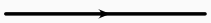
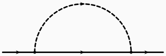

- The many-body problem is reformulated in terms of a self-interacting free molecule.
- Time-non-local interaction (cf. Caldeira-Leggett, polaron, more generally: open quantum systems)
- The interaction term is very difficult to treat: it encodes exactly the many-body nature of the problem.

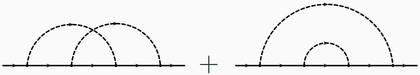
# Diagrammatic theory of angular momentum in a many-body bath

We treat the interaction as a perturbation

$$G = \int \mathcal{D}\theta\mathcal{D}\phi\, e^{\mathrm{i}S_0 + \mathrm{i}S_{\mathrm{int}}} = \int \mathcal{D}\theta\mathcal{D}\phi\, e^{\mathrm{i}S_0} \left(1 + \mathrm{i}S_{\mathrm{int}} - \frac{1}{2}S_{\mathrm{int}}^2 + \ldots\right) = G_0 + G_1 + G_2 + \ldots$$

The result can be interpreted as a diagrammatic expansion (solid lines represent a free rotor, dashed lines are the interaction)

- $G_0(\theta_i, \phi_i \to \theta_f, \phi_f; T)$ is the Green's function for a free rotor 
- $G_1(\theta_i, \phi_i \to \theta_f, \phi_f; T)$ is the one-loop correction 
- $G_2(\theta_i, \phi_i \to \theta_f, \phi_f; T)$ is the two-loop correction

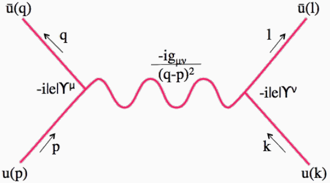


- and so on...

# Feynman rules

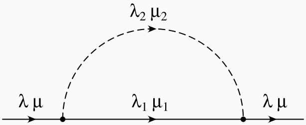
## "Standard" Feynman rules

- Start with real-space Green's function $G(\mathbf{r}, \mathbf{r}')$
- Fourier transform
- Assign a momentum $\mathbf{p}_i$ to every line
- Each loop: integral over momenta
- Enforce momentum conservation

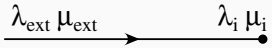
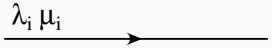
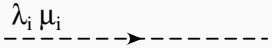
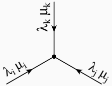



## Feynman rules for the angulon

- Green's function depends on angles $G(\theta, \phi, \theta', \phi')$
- Spherical harmonics $Y_{\lambda\mu}(\theta, \phi)$ expansion
- Assign an angular momentum $(\lambda_i, \mu_i)$ to every line
- Each line: sums over angular momenta
- Enforce angular momentum conservation



# Feynman rules for the angulon

| | |
|---|---|
| Each external line  | $\sum_{\lambda_i\mu_i}(-1)^{\mu_i} G_{0,\lambda_i}\delta_{\lambda_{\text{ext}},\lambda_i}\delta_{\mu_{\text{ext}},\pm\mu_i}$ |
| Each internal $G_0$ line  | $\sum_{\lambda_i\mu_i}(-1)^{\mu_i} G_{0,\lambda_i}$ |
| Each internal $\chi$ line  | $\sum_{\lambda_i\mu_i}(-1)^{\mu_i} \chi_{\lambda_i}$ |
| Each vertex  | $(-1)^{\lambda_i}\langle\lambda_i\|\|Y^{(\lambda_j)}\|\|\lambda_k\rangle \begin{pmatrix}\lambda_i & \lambda_j & \lambda_k \\ \mu_i & \mu_j & \mu_k\end{pmatrix}$ |

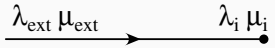
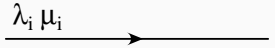
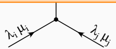
Free rotor propagator

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

Interaction propagator

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

# Feynman rules for the angulon

| | |
|---|---|
| Each external line   | $\sum_{\lambda_i \mu_i} (-1)^{\mu_i} G_{0,\lambda_i} \delta_{\lambda_{\text{ext}},\lambda_i} \delta_{\mu_{\text{ext}},\pm\mu_i}$ |
| Each internal $G_0$ line   | $\sum_{\lambda_i \mu_i} (-1)^{\mu_i} G_{0,\lambda_i}$ |
| Each internal $\chi$ line | $\sum_{\lambda_i \mu_i} (-1)^{\mu_i} \chi_{\lambda_i}$ |
|   | $(-1)^{\lambda_i} \langle \lambda_i \| \|Y^{(\lambda_j)}\| \| \lambda_k \rangle \begin{pmatrix} \lambda_i & \lambda_j & \lambda_k \\ \mu_i & \mu_j & \mu_k \end{pmatrix}$ |

3*j* symbol: momentum conservation

Free rotor propagator

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

Molecule-bath interaction

Bath dispersion relation

Interaction propagator

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

# Feynman rules for the angulon

Dressed diagrams:

$$\Big(\text{Diagram}\Big) = \Big(\text{Skeleton diagram}\Big) \times \Big(\text{Dress}\Big)$$

Conservation of angular momentum/geometry

Many-body excitations

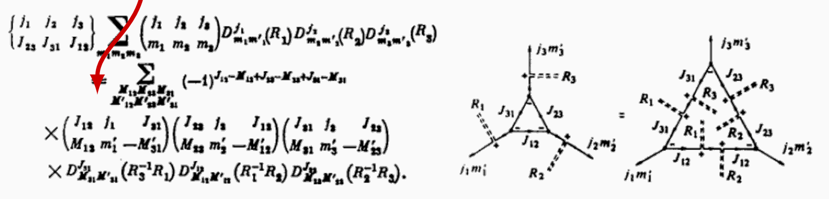
# Feynman rules for the angulon

Dressed diagrams:

$$\Big(\text{Diagram}\Big) = \Big(\text{Skeleton diagram}\Big) \times \Big(\text{Dress}\Big)$$

Conservation of angular momentum/geometry

Many-body excitations



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

The first part coincides with the diagrammatic theory of angular momentum, describing coupling of many angular momenta – in an abstract way – in the context of theoretical atomic spectroscopy.

# Feynman rules for the angulon

Dressed diagrams:

$$\Big(\text{Diagram}\Big) = \Big(\text{Skeleton diagram}\Big) \times \Big(\text{Dress}\Big)$$

Conservation of angular momentum/geometry

Many-body excitations

# Angulon spectral function

Let us use the theory! The plan is simple:

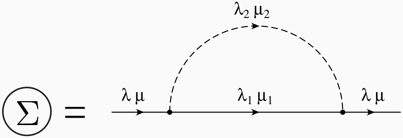
1. Self-energy ($\Sigma$)
2. Dyson equation to obtain the angulon Green's function (G)
3. Spectral function ($\mathcal{A}$)

# Angulon spectral function

Let us use the theory! The plan is simple:

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

First order:




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

$$|\psi\rangle = Z_{LM}^{1/2} |0\rangle \, |LM\rangle + \sum_{\substack{k\lambda\mu \\ jm}} \beta_{k\lambda j} C^{LM}_{jm,\lambda\mu} b^\dagger_{k\lambda\mu} |0\rangle \, |jm\rangle$$

# Angulon spectral function

Let us use the theory! The plan is simple:

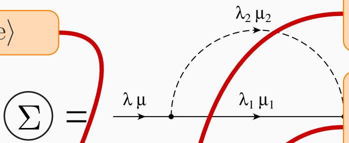
1. Self-energy ($\Sigma$)
2. Dyson equation to obtain the angulon Green's function (G)
3. Spectral function ($\mathcal{A}$)

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

Variational coeffi- cients

Quasiparticle weight

Fi




Clebsch-Gordan to couple angular momenta

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

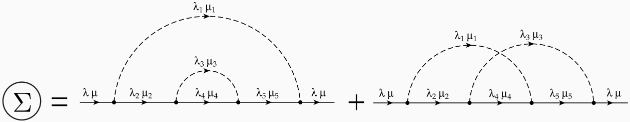
$$|\psi\rangle = Z_{LM}^{1/2}\,|0\rangle\,|LM\rangle + \sum_{\substack{k\lambda\mu\\ jm}} \beta_{k\lambda j} C^{LM}_{jm,\lambda\mu} b^{\dagger}_{k\lambda\mu}\,|0\rangle\,|jm\rangle$$

# Angulon spectral function

Let us use the theory! The plan is simple:

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

Second order:

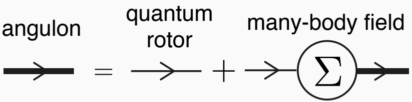


# Angulon spectral function

Let us use the theory! The plan is simple:

1. Self-energy ($\Sigma$)
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 theory! The plan is simple:

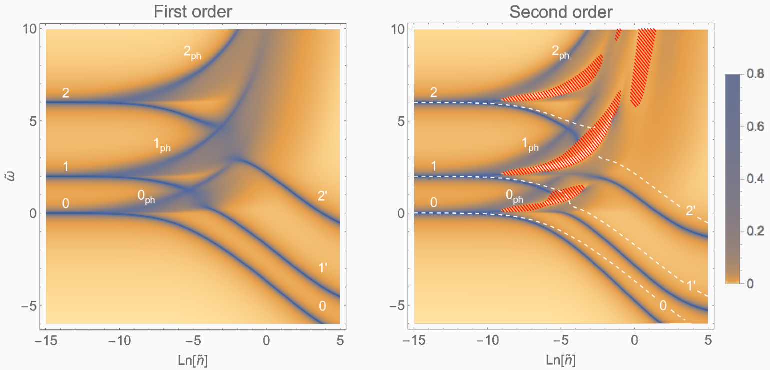
1. Self-energy ($\Sigma$)
2. Dyson equation to obtain the angulon Green's function (G)
3. Spectral function ($\mathcal{A}$)

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

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

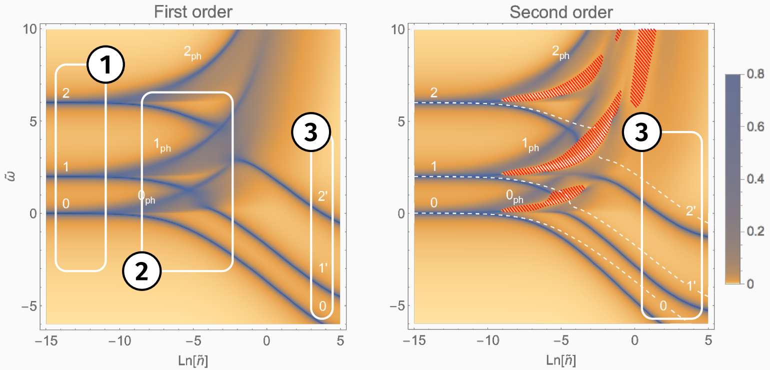
# Angulon quasiparticle spectrum

Angulon quasiparticle spectrum as a function of the density:



# Angulon quasiparticle spectrum

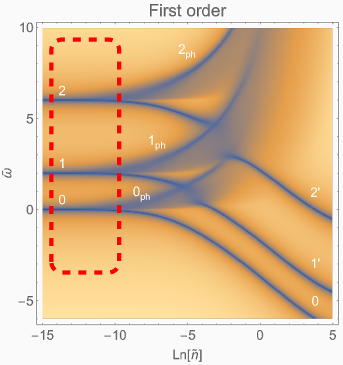
Angulon quasiparticle spectrum as a function of the density:




**Key features**:

1. Low density
2. Intermediate instability
3. High density

# Angulon quasiparticle spectrum: low density

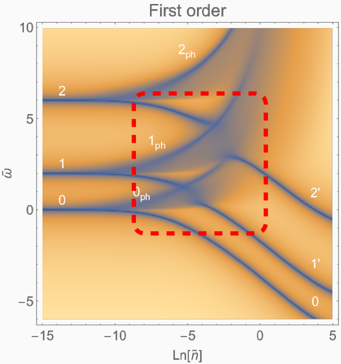



Density range: from ultra-cold atoms to superfluid helium.

Low density: free rotor spectrum, $E \sim L(L+1)$.

Many-body-induced fine structure

# Angulon quasiparticle spectrum: instability

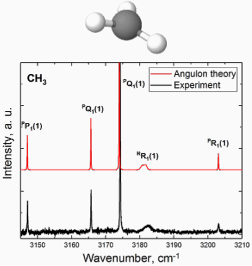



Intermediate region: angulon instability.

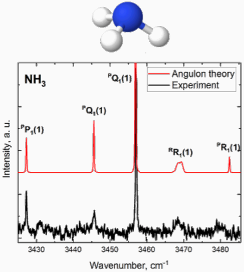
Corresponding to the emission of a phonon with $\lambda = 1$.

# Experimental observation of angulon instabilities?

I. N. Cherepanov, M. Lemeshko, *"Fingerprints of angulon instabilities in the spectra of matrix-isolated molecules"*, arXiv:1705.09220.

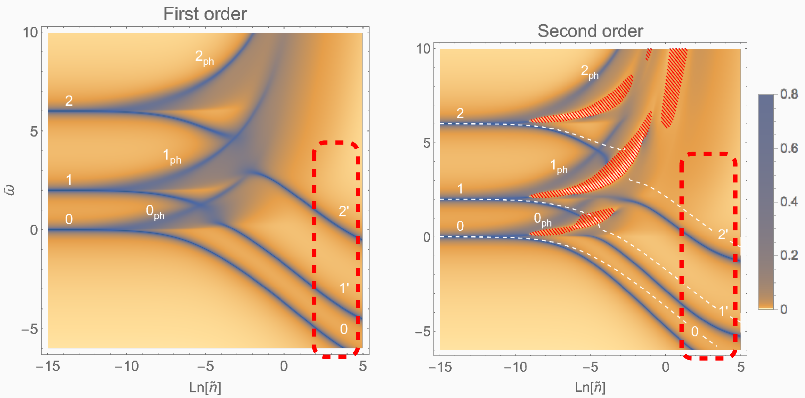



Douberly group, J. Phys. Chem. A **117**, 11640 (2013)




Vilesov group, Chem. Phys. Lett. **412**, 176 (2005).

# Angulon quasiparticle spectrum: high density

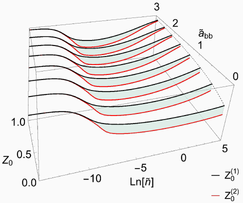


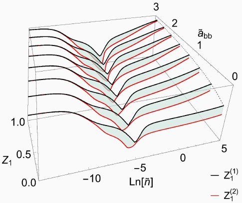

High density: the two-loop corrections start to be relevant.

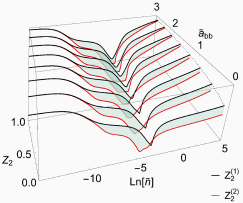
# Quasiparticle weight

Location of the quasiparticle pole

$$Z_\lambda = \frac{1}{1 - \left.\frac{\partial \operatorname{Re} \Sigma_\lambda(E)}{\partial E}\right|_{E=E_P}}$$



# Conclusions

- The problem of angular momentum redistribution in a many-body environment has been treated through the path integral formalism and reformulated in terms of diagrams.
- It allows for a simple, compact derivation of angulon properties, including higher order terms.
- It can be extended, to include e.g. the angulon-angulon interaction, or the interaction with external fields.
- It connects the angulon theory with advanced diagrammatic techniques (higher orders, different summation schemes, Diagrammatic Monte Carlo).


Conference on Controllable Quantum Impurities in Physics and Chemistry (CoQIPC 2017).

August 16-18, 2017, at IST Austria, Klosterneuburg, near Vienna.

# Thank you for your attention.

Der Wissenschaftsfonds.

This work was supported by the Austrian Science Fund (FWF), project Nr. P29902-N27.

# Backup slide # 1

# Backup slide # 2

# Backup slide # 3