Quasiparticle approach to far-from-equilibrium dynamics of molecules in helium nanodroplets

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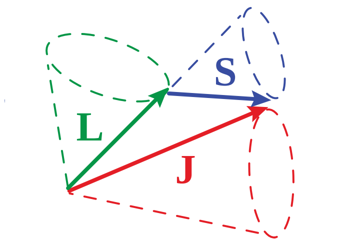
Institute of Science and Technology (IST) Austria

Quantum Fluid Clusters

May 20, 2019

The problem

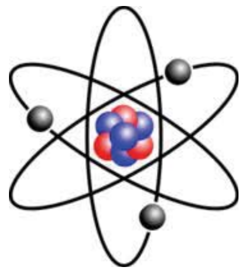


$J = |L-S| \ldots |L+S|$

Quantum angular momentum

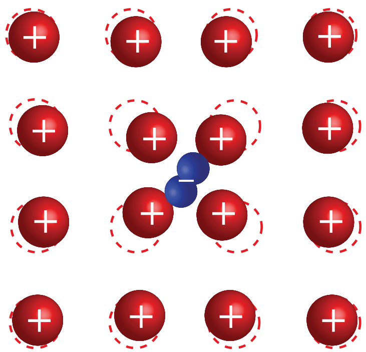
Small systems: extremely challenging

(electrons in an atom)



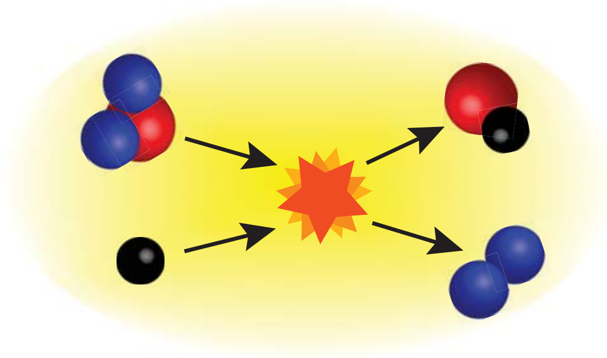
Condensed-matter systems: is it possible at all?

Electrons in solids



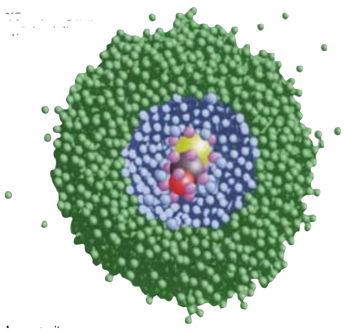
Ultrafast magnetism, Data storage, Spintronics, Quantum computation, …

Molecules in solvents



Controlling chemical reactions, Catalysis, Energy research, …

Initial motivation:
molecules in superfluid helium nanodroplets



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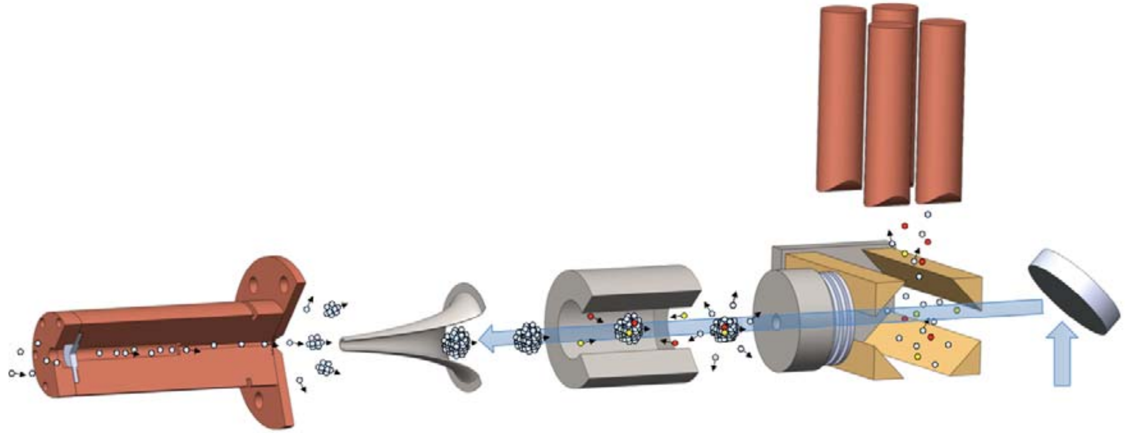


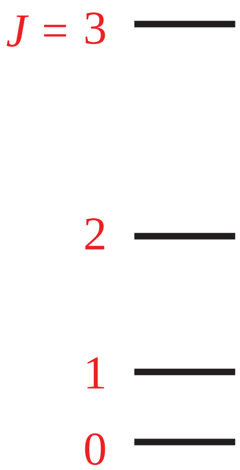
Andrew M. Ellis webpage

Reasons people do it:

- **Spectroscopy (0.4 K, no doppler shift)**
- **Studying unstable species (radicals)**

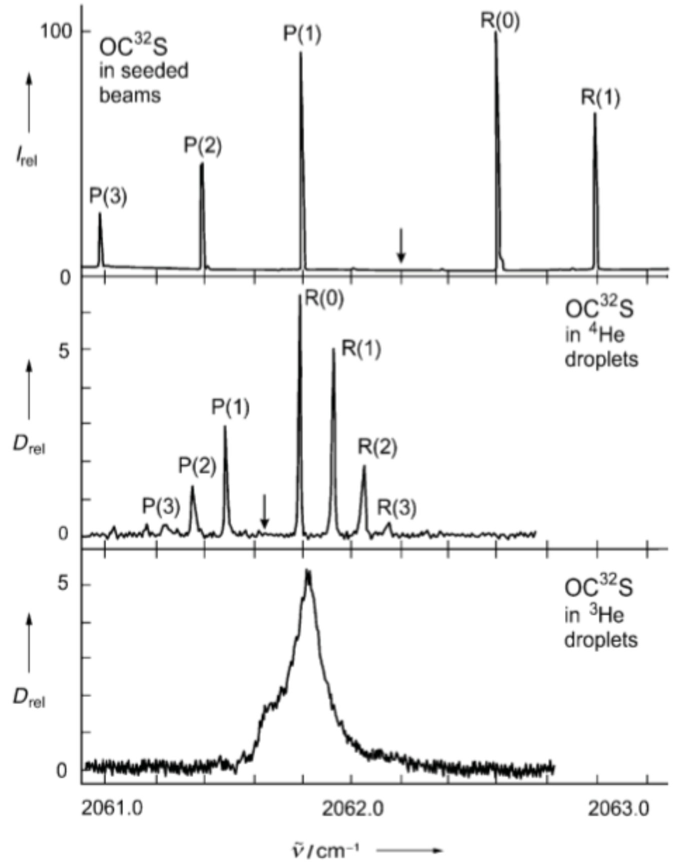
Initial motivation:
molecules in superfluid helium nanodroplets

"Clean" rotational spectra,
but renormalized rotational constant



$$E_J = BJ(J+1)$$

$$B = \frac{\hbar^2}{2I}$$



Free

in ^{4}He

in ^{3}He

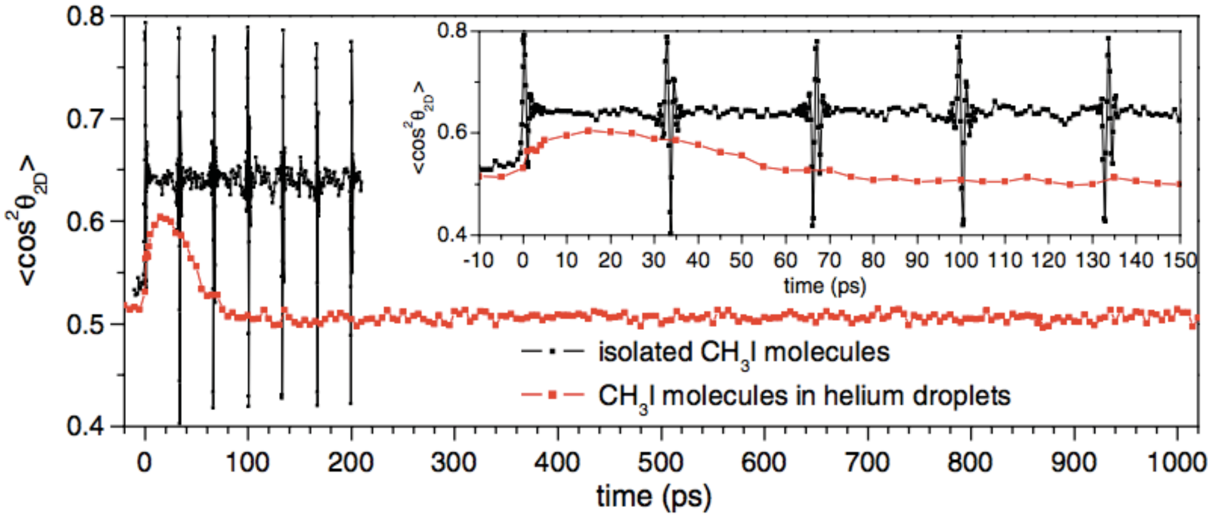
There are qualitative explanations (two-fluid model, etc.),
Quantum Monte Carlo calculations for several molecules (Zillich, Whaley, …)
However, no general microscopic understanding

Initial motivation:
molecules in superfluid helium nanodroplets

Dynamics of molecules in droplets:

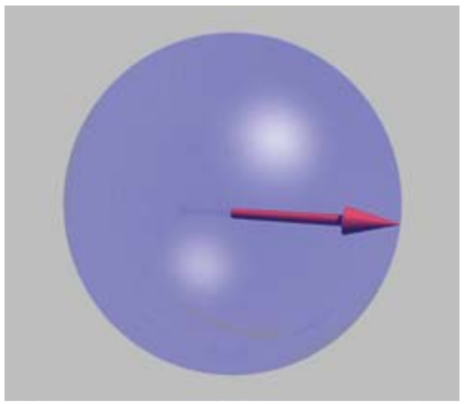
even qualitative understanding was absent

Revivals of rotational wavepackets



Stapelfeldt group, PRL **110**, 093002 (2013)

Bloch sphere analogy

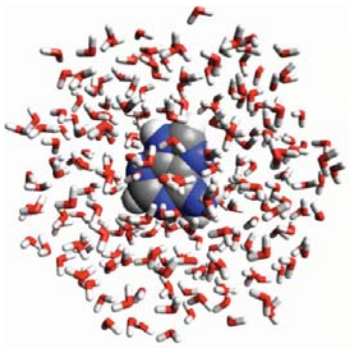


Create $|\uparrow\rangle + |\downarrow\rangle$

Measure $\sigma_x(t)$

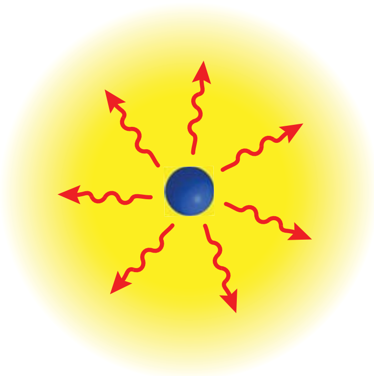
Molecule in He droplet
as a quantum impurity problem

Molecules in solution

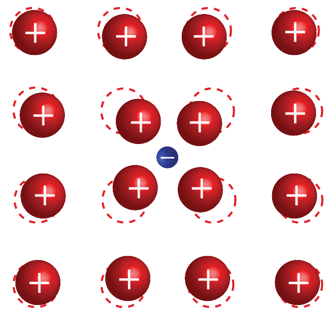


Molecular Modelling Basics (2010)

Atoms in BEC



Electrons in solids



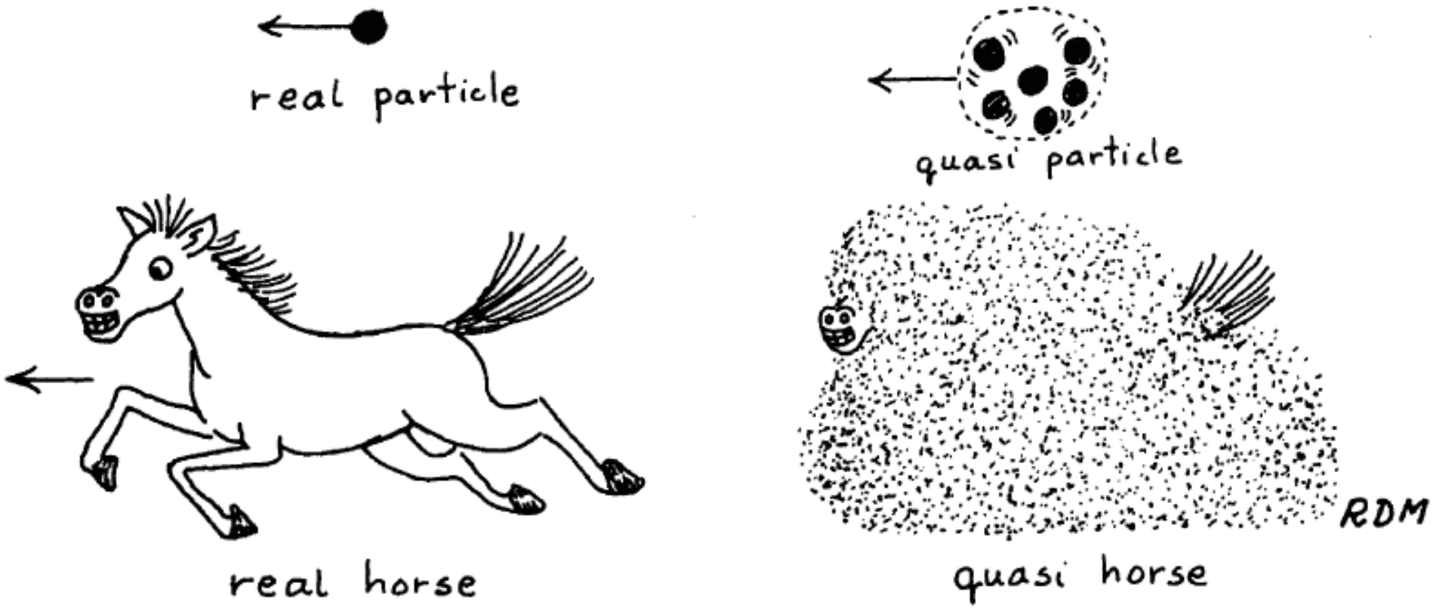
Impurity problems: 1 particle + its many-body environment

Still ~10^{23} degrees of freedom – challenging to understand

A physicist's trick: introducing "quasiparticles"

Strongly interacting system of real particles

→ Almost free motion of imaginary quasiparticles

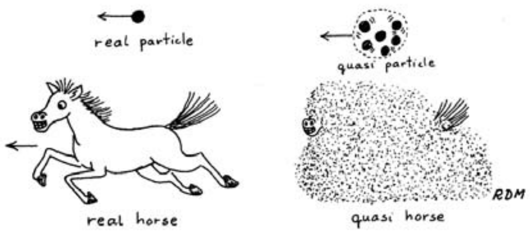


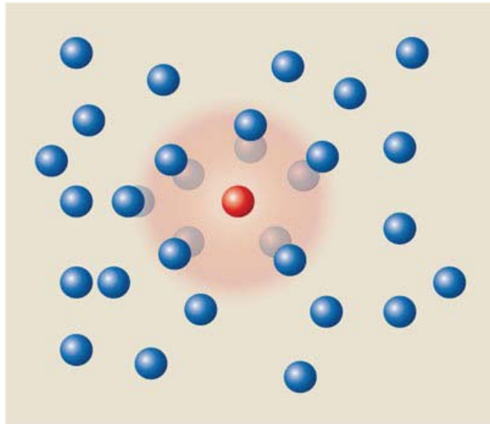
R. Mattuck, "A Guide to Feynman Diagrams in the Many-body Problem"

Quasiparticles: examples

Polaron:

an impurity whose linear motion
is "dressed" by a cloud of excitations

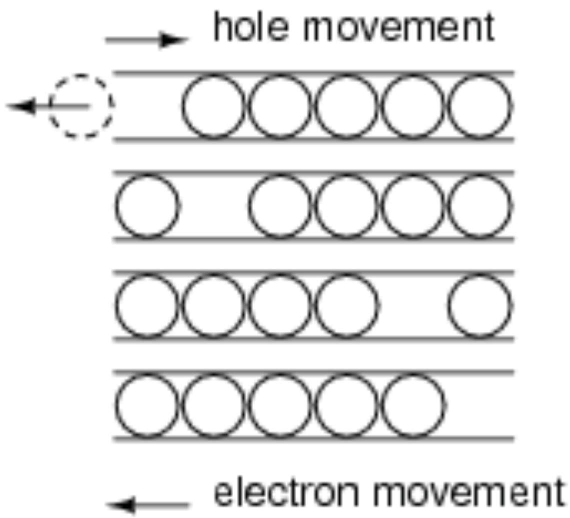




Nature **485**, 588–589

Hole:

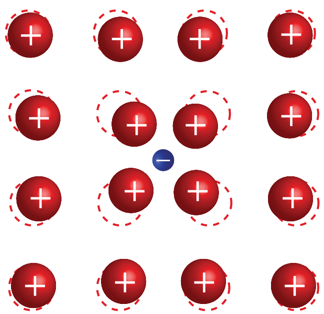
an empty spot in a sea of electrons



http://www.allaboutcircuits.com

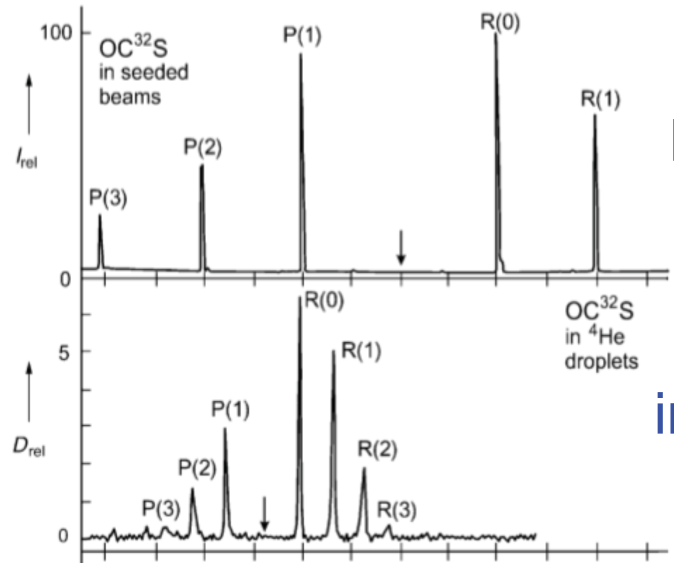
Can molecules in superfluids be described as quasiparticles?

Electrons in solids: effective mass



Polaron quasiparticle

Molecules in He: effective rotational constant



Free

in ^{4}He

Angew. Chem. Int. Ed. **43,** 2622 (2004)

‘Angulon’ quasiparticle?

Why do we need another one?

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List of quasiparticles

From Wikipedia, the free encyclopedia

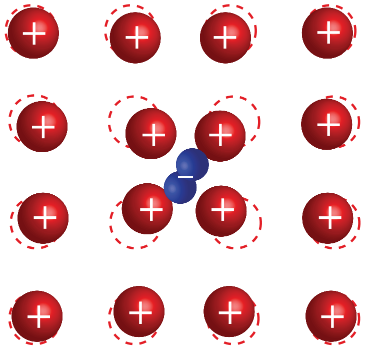
This is a **list of quasiparticles**.

Quasiparticles

Quasiparticle	Signification	Underlying particles
Bipolaron	A bound pair of two polarons	polaron (electron, phonon)
Bogoliubon	Broken Cooper pair	electron
Configuron[1]	An elementary configurational excitation in an amorphous material which involves breaking of a chemical bond	
Dropleton	The first known quasiparticle that behaves like a liquid[2]	
Electron quasiparticle	An electron as affected by the other forces and interactions in the solid	electron
Electron hole (hole)	A *lack* of electron in a valence band	electron, cation
Exciton	A bound state of an electron and a hole	electron, hole
Fracton	A collective quantized vibration on a substrate with a fractal structure.	
Holon (chargon)	A quasi-particle resulting from electron spin-charge separation	
Leviton	A collective excitation of a single electron within a metal	
Magnon	A coherent excitation of electron spins in a material	
Majorana fermion	A quasiparticle equal to its own antiparticle, emerging as a midgap state in certain superconductors	
Orbiton[3]	A quasiparticle resulting from electron spin-orbital separation	
Phason	Vibrational modes in a quasicrystal associated with atomic rearrangements	
Phoniton	A theoretical quasiparticle which is a hybridization of a localized, long-living phonon and a matter excitation[4]	
Phonon	Vibrational modes in a crystal lattice associated with atomic shifts	
Plasmaron	A quasiparticle emerging from the coupling between a plasmon and a hole	
Plasmon	A coherent excitation of a plasma	
Polaron	A moving charged quasiparticle that is surrounded by ions in a material	electron, phonon
Polariton	A mixture of photon with other quasiparticles	photon, optical phonon
Roton	Elementary excitation in superfluid helium-4	
Soliton	A self-reinforcing solitary excitation wave	
Spinon	A quasiparticle produced as a result of electron spin-charge separation that can form both quantum spin liquid and strongly correlated quantum spin liquid	
Trion	A coherent excitation of three quasiparticles (two holes and one electron or two electrons and one hole)	
Wrinklon	A localized excitation corresponding to wrinkles in a constrained two dimensional system[5][6]	

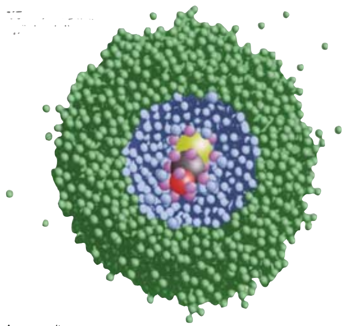
It'd describe (in principle) any many-body system with angular momentum

Electrons in solids



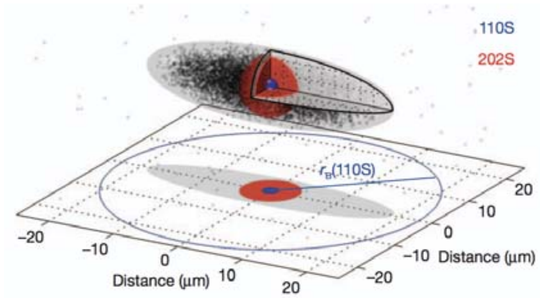
(Einstein-de Haas effect)

Chemistry in solvents



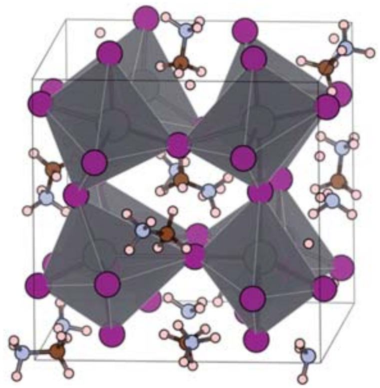
Angew. Chem. Int. Ed. **43,** 2622 (2004)

Rydberg atoms / cold molecules in a BEC / Fermi gas



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

Hybrid organic/inorganic perovskites

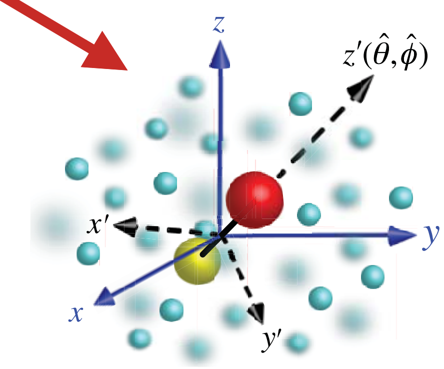


Bakulin *et al.*, J. Phys. Chem. Lett. **6**, 3663 (2015)

The angulon Hamiltonian

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

molecule **phonons** **molecule-phonon interactions**



- Was derived exactly for a molecule in a weakly-interacting superfluid (BEC)

R. Schmidt and ML, Phys. Rev. Lett. **114**, 203001 (2015)
R. Schmidt and ML, Phys. Rev. X **6**, 011012 (2016)

- Can be used as a phenomenological model for any bosonic bath

ML, Phys. Rev. Lett. **118**, 095301 (2017)

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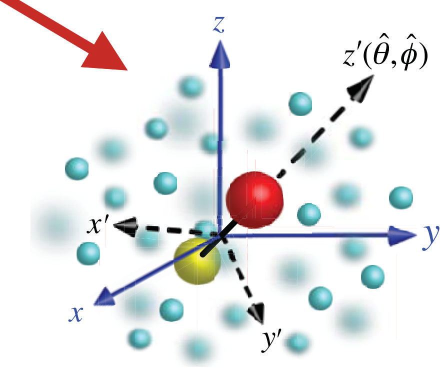
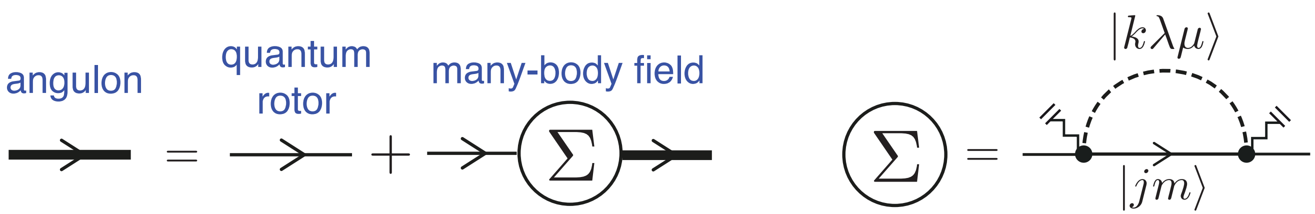
$$\hat{H} = B\hat{\mathbf{J}}^2 + \sum_{k\lambda\mu} \omega_k \hat{b}^\dagger_{k\lambda\mu}\hat{b}_{k\lambda\mu} + \sum_{k\lambda\mu} U_\lambda(k) \left[Y_{\lambda\mu}(\hat{\theta},\hat{\phi})\hat{b}_{k\lambda\mu} + Y^*_{\lambda\mu}(\hat{\theta},\hat{\phi})\hat{b}^\dagger_{k\lambda\mu} \right]$$

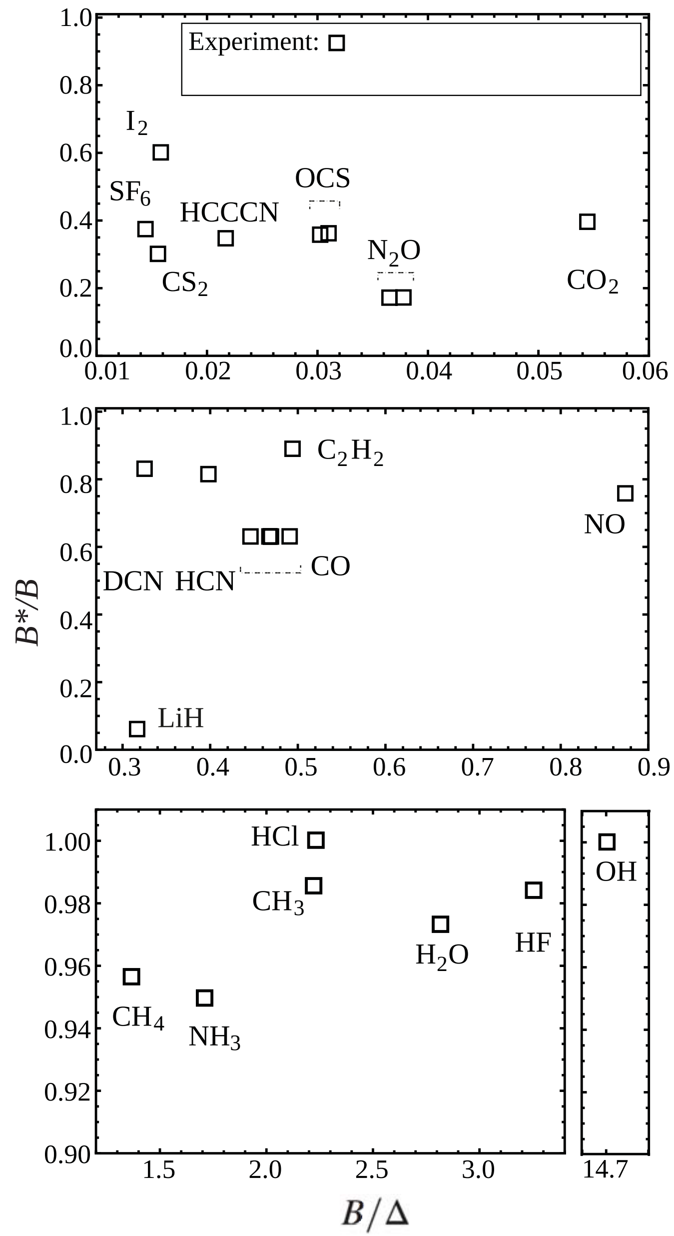
molecule — **phonons** — **molecule-phonon interactions**



The “angulon” quasiparticle



Existence of angulons



ML, Phys. Rev. Lett. **118**, 095301 (2017) *Viewpoint: Yu. Shchadilova, Physics* ***10****, 20 (2017)*

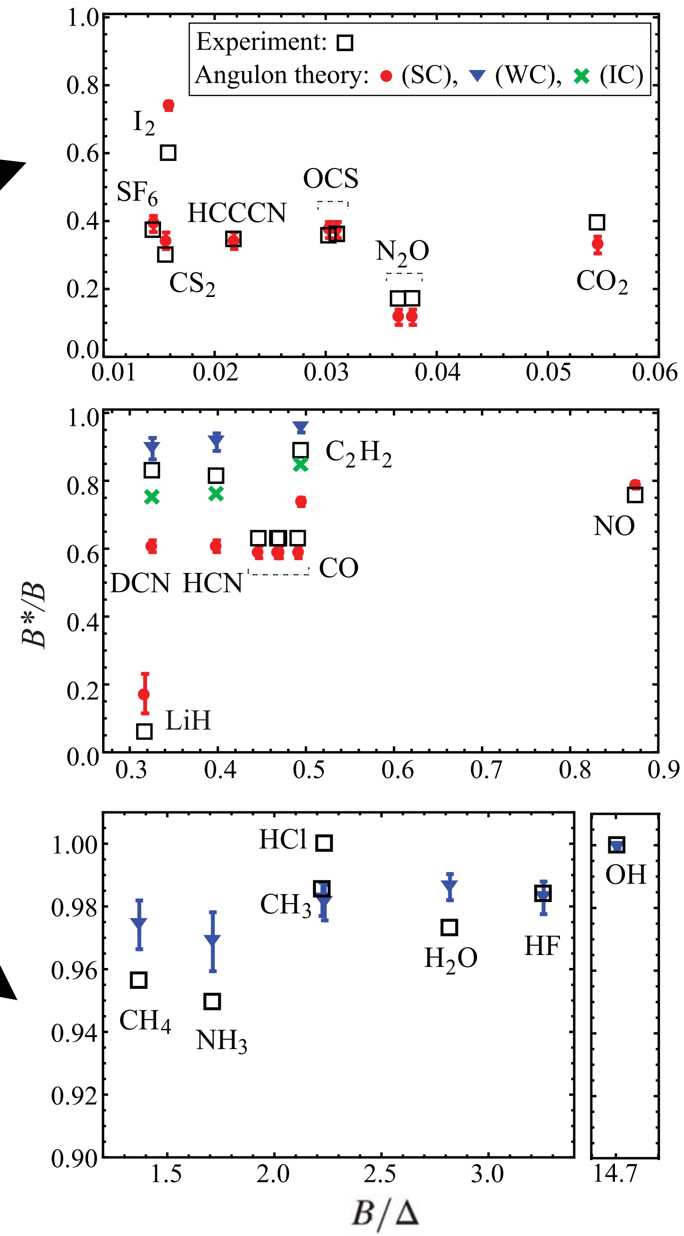
Existence of angulons

Heavy molecules: strong-coupling
(real deformation of the superfluid)

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

Light molecules: weak-coupling
(rotational Lamb shift)

R. Schmidt and ML, Phys. Rev. Lett. **114**, 203001 (2015)

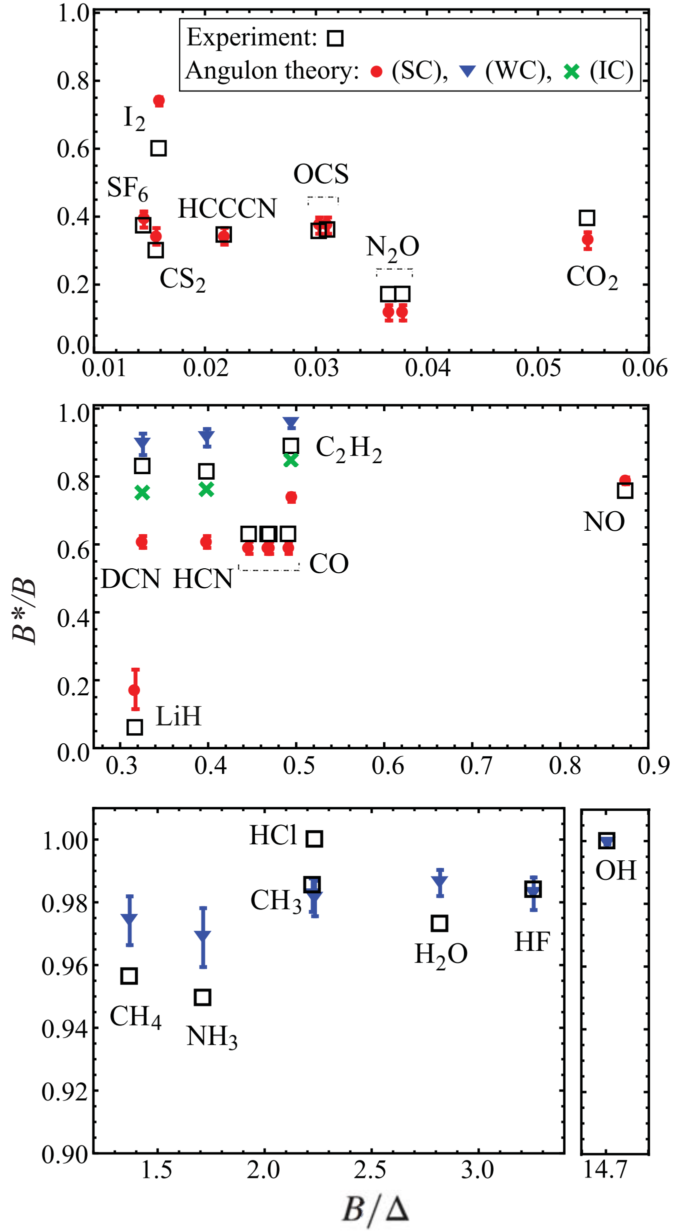


ML, Phys. Rev. Lett. **118**, 095301 (2017)   *Viewpoint: Yu. Shchadilova, Physics **10**, 20 (2017)*

Existence of angulons

$$\frac{B^*_{\mathrm{SC}}}{B} = (1 - \eta\,\Delta)^2 \qquad \frac{B^*_{\mathrm{WC}}}{B} = 1 - \xi \frac{\Delta^2}{B}$$

(instead of expensive MC computations)



ML, Phys. Rev. Lett. **118**, 095301 (2017) *Viewpoint: Yu. Shchadilova, Physics **10**, 20 (2017)*

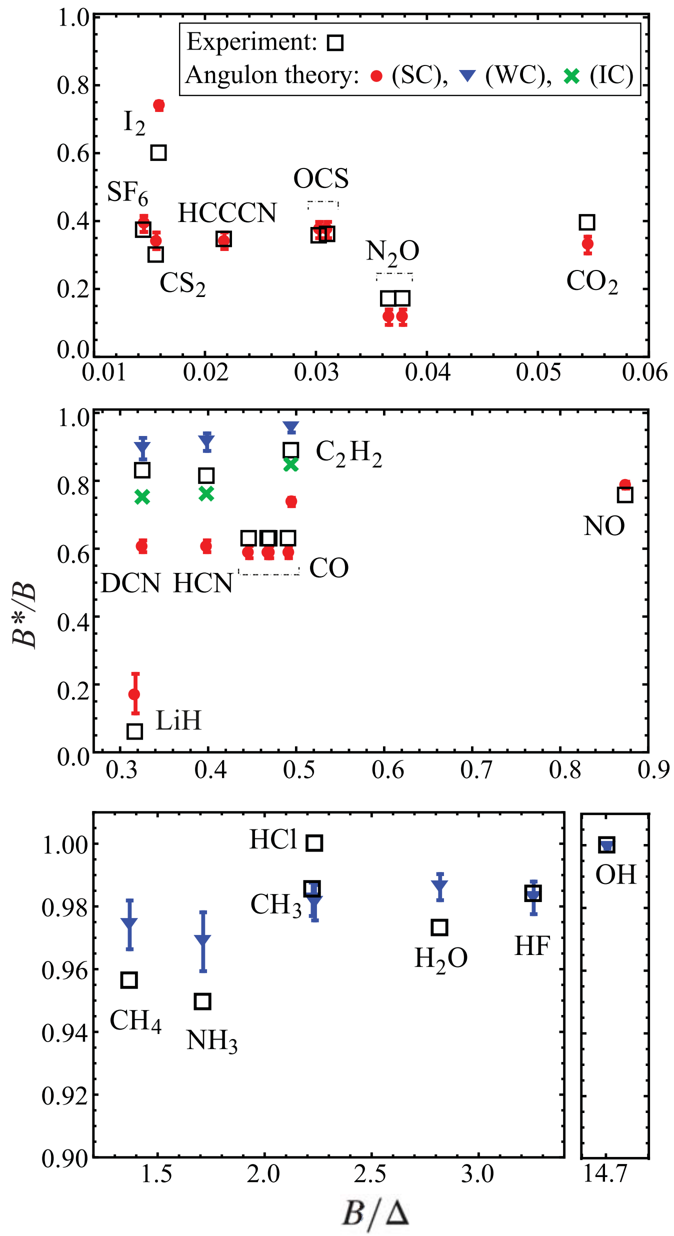
Existence of angulons

anisotropy of molecule-He potential (|| minus ⊥)

$$\frac{B^*_{\rm SC}}{B} = (1 - \eta\,\Delta)^2 \qquad \frac{B^*_{\rm WC}}{B} = 1 - \xi \frac{\Delta^2}{B}$$

phenomenological parameters

(expressed through He properties, same for all molecules)



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Existence of angulons

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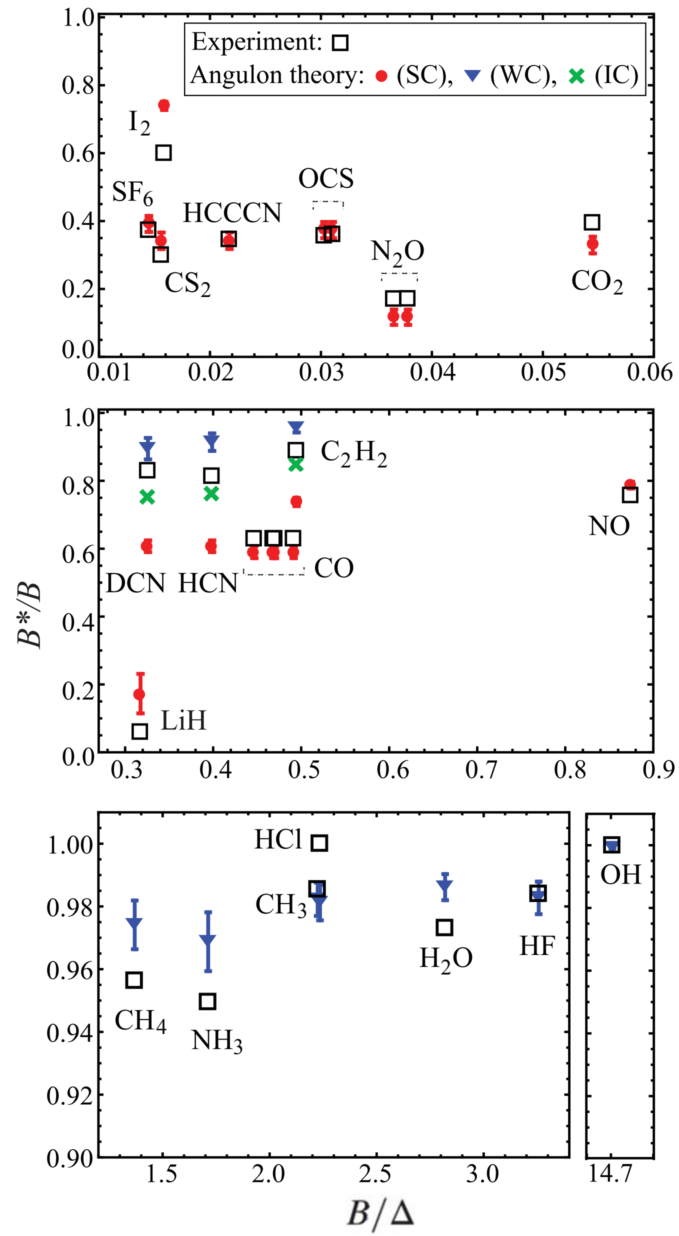
phenomenological parameters

(expressed through He properties, same for all molecules)

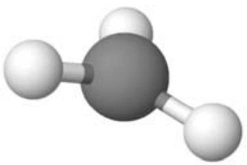
Strong evidence that angulons are formed in experiment



ML, Phys. Rev. Lett. **118**, 095301 (2017) *Viewpoint: Yu. Shchadilova, Physics* ***10****, 20 (2017)*

Angulon instabilities

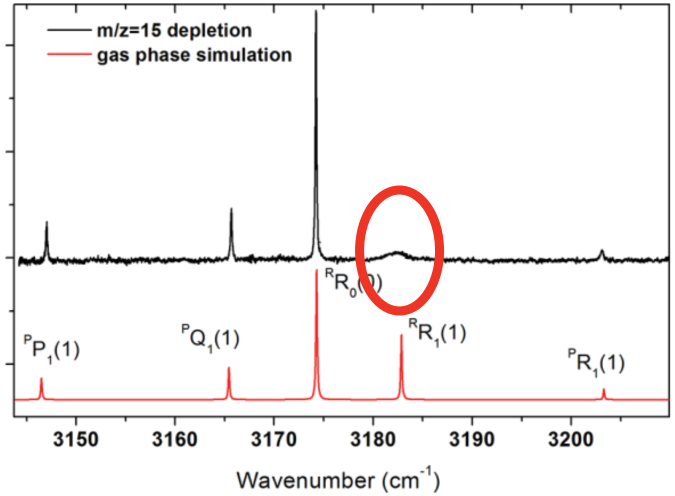
Explain ‘anomalous broadening’ in molecular spectra in He droplets



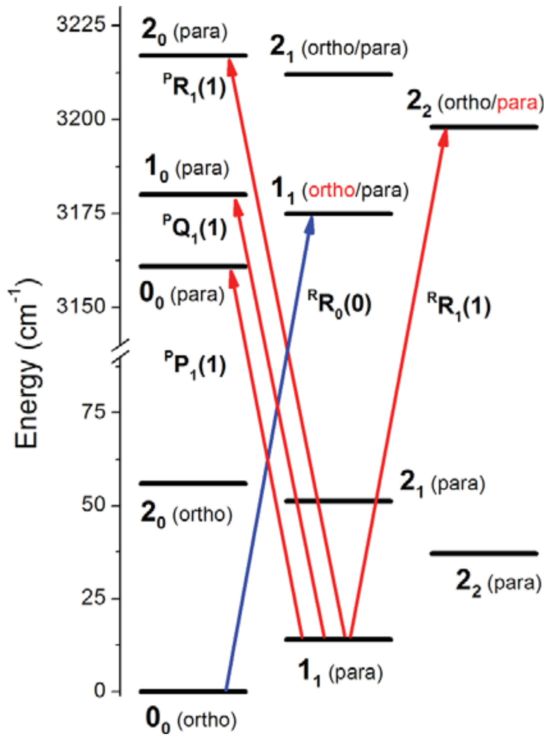
CH_3

He droplets →

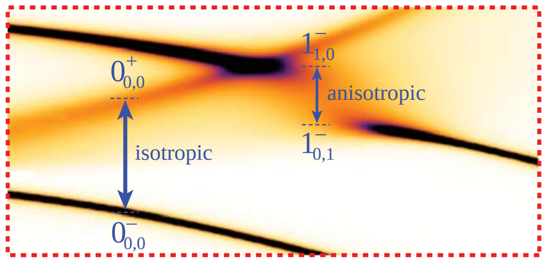
gas phase →



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



Are those the angulon instabilities?

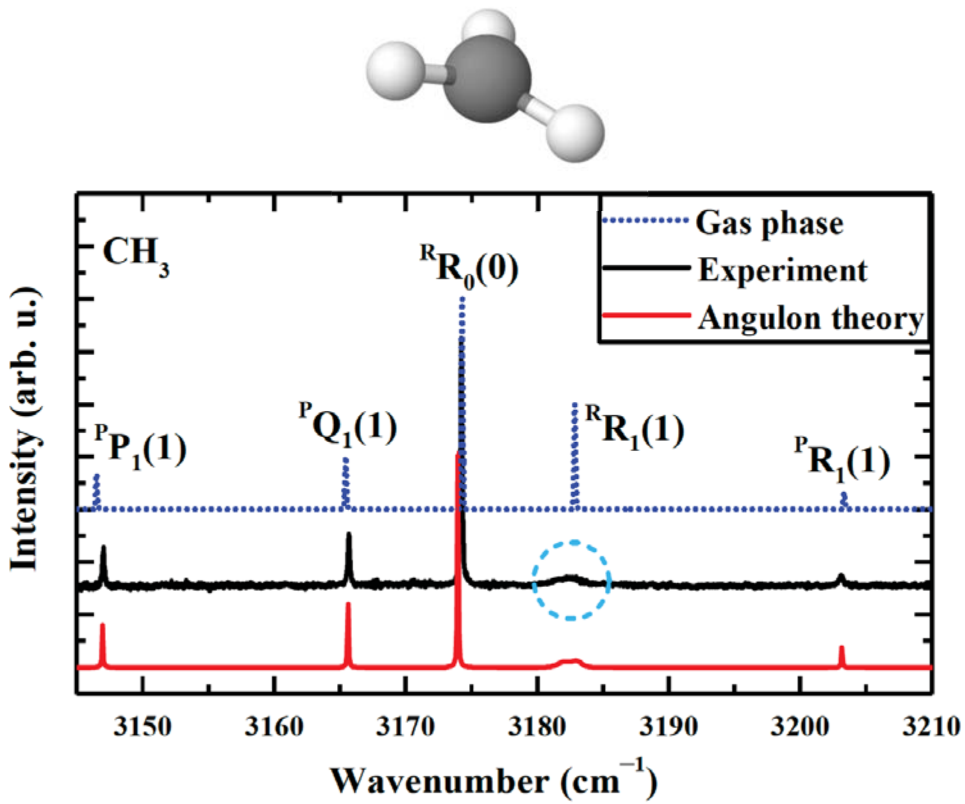


Angulon instabilities

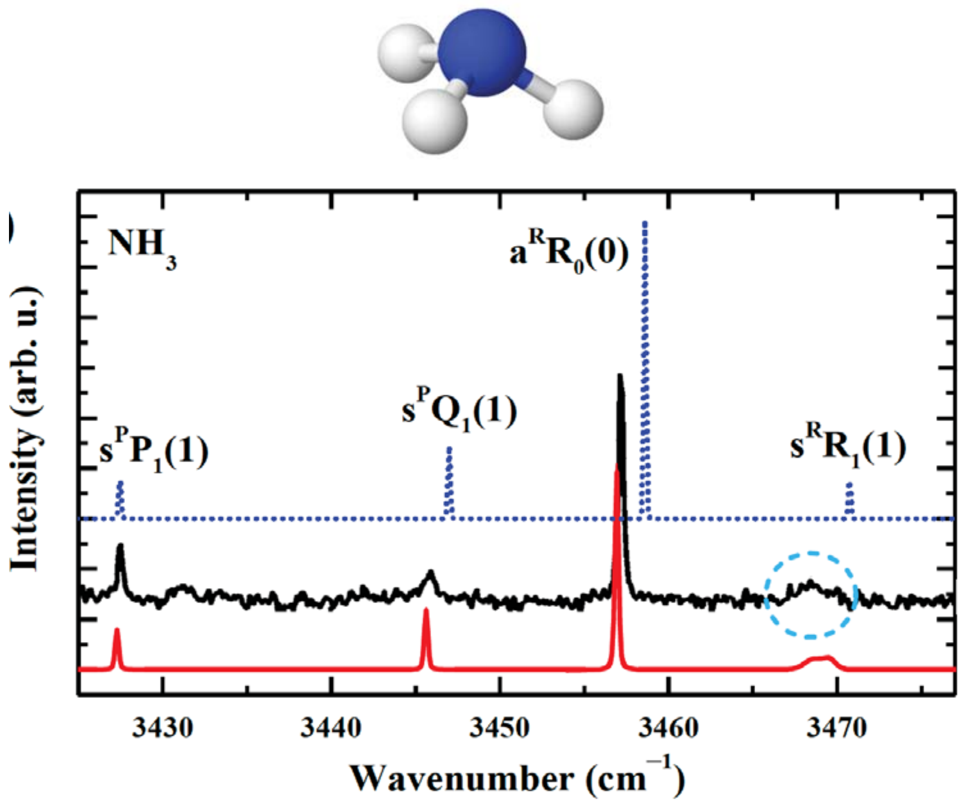
We have developed a theory for symmetric-top angulon

I. Cherepanov and ML, Phys. Rev. Materials **1**, 035602 (2017)

Igor Cherepanov



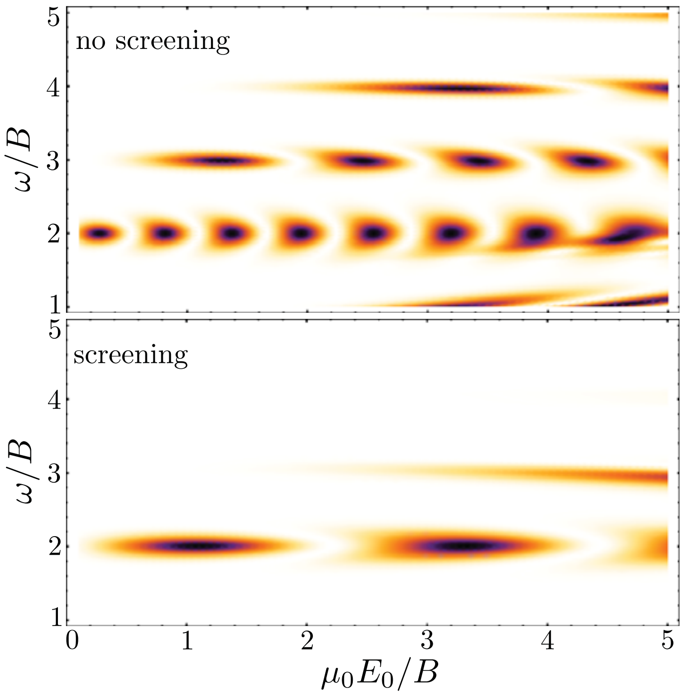
Douberly group, J. Phys. Chem. A **117**, 11640 (2013)



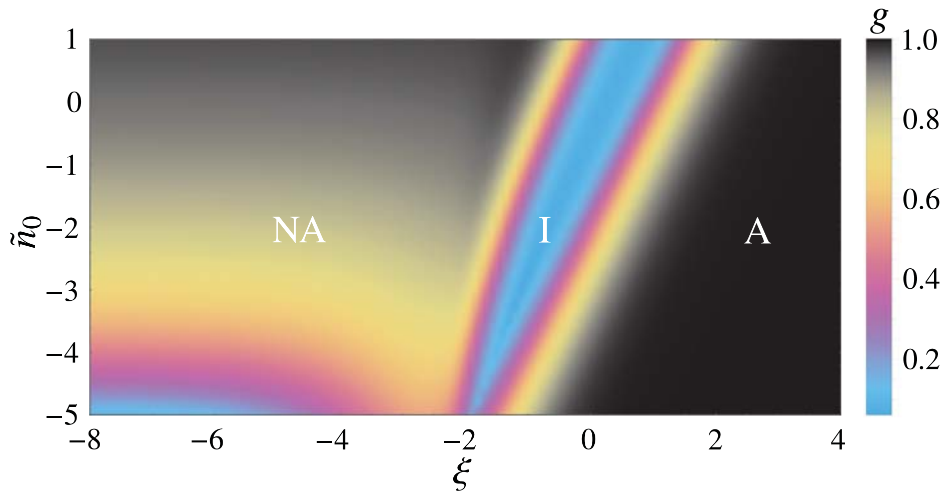
Vilesov group, Chem. Phys. Lett. **412**, 176 (2005)

New emergent phenomena

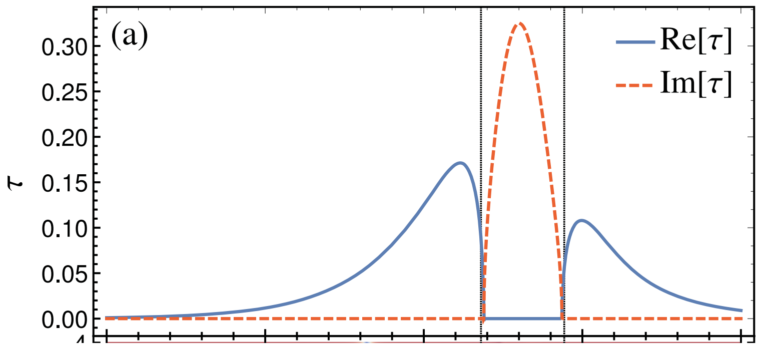
‘Anomalous screening’



Non-Abelian magnetic monopoles



Quantum groups



Enderalp Yakaboylu

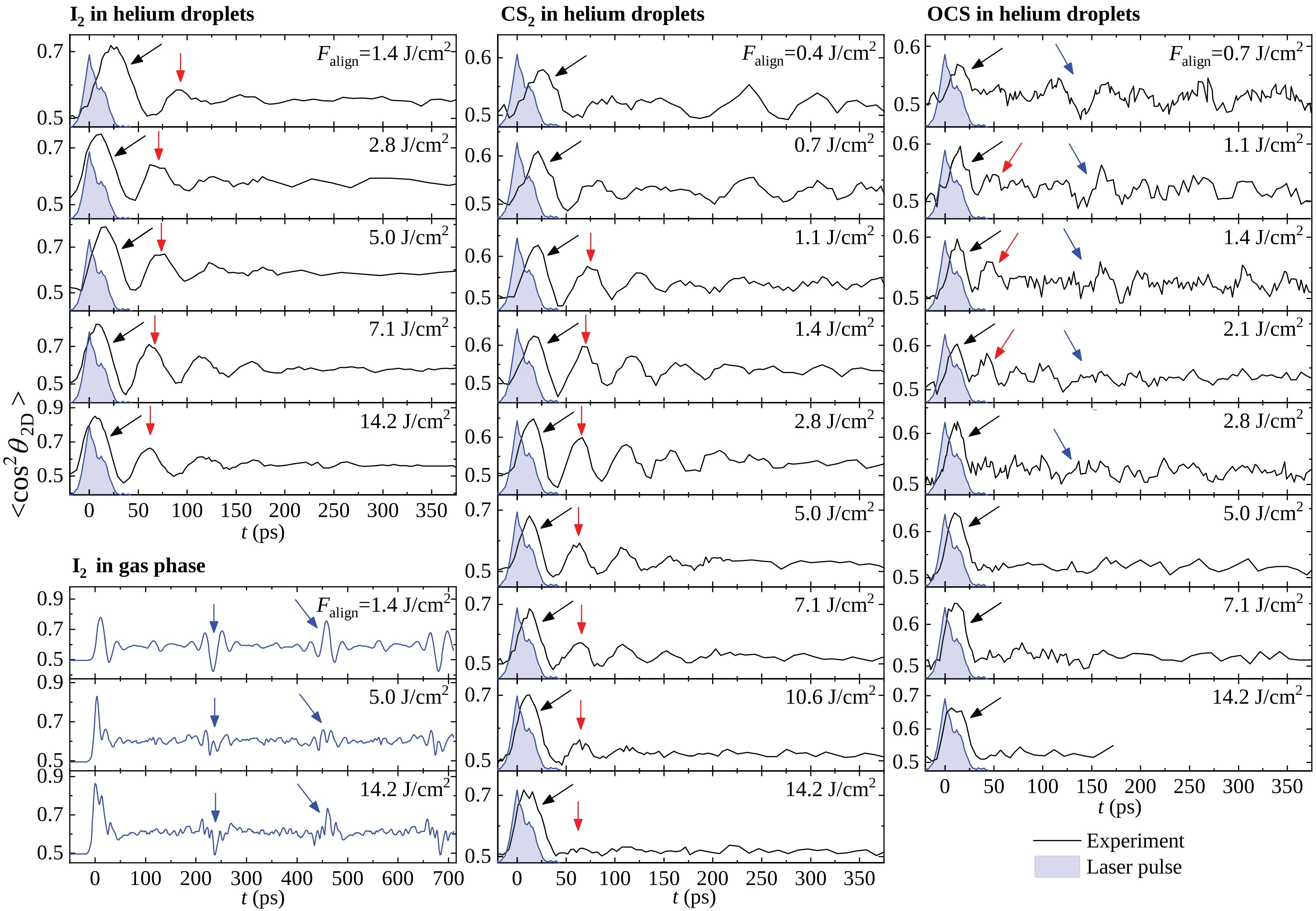
E. Yakaboylu, M. Shkolnikov, and ML, Phys. Rev. Lett., **121**, 255302 (2018)

E. Yakaboylu and ML, Phys. Rev. Lett. **118**, 085302 (2017)

E. Yakaboylu, A. Deuchert, and ML, Phys. Rev. Lett. **119**, 235301 (2017)

Far from equilibrium dynamics
of molecules in superfluid helium

Far from equilibrium dynamics of molecules in helium



Igor Cherepanov

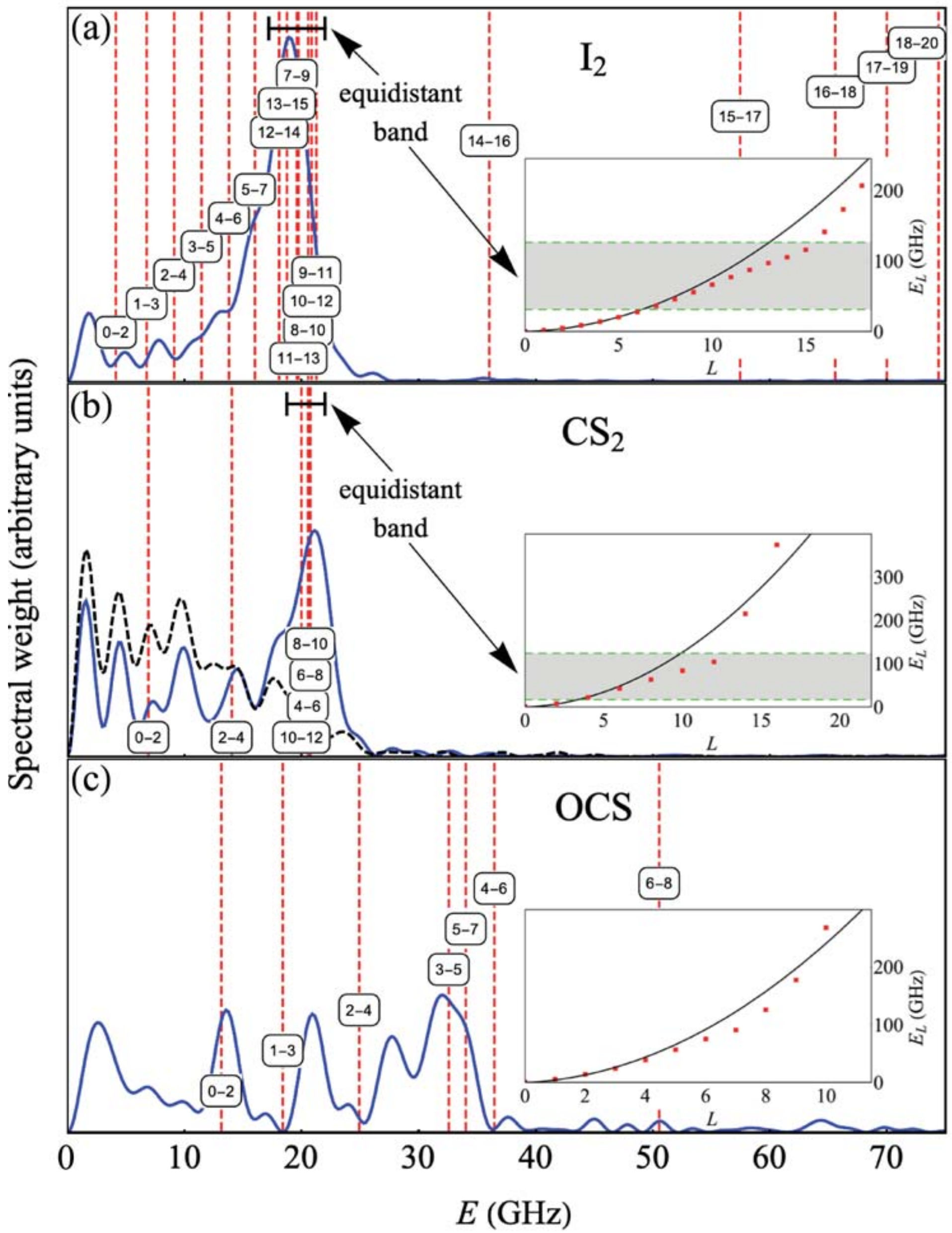
Giacomo Bighin

Posters

I. Cherepanov, G. Bighin, L. Christiansen, A.V. Jørgensen, R. Schmidt, H. Stapelfeldt, ML, submitted (2019)
(also see PRL **118**, 203203 (2017))

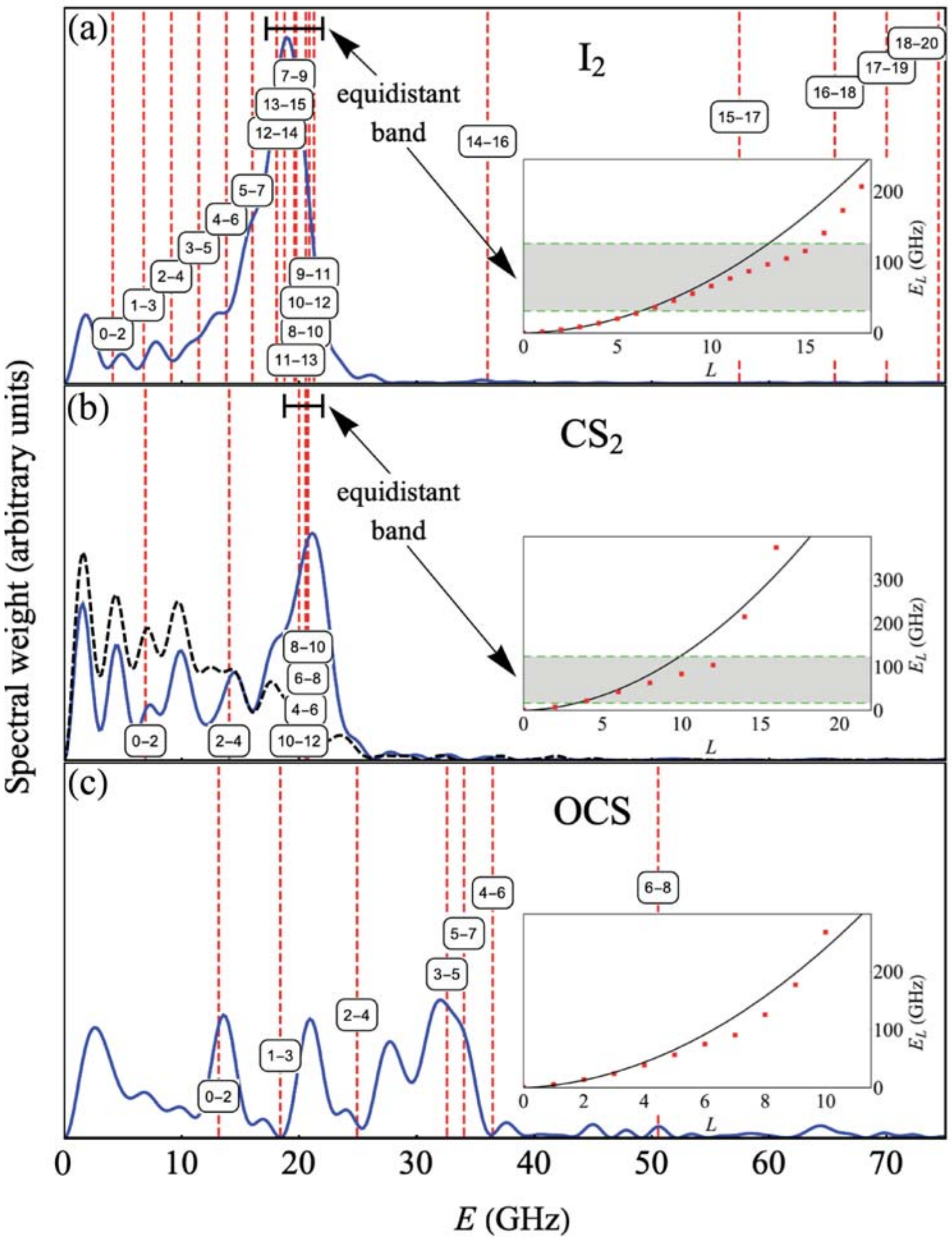
What is the physics behind it?

1. “Equidistant band” of states

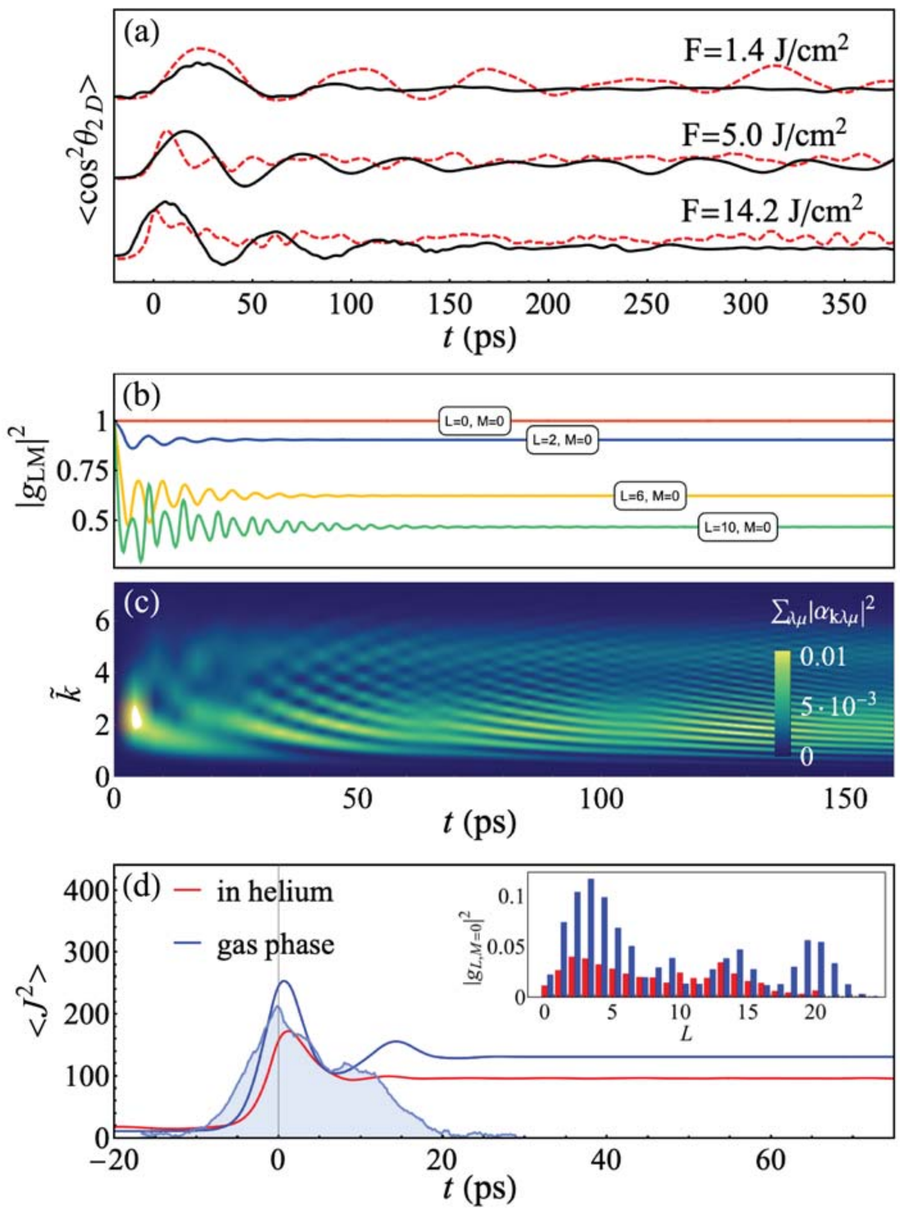


What is the physics behind it?

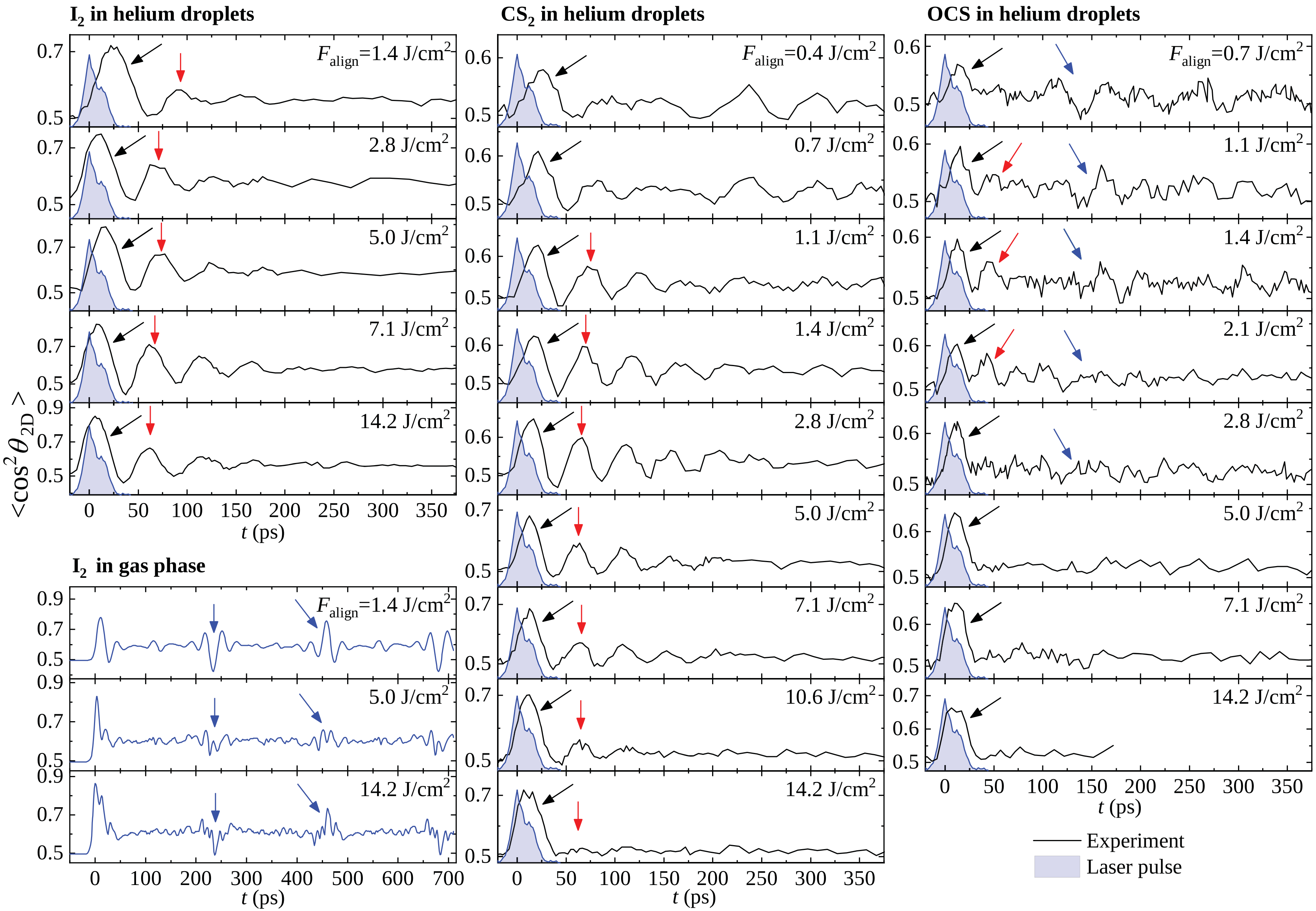
1. “Equidistant band” of states



2. Dynamical transfer of angular momentum



Far from equilibrium dynamics of molecules in helium



Igor Cherepanov

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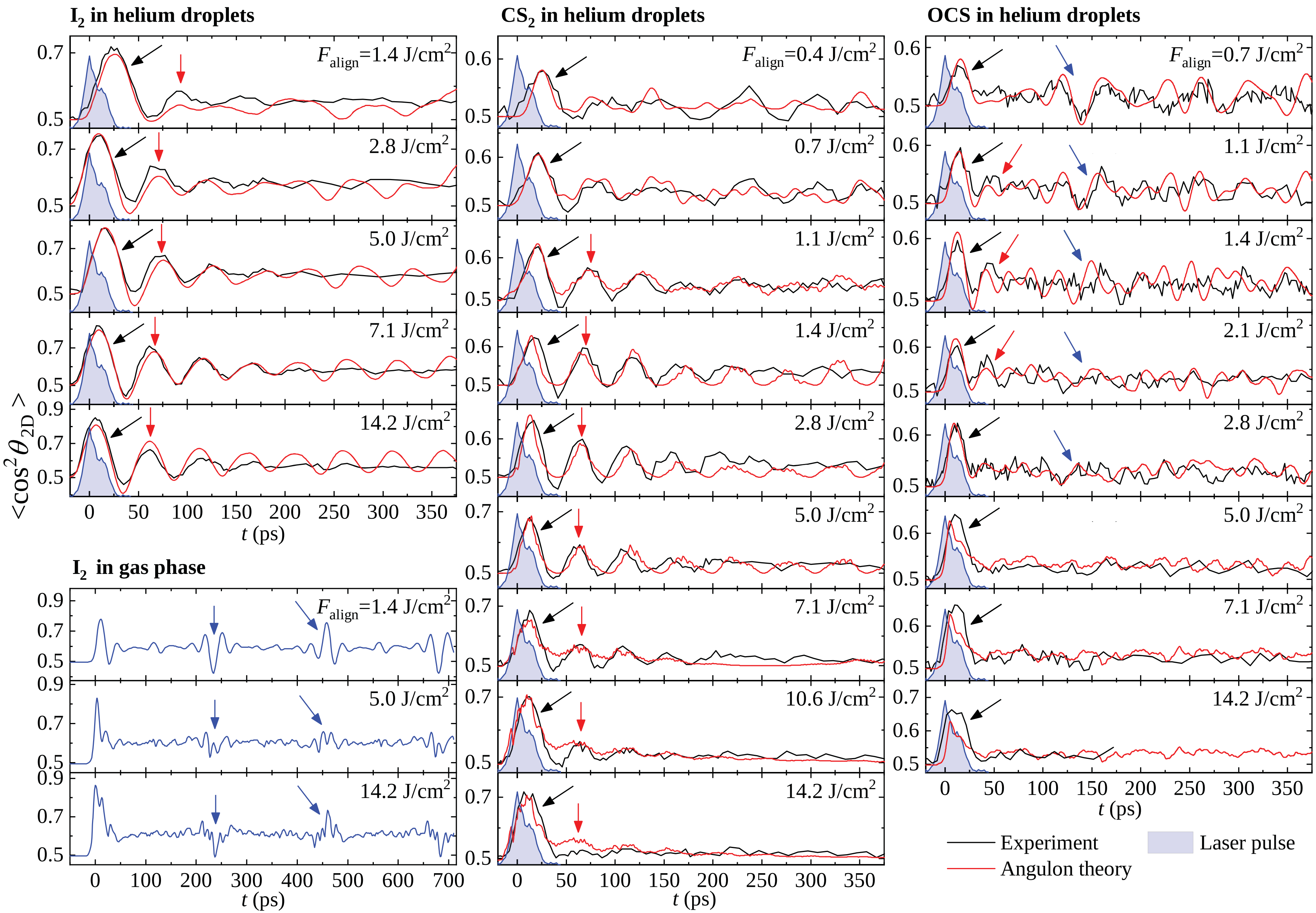
I. Cherepanov, G. Bighin, L. Christiansen, A.V. Jørgensen, R. Schmidt, H. Stapelfeldt, ML, submitted (2019)
(also see PRL **118**, 203203 (2017))

Far from equilibrium dynamics of molecules in helium



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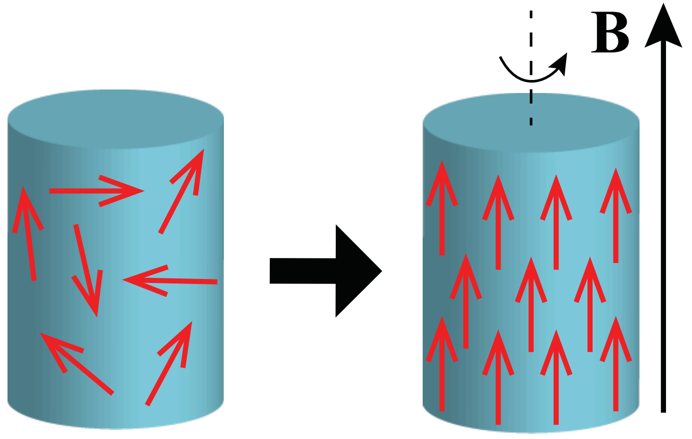
I. Cherepanov, G. Bighin, L. Christiansen, A.V. Jørgensen, R. Schmidt, H. Stapelfeldt, ML, submitted (2019)

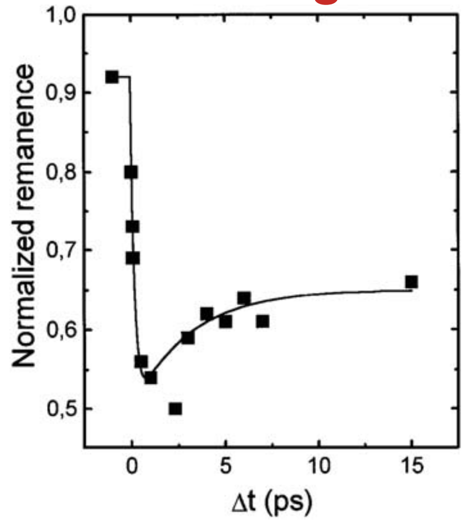
(also see PRL **118**, 203203 (2017))

Angulons in 'real' solid state systems

Einstein-de Haas effect (1915)



Ultrafast magnetism



Beaurepaire et al., PRL 1996

Spin-phonon relaxation and phonon spin
Chudnovskii, Garanin PRL 2005; Niu, Zhang, PRL 2014; Garanin & Chudnovskii PRB 2015

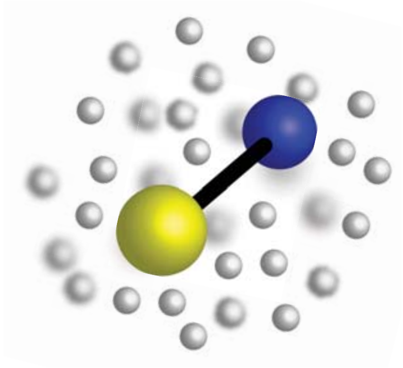
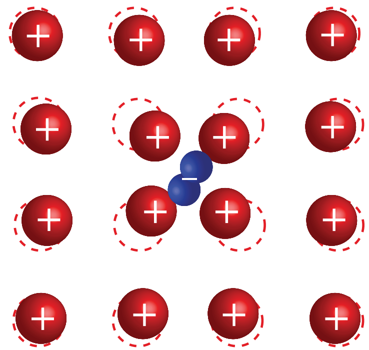
Nano-magneto-mechanical systems
Wernsdorfer group Nature Comm. 2016

Spin mechatronics
Matsuo, Saitoh, and Maekawa Frontiers of physics 2015

…and many many more

RU Nijmegen: Johan Mentink & Mikhail Katsnelson

Angulons in 'real' solid state systems

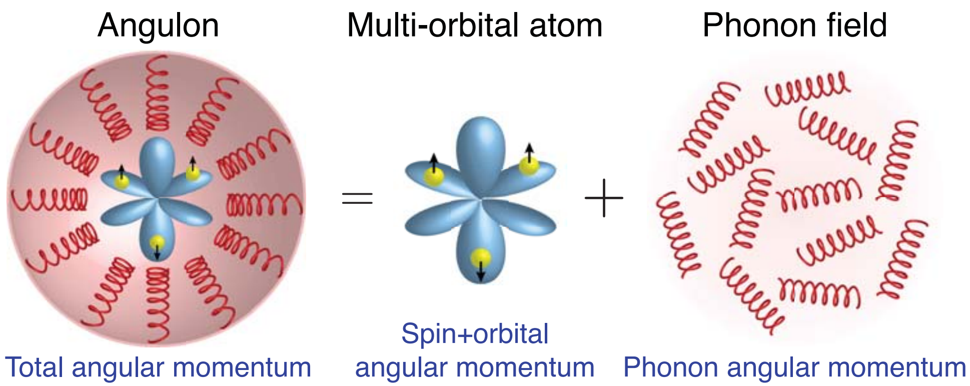
Molecule + helium		Electron + phonons
	⟷	
“Easy”		**“Hard”**
Rotational angular momentum	⟷	Orbital angular momentum
Superfluid excitations	⟷	Lattice phonons
...		...

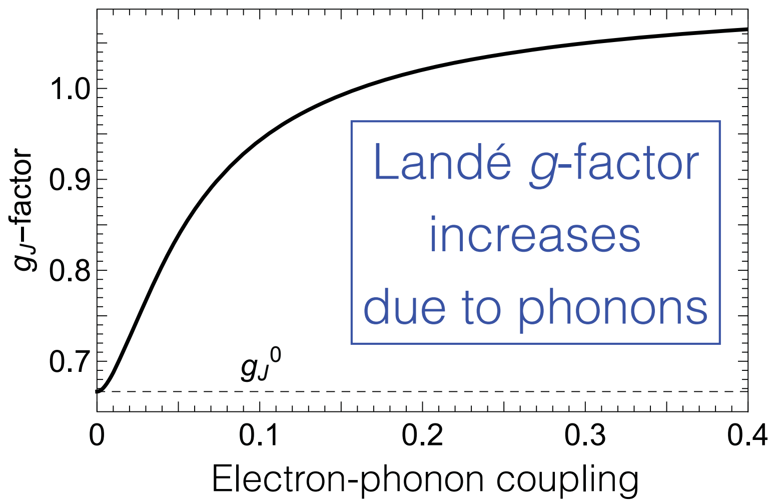
At every step

Compare to fully controlled experiments on molecules

Example:
renormalisation of Landé g-factor

Angulons in solids





Experiments on OH molecules in ^{4}He show exactly the same effect

(Douberly group, unpublished)

J. Mentink, M. I. Katsnelson, and ML, Phys. Rev. B **99**, 064428 (2019)

W. Rzadkowski and ML, J. Chem. Phys. **148**, 104307 (2018)

Summary

- Our claim: angulons provide a general framework to study angular momentum dynamics in quantum many particle systems
- We have shown: it works for molecules in superfluids

Tutorial chapter: ML, R. Schmidt, arXiv:1703.06753

Future directions

- Many-body techniques for the angulon problem: path integral, diagrammatic Monte Carlo, …

G. Bighin and ML, Phys. Rev. B **96**, 085410 (2017)
G. Bighin, T. Tscherbul, and ML, Phys. Rev. Lett., **121**, 165301 (2018)

- Applications to chemical reaction dynamics
- Applications to transport in hybrid organic/inorganic perovskites
- …

Any ideas and suggestions are welcome!

The group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Funding

FWF Der Wissenschaftsfonds.

2017–2020

erc

2019–2024

The group

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Don't miss their posters!

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Der Wissenschaftsfonds.

2017–2020

erc

2019–2024

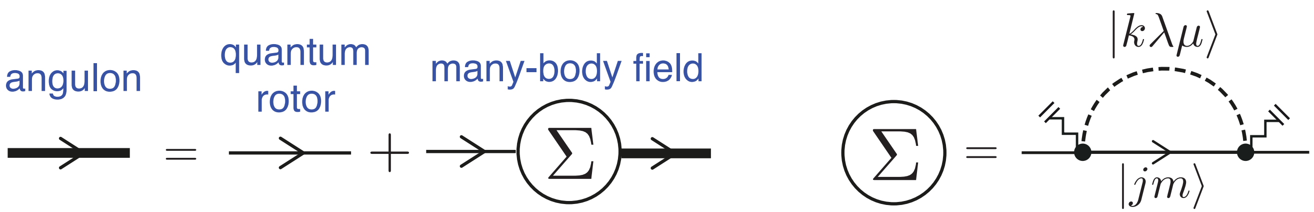
Spectrum of the angulon (an example)

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

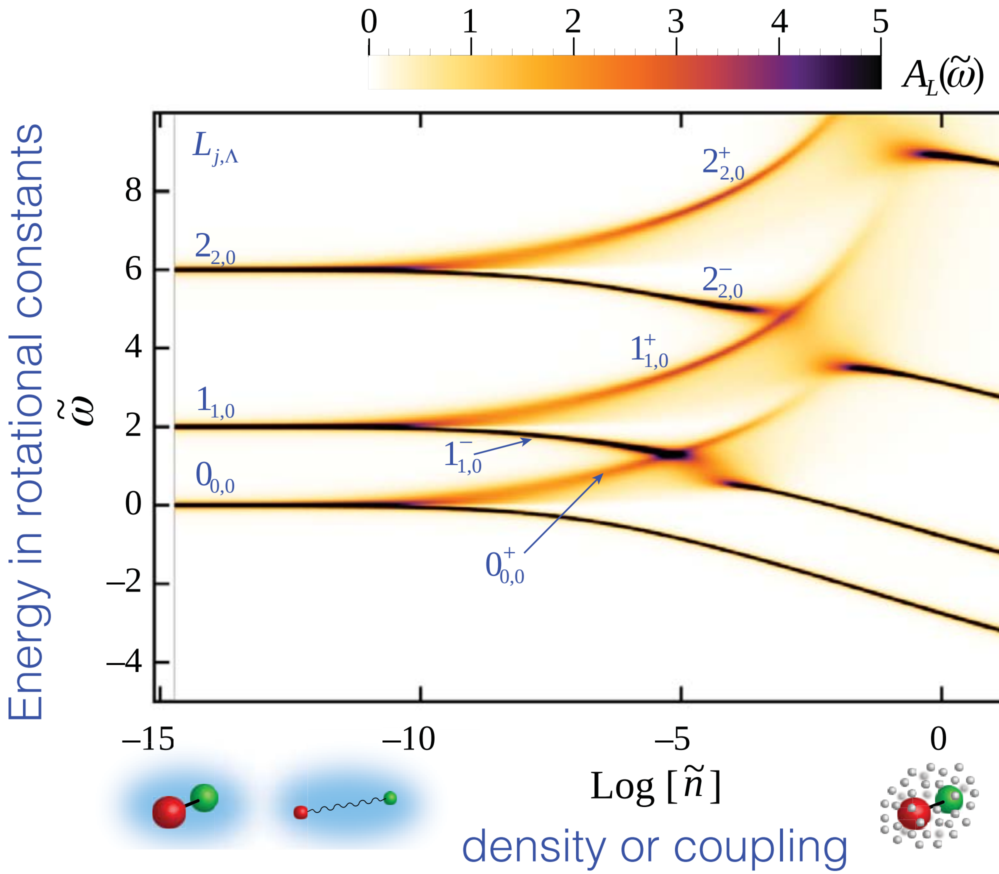
Simple variational ansatz: single bath excitations only

$$|\psi\rangle = Z^{1/2}_{LM}\,|0\rangle\,\underbrace{|LM\rangle}_{\text{total angular momentum}} + \sum_{\substack{k\lambda\mu\\ jm}} \beta_{k\lambda j}\,\underbrace{C^{LM}_{jm,\lambda\mu}}_{\text{angular momentum conservation}}\,\hat{b}^\dagger_{k\lambda\mu}\,|0\rangle\,|jm\rangle$$

The "angulon" quasiparticle



Spectrum of the angulon (an example)



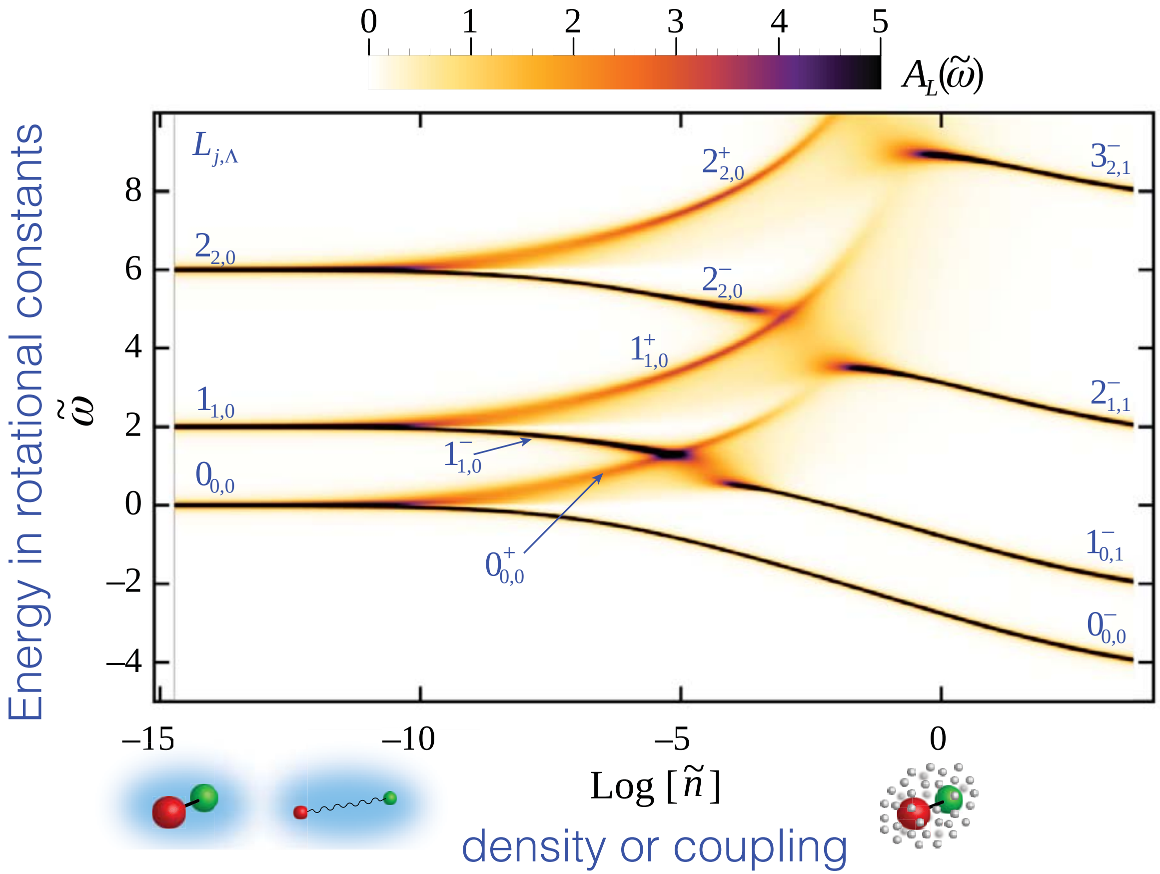
The only good quantum number: total angular momentum L

“Approximate” quantum numbers: angular momenta of molecule, j, and bosons, Λ

R. Schmidt and ML, Phys. Rev. Lett. **114**, 203001 (2015)

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

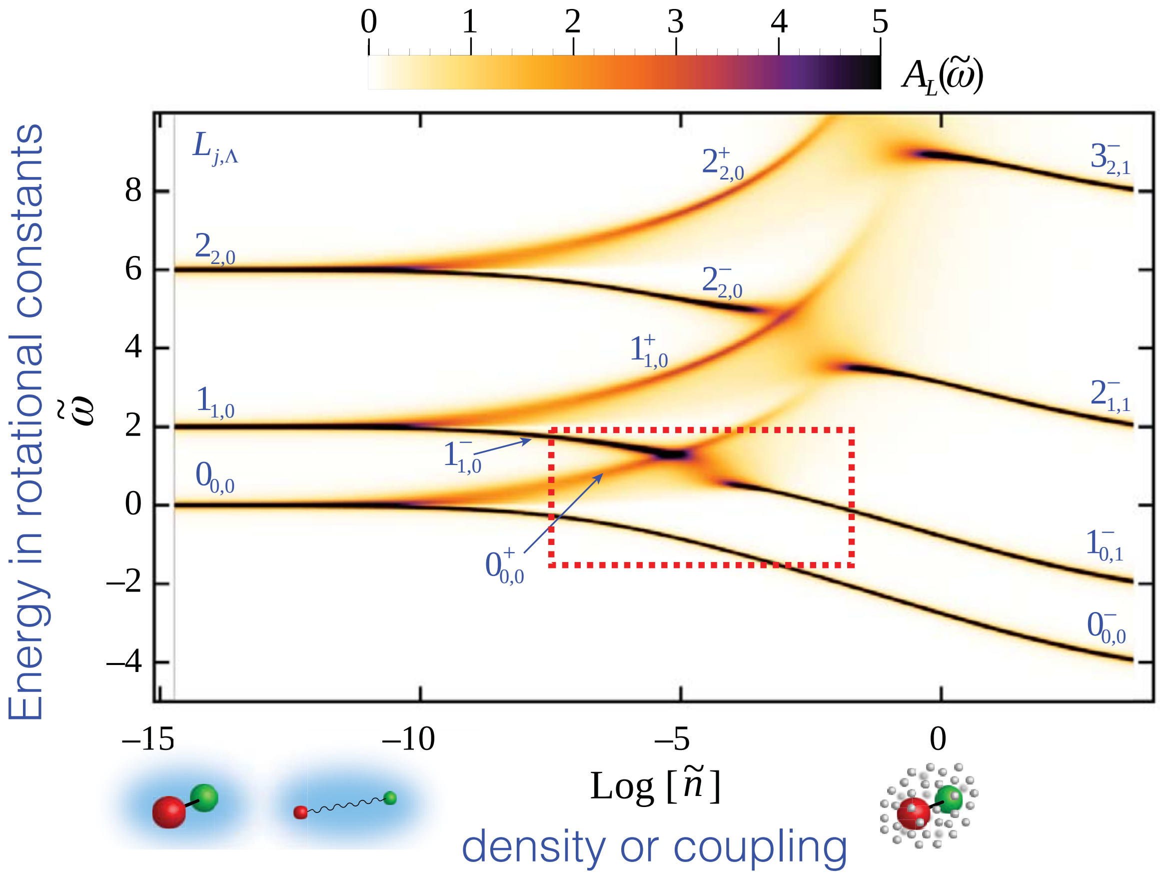
1. For different L, the energies change differently



Rotational Lamb shift due to the boson field

R. Schmidt and ML, Phys. Rev. Lett. **114**, 203001 (2015)
R. Schmidt and ML, Phys. Rev. X **6**, 011012 (2016)

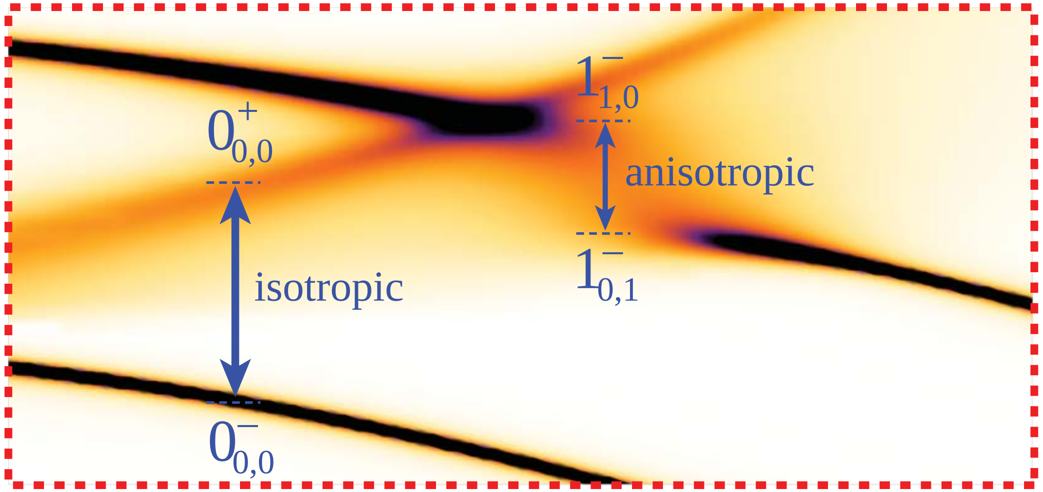
2. There are splittings of rotational lines



Let's zoom in…

R. Schmidt and ML, Phys. Rev. Lett. **114**, 203001 (2015)
R. Schmidt and ML, Phys. Rev. X **6**, 011012 (2016)

Angulon Fine Structure



Phonon wing (isotropic impurity-boson interactions)

Splitting between |*j*=*L*, no phonons> and |*j*=*L*, 1 phonon with $\lambda = 0$>

'Angulon instabilities' (anisotropic impurity-boson interactions)

Splitting between |*j*=*L*, no phonons> and |*j*=*L*–1, 1 phonon with $\lambda = 1$>

R. Schmidt and ML, Phys. Rev. Lett. **114**, 203001 (2015)

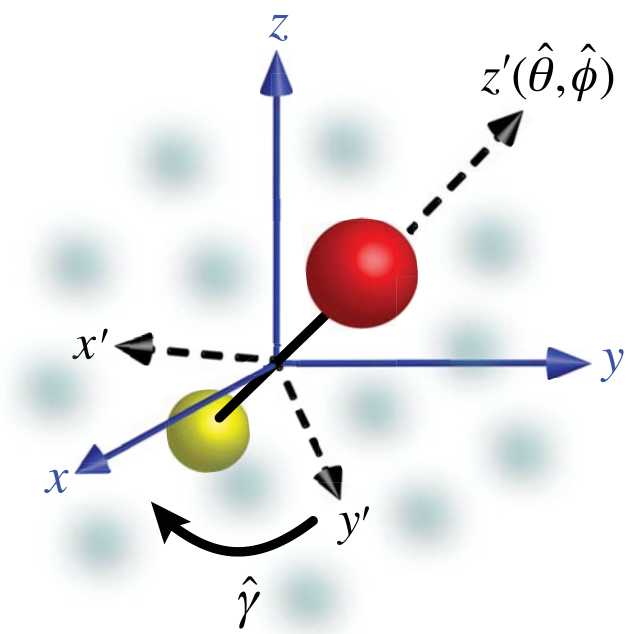
R. Schmidt and ML, Phys. Rev. X **6**, 011012 (2016)

The canonical transformation

Bosons: laboratory frame (x, y, z)

Molecule: rotating frame (x', y', z'),

defined by Euler angles $(\hat{\phi}, \hat{\theta}, \hat{\gamma})$



The canonical transformation

Bosons: laboratory frame (x, y, z)

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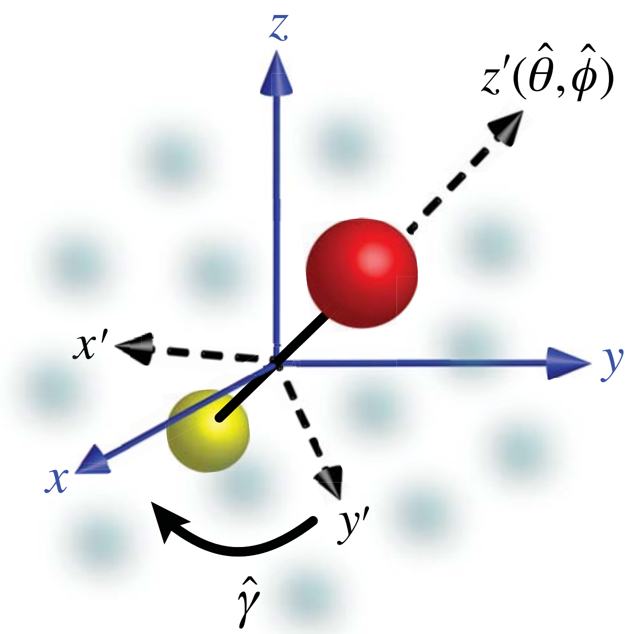

$$\hat{S} = e^{-i\hat{\phi}\otimes\hat{\Lambda}_z} e^{-i\hat{\theta}\otimes\hat{\Lambda}_y} e^{-i\hat{\gamma}\otimes\hat{\Lambda}_z}$$

With $\hat{\mathbf{\Lambda}} = \sum_{k\lambda\mu\nu} \hat{b}^\dagger_{k\lambda\mu} \boldsymbol{\sigma}^\lambda_{\mu\nu} \hat{b}_{k\lambda\nu}$ the total angular momentum of bosons

angular momentum matrices

Transformed Hamiltonian

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

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

Why is it better?

1. It does not contain the molecular coordinates (angles)

Original H: coupling mixes 3D angular momenta, leading to 3*jn*-symbols

Transformed H: coupling ~ $\mathbf{L} \cdot \Lambda$ (as e.g. in spin-orbit interaction)

Addition of 3D angular momenta is replaced by addition of “spins”

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

Transformed Hamiltonian

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

$$\downarrow$$

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

Why is it better?

2. The transformation singles out total angular momentum,

$\hat{\mathbf{L}} = \hat{\mathbf{J}} + \hat{\mathbf{\Lambda}}$, the only conserved quantity of the problem

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

Transformed Hamiltonian

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

Why is it better?

3. In the regime of *B*=0, *H* can be diagonalized exactly:

$$\hat{\mathscr{H}} = \hat{U}^{-1}\hat{\mathcal{H}}\hat{U} \qquad \hat{U} = \exp\left[\sum_{k\lambda} \frac{U_\lambda(k)}{\omega_k}(\hat{b}_{k\lambda 0} - \hat{b}^\dagger_{k\lambda 0})\right]$$

Thus, one can look for perturbative solutions near the state which already contains an **infinite number of phonon excitations**:

$$|\psi\rangle = g_{LM}|0\rangle|LM0\rangle + \sum_{k\lambda n} \alpha_{k\lambda n} \hat{b}^\dagger_{k\lambda n}|0\rangle|LMn\rangle$$

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