Ultracold molecules, from dilute to quantum degenerate gases

Charbel Karam

Laboratoire interdisciplinaire Carnot de Bourgogne

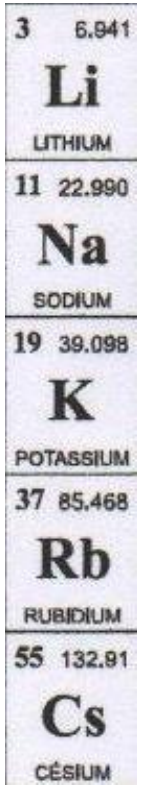
Winter school – Physics and Mathematics of Bose-Einstein Condensates

25th of February 2025

Outline

- Introduction: System – Goal – Applications.
- Planning a BEC of ground state molecules
- Dealing with losses: Shielding of collisions
- Results and First observation of molecular BEC

Introduction: system

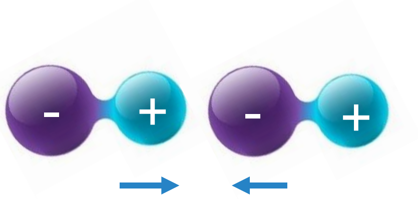

3 6.941
Li
LITHIUM
11 22.990
Na
SODIUM
19 39.098
K
POTASSIUM
37 85.468
Rb
RUBIDIUM
55 132.91
Cs
CÉSIUM

Polar bi-alkali metal ultracold molecules:

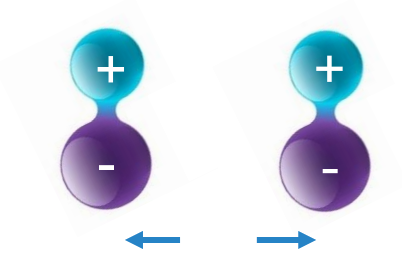
- Strong permanent electric dipole moment (body fixed frame)

→ Easily manipulated by external electric fields

→ Long range anisotropic interactions:
dipole-dipole interaction (DDI)

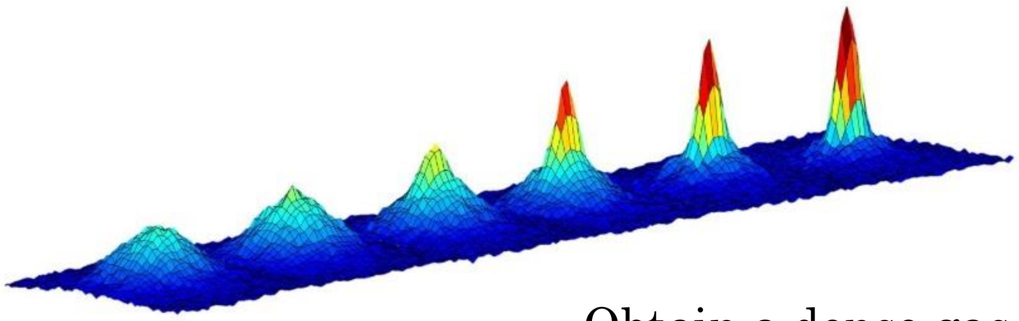


Attractive



Repulsive

Introduction: Goal

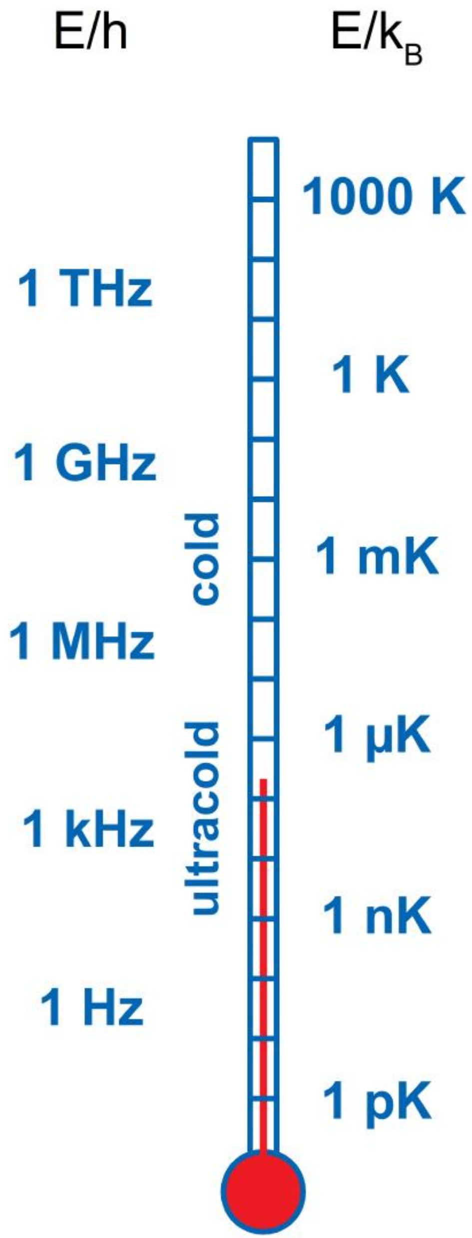


Obtain a dense gas ($10^{12} – 10^{15}$ cm^{-3})

of polar ultracold molecules (<<1mK)

in their absolute ground state.

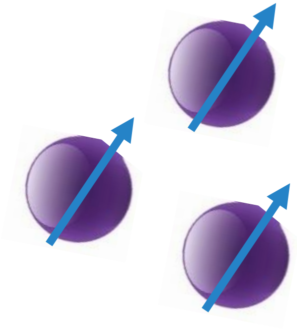
Aiming to reach quantum degeneracy.



Introduction: Cold dipolar systems

Dipole moment →

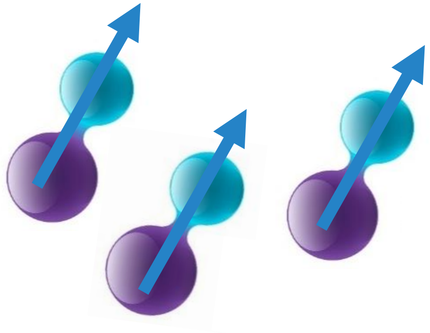
Magnetic Atoms



e.g.: Cr, Er, Dy, etc…

Long-lived, very cold,
Weaker dipole moment

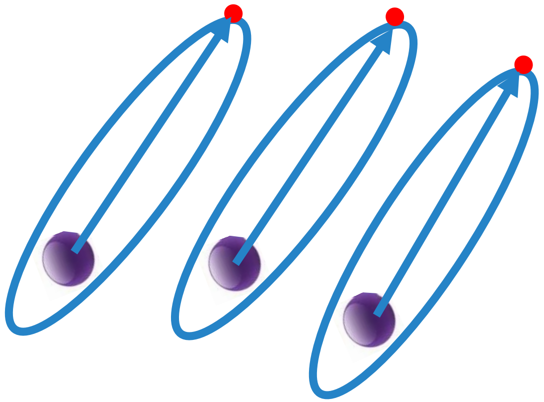
Ultracold Polar Molecules



e.g.: NaK, NaRb, NaCs, RbCs,
SrF, BaF, YO, CaOH, etc…

Intermediate lifetime
Intermediate to Strong dipole moment

Rydberg Atoms



Short lifetime
Strong dipole moment,

← Lifetime

Introduction: Applications

Molecular quantum degenerate gases bring many of the theoretical predictions on dipolar gases into experimental reach

Macroscopic manifestation of microscopic anisotropic interactions

→ Simulation of condensed matter systems.

→ Transitions to a variety of exotic supersolid states.

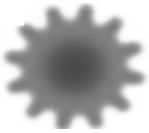
Supersolid states of trapped molecular BECs


Droplet arrays


Honeycomb


Stripe


Labyrinth

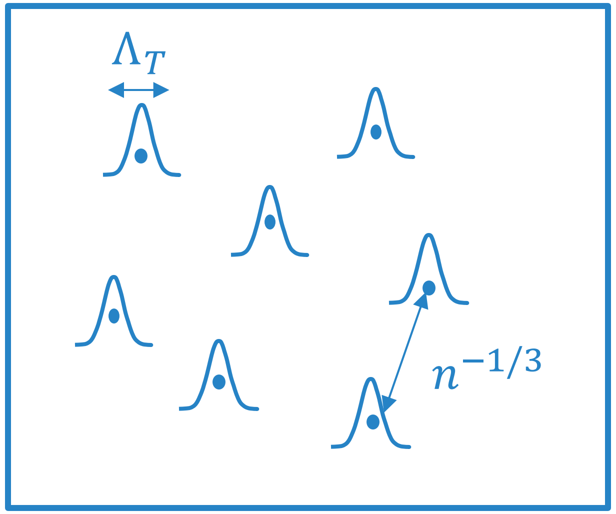

Pumpkin

Schmidt *et al* PRR4,013235 (2022)

Introduction: Reminder on degenerate gases: $n\,{\Lambda_T}^3 \approx 1$

What do we need to reach quantum degeneracy?

$\Lambda_T \propto 1/\sqrt{T}$



Classical regime

(dilute gas)

$\Lambda_T = \sqrt{\frac{2\pi\hbar^2}{mk_BT}} \equiv$ De Broglie wavelength

$\mathrm{T} \equiv$ gas temperature

$\mathrm{n} = \frac{N}{V} \equiv$ gas density

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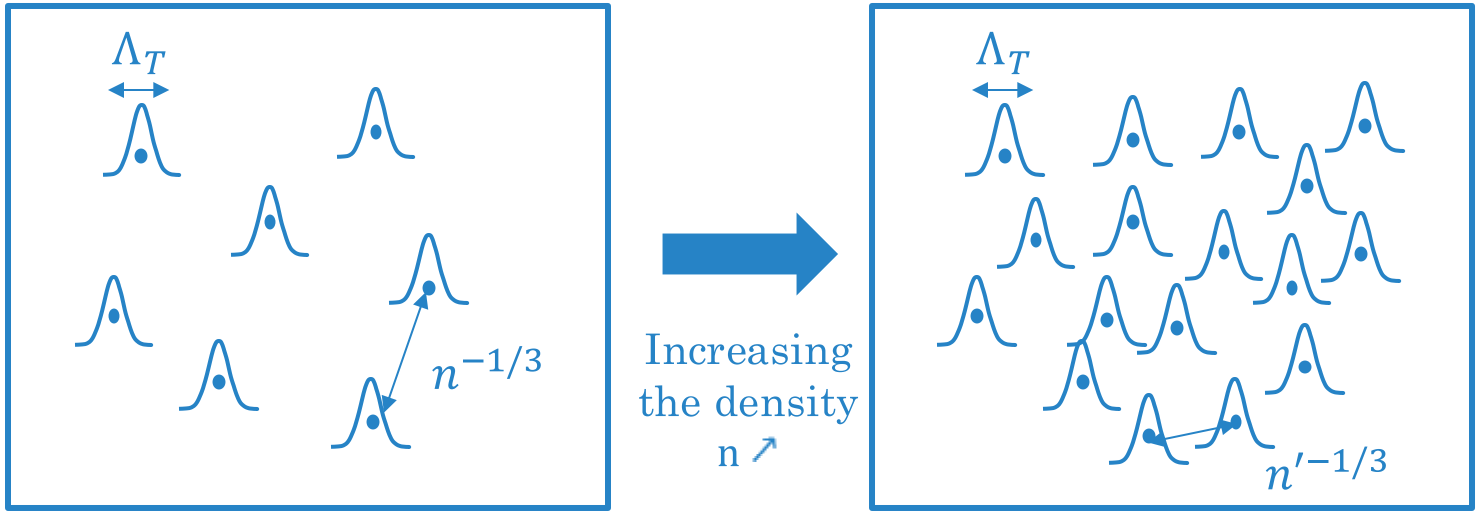

Increasing the density
n ↗

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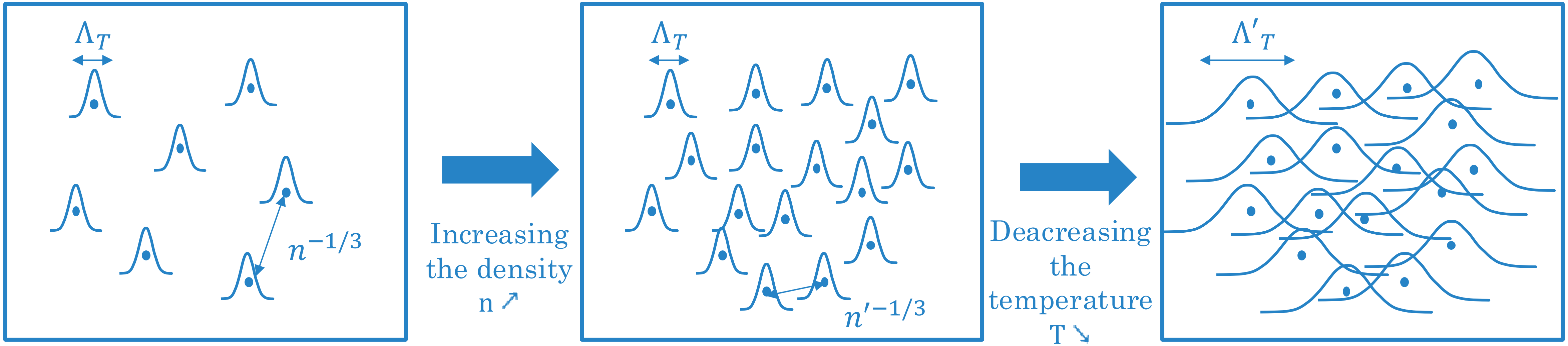

Increasing the density
n ↗

Deacreasing the temperature
T ↘

Classical regime
(dilute gas)

Quantum regime
(correlated gas)

$\Lambda_T = \sqrt{\frac{2\pi\hbar^2}{mk_BT}} \equiv$ De Broglie wavelength

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Introduction: Reminder on degenerate gases: $n\,\Lambda_T{}^{3} \approx 1$

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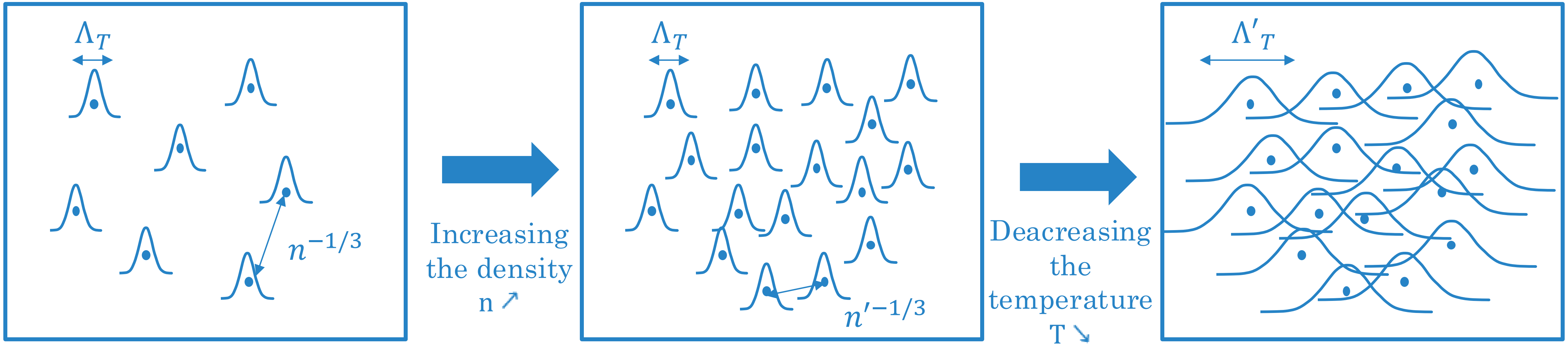


Classical regime

(dilute gas)

Quantum regime

(correlated gas)

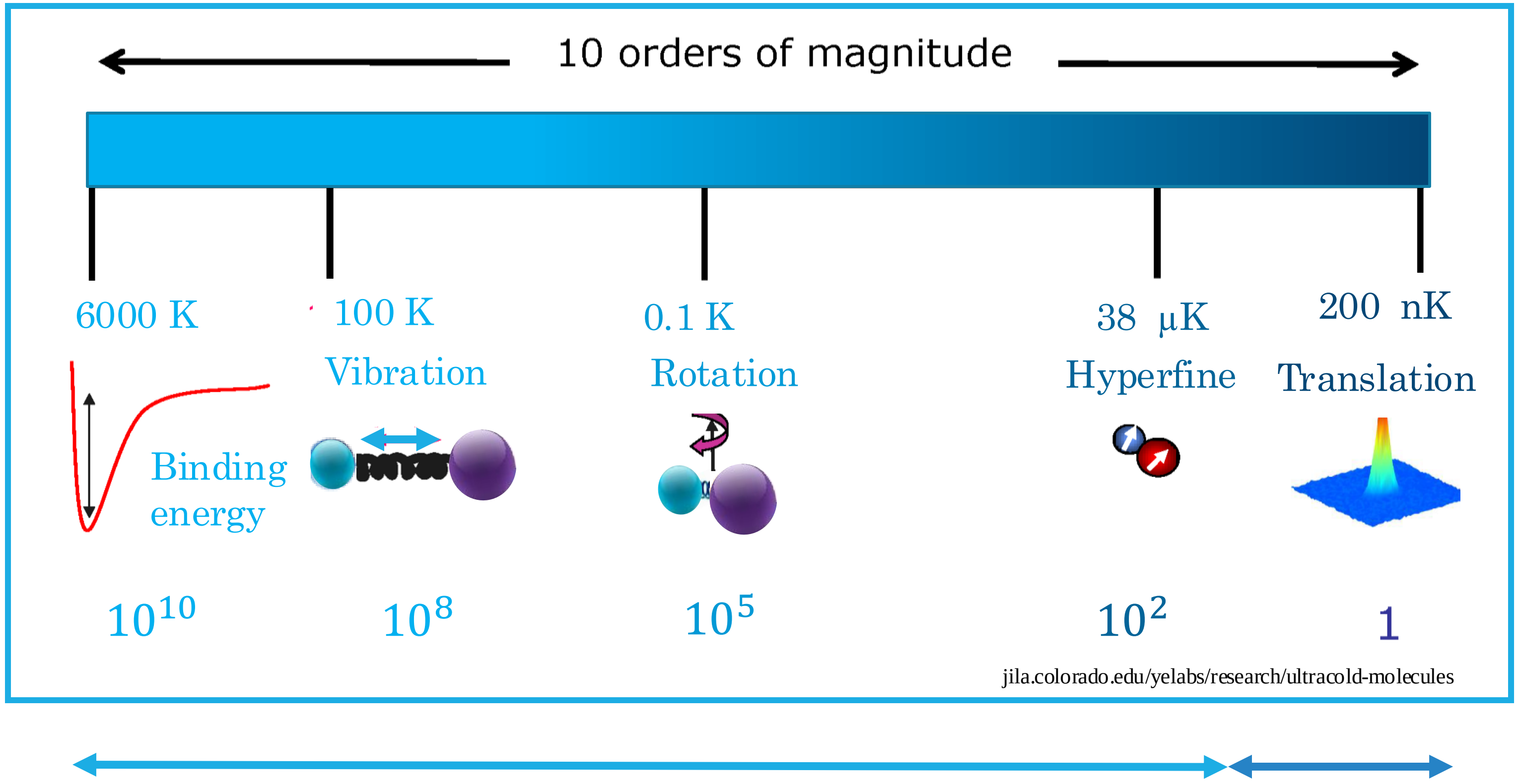
Two main ingredients:
Temperature and Density

$\Lambda_T = \sqrt{\frac{2\pi\hbar^2}{m k_B T}} \equiv$ De Broglie wavelength

T ≡ gas temperature

$n = \frac{N}{V} \equiv$ gas density

Temperature/Energy orders of magnitude in bi-alkali molecules



10 orders of magnitude

6000 K	100 K	0.1 K	38 μK	200 nK
Binding energy	Vibration	Rotation	Hyperfine	Translation
10^{10}	10^{8}	10^{5}	10^{2}	1

jila.colorado.edu/yelabs/research/ultracold-molecules

Internal
degrees of freedom

External
degree of freedom

Lifetime of ground state $^{23}Na^{39}K$ gas:

Density 10^{12} cm^{-3}
Temperature 300 nK

$^{23}Na^{39}K$ is not chemically reactive:

$$NaK + NaK \not\rightarrow Na_2 + K_2$$

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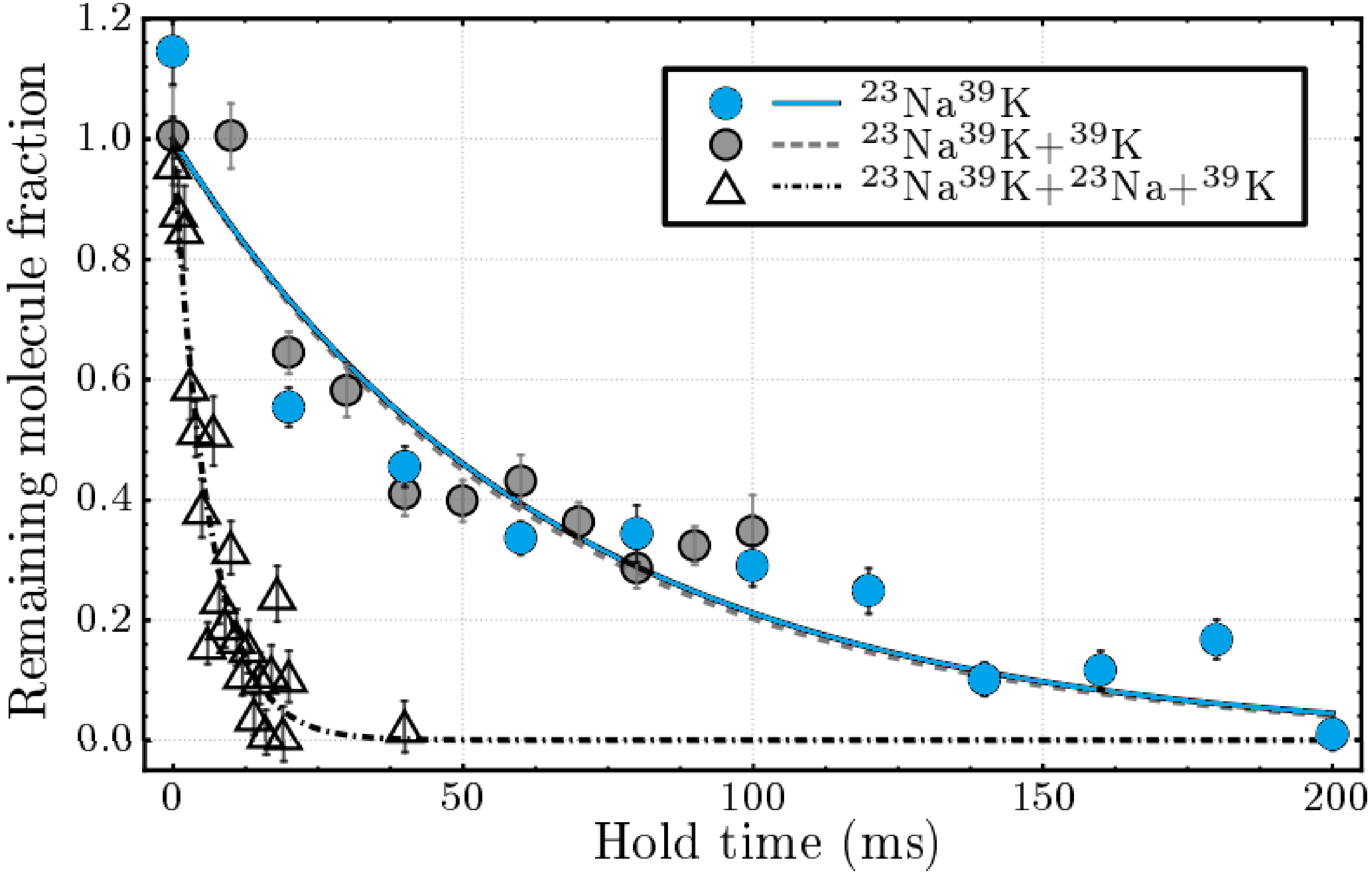
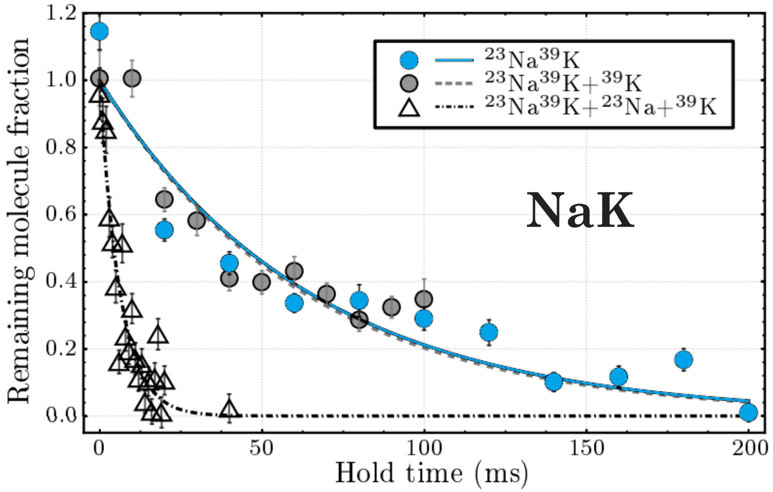


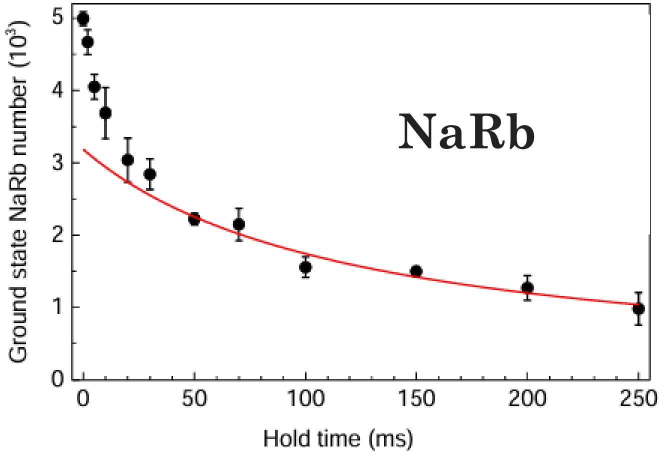
Loss rate:
$4.49\ (\pm 1.18) \times 10^{-10}\ \mathrm{cm^3\ s^{-1}}$

Hannover
PRL 125, 083401 (2020)

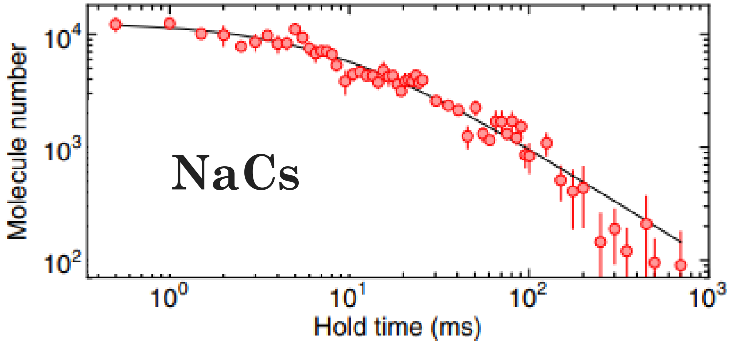
Not only $^{23}Na^{39}K$ gas:



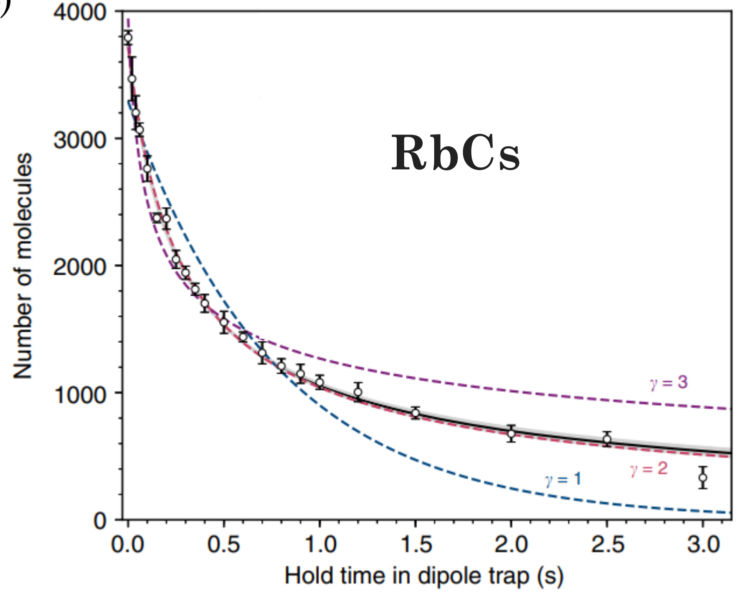
Hannover
PRA 101, 042704 (2020)



Hong Kong
PRL **116**, 205303 (2016)



Columbia U
PRL 130, 113002 (2023)

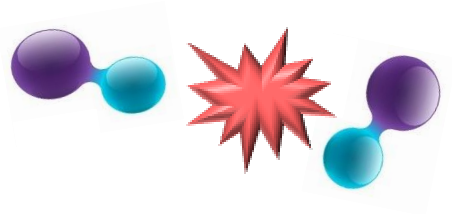


Durham
Nature commun 10, 3104 (2019)

Consequence: the density decreases preventing the formation of a degenerate gas…

Shielding collisions

Shielding



Engineering the long-range interaction
between ultracold molecules
using external fields:

Changing attractive interactions into repulsive ones
→ Shield the collisions
→ Prevent the loss.

Shielding

Theoretical Proposals:

→ Static electric field

Avdeenkov *et al* PRA 73 022707 **(2006)**
Wang *et al* New J.Phys 17 035015 **(2015)**

Experimental validation:

→ On fermionic KRb
For E= 12 kV/cm

Li *et al* Nature Phys. 17, 1144 **(2021)**

Shielding

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On bosonic NaRb

Lin *et al* PRX 13, 021032 **(2023)**

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Bigagli *et al* Nature Phys. 19, 1579 **(2024)**

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Optical field

X.Tie *et al* PRL 125, 153202 **(2020)**

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Two-photon Optical field

Charbel Karam *et al.* PRR 5 033074 (2023)

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In progress
on bosonic NaK

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Li *et al* Nature Phys. 17, 1144 **(2021)**

→ On fermionic NaK → Degenerate Fermi Gas

Schindewolf *et al* Nature 607, 677 **(2022)**

On bosonic NaRb

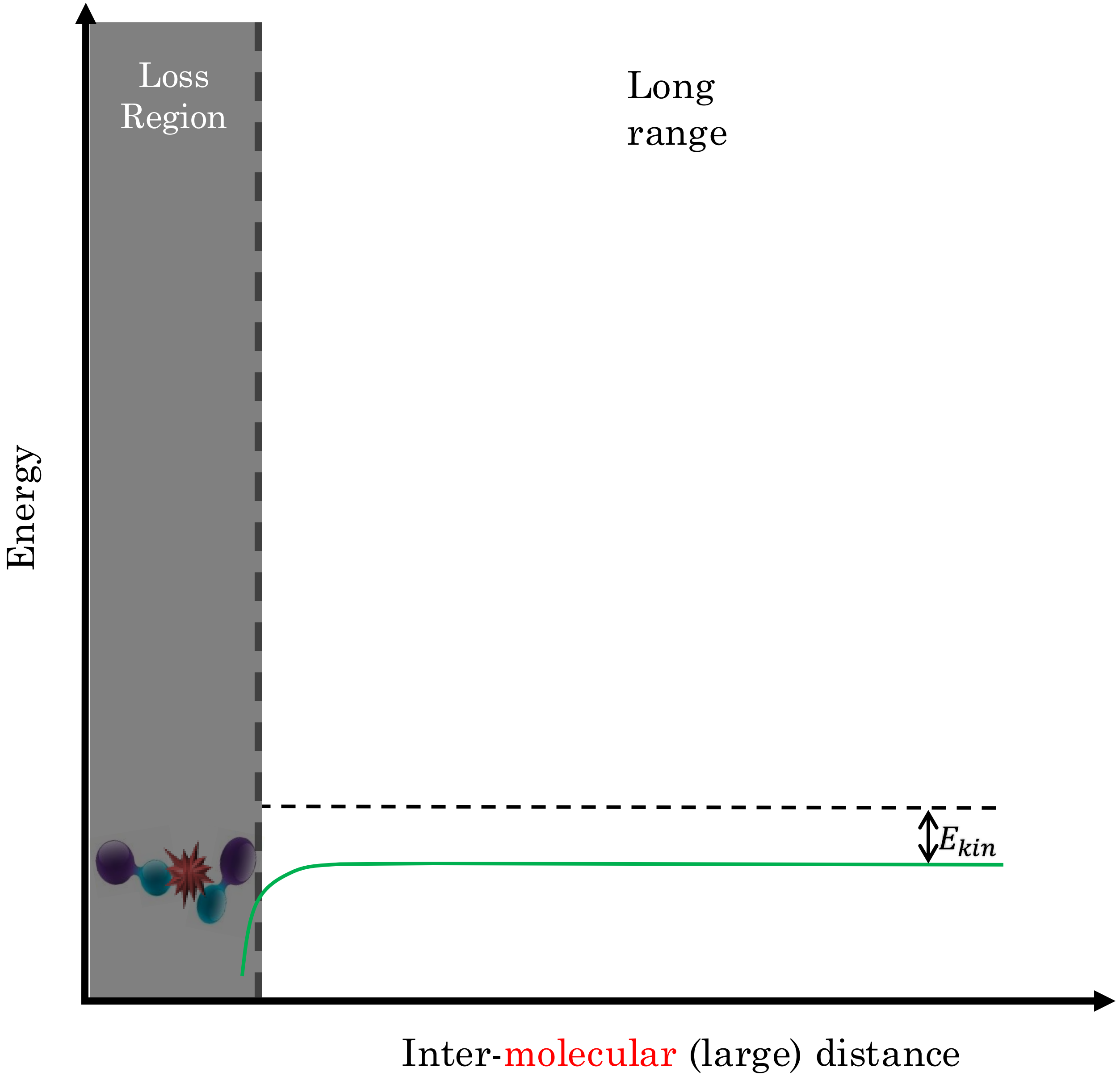
Lin *et al* PRX 13, 021032 **(2023)**

On bosonic NaCs → First BEC of ground state dipolar molecules

Bigagli *et al* Nature Phys. 19, 1579 **(2024)**

→ In progress
on bosonic NaK

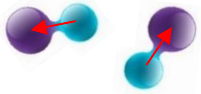
How to shield ?



$$\hat{H} = \hat{T} + \frac{\widehat{L^2}}{2\mu R^2} + \hat{H}_{rot} + \boxed{\hat{V}_{mol-mol}(R)}$$

Mainly anisotropic dipole-dipole interaction $\propto \frac{1}{R^3}$.

Ground + Ground



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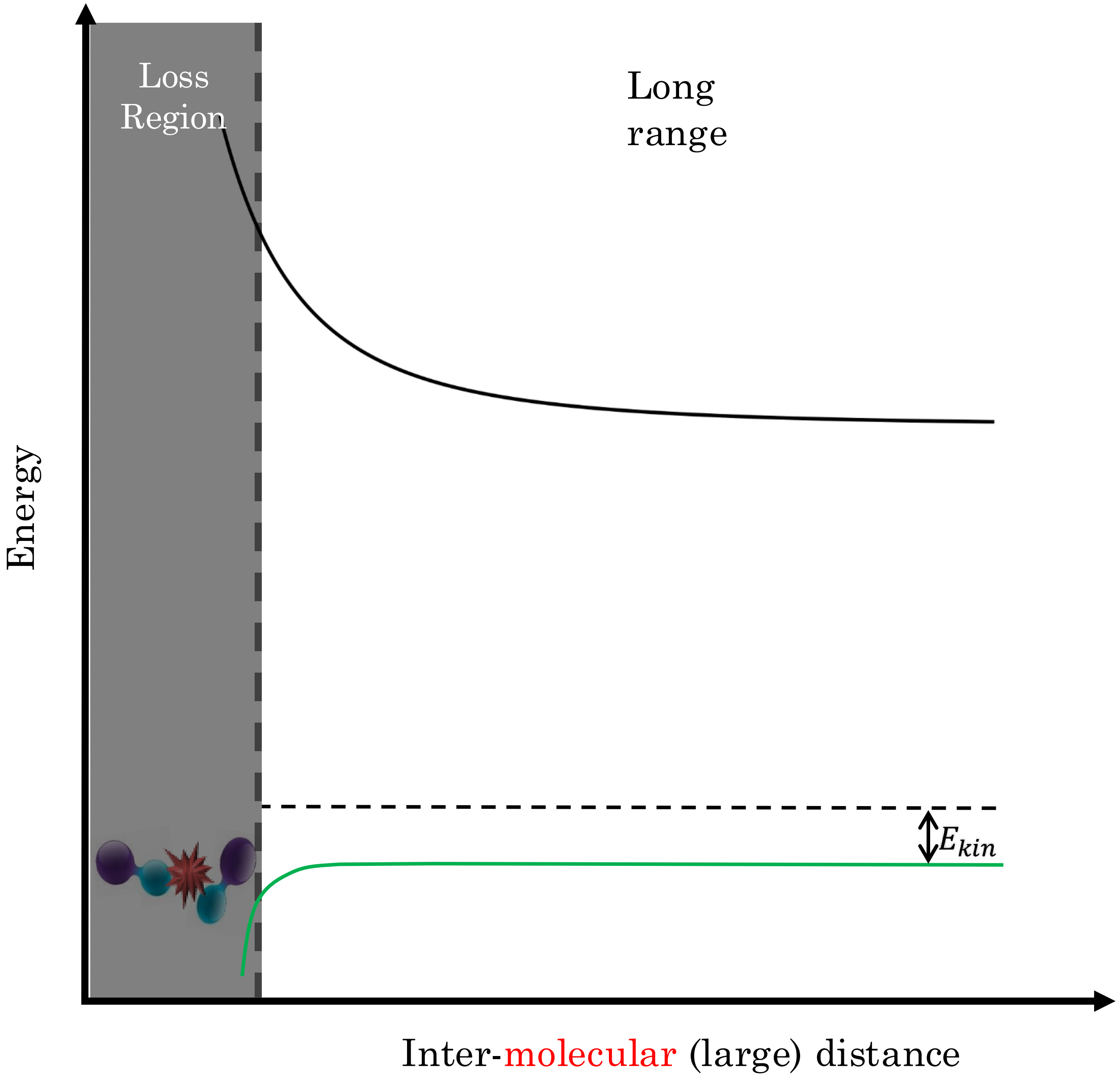

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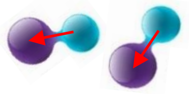
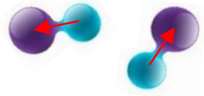
Ground + Excited



Ground + Ground



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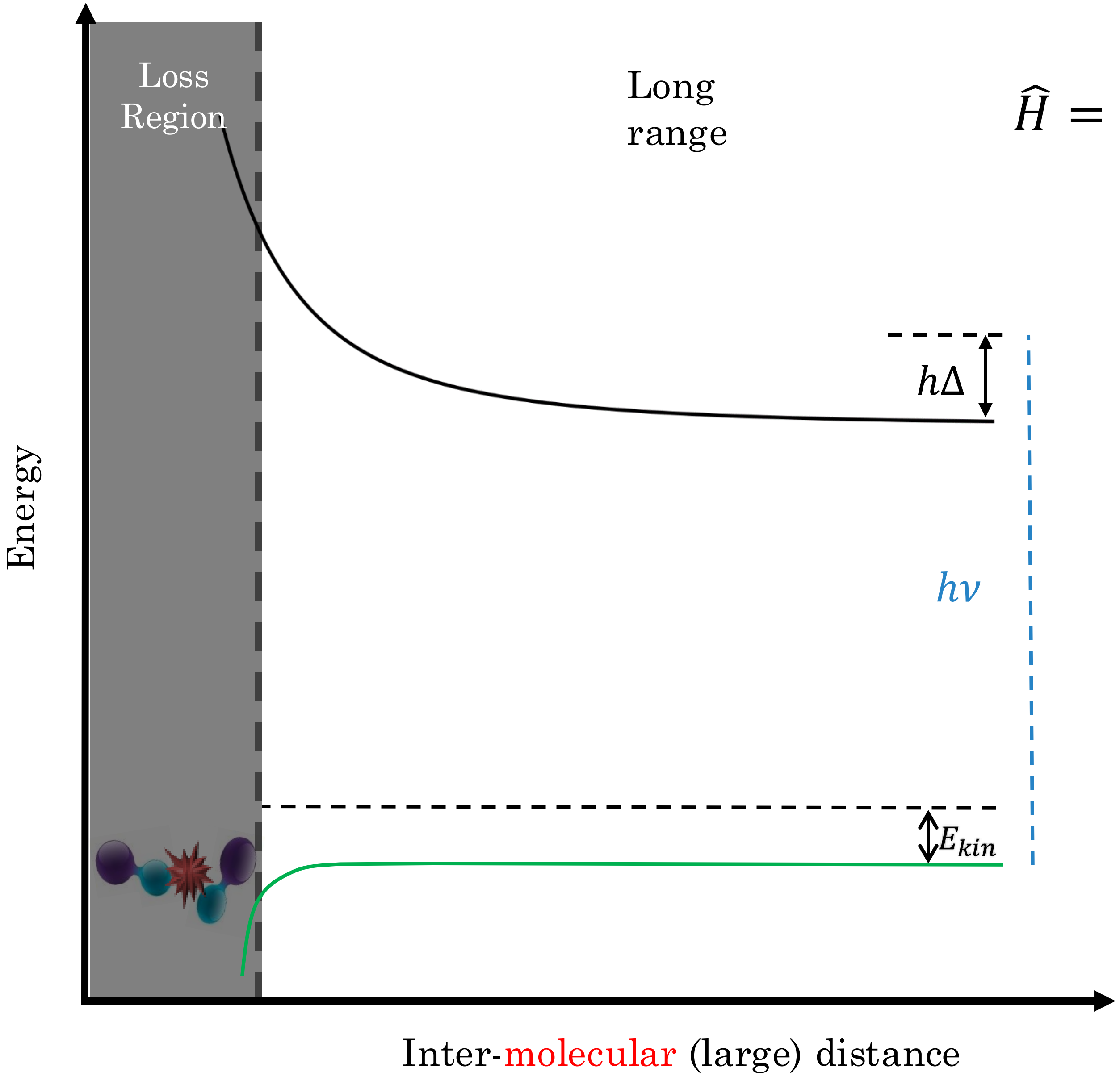
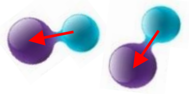


$$\hat{H} = \hat{T} + \frac{\hat{L}^2}{2\mu R^2} + \hat{H}_{rot} + \hat{V}_{mol-mol}(R) + \boxed{\hat{H}_f}$$

$$\boxed{\hbar\omega\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right)}$$

Ground + Excited



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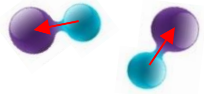
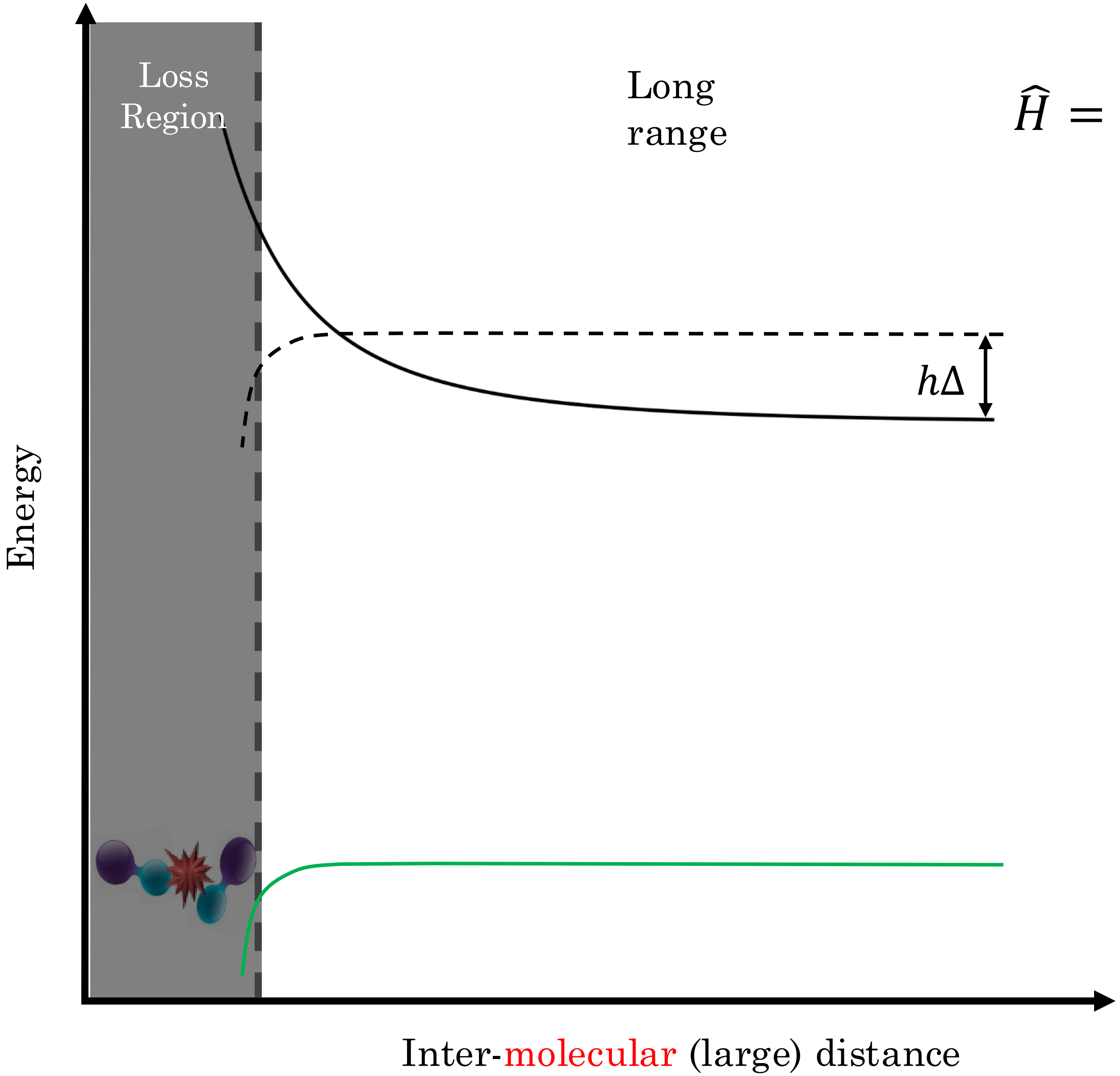


We couple the ground state to a repulsive excited state using a blue detuned photon by respect to the transition.

How to shield ?



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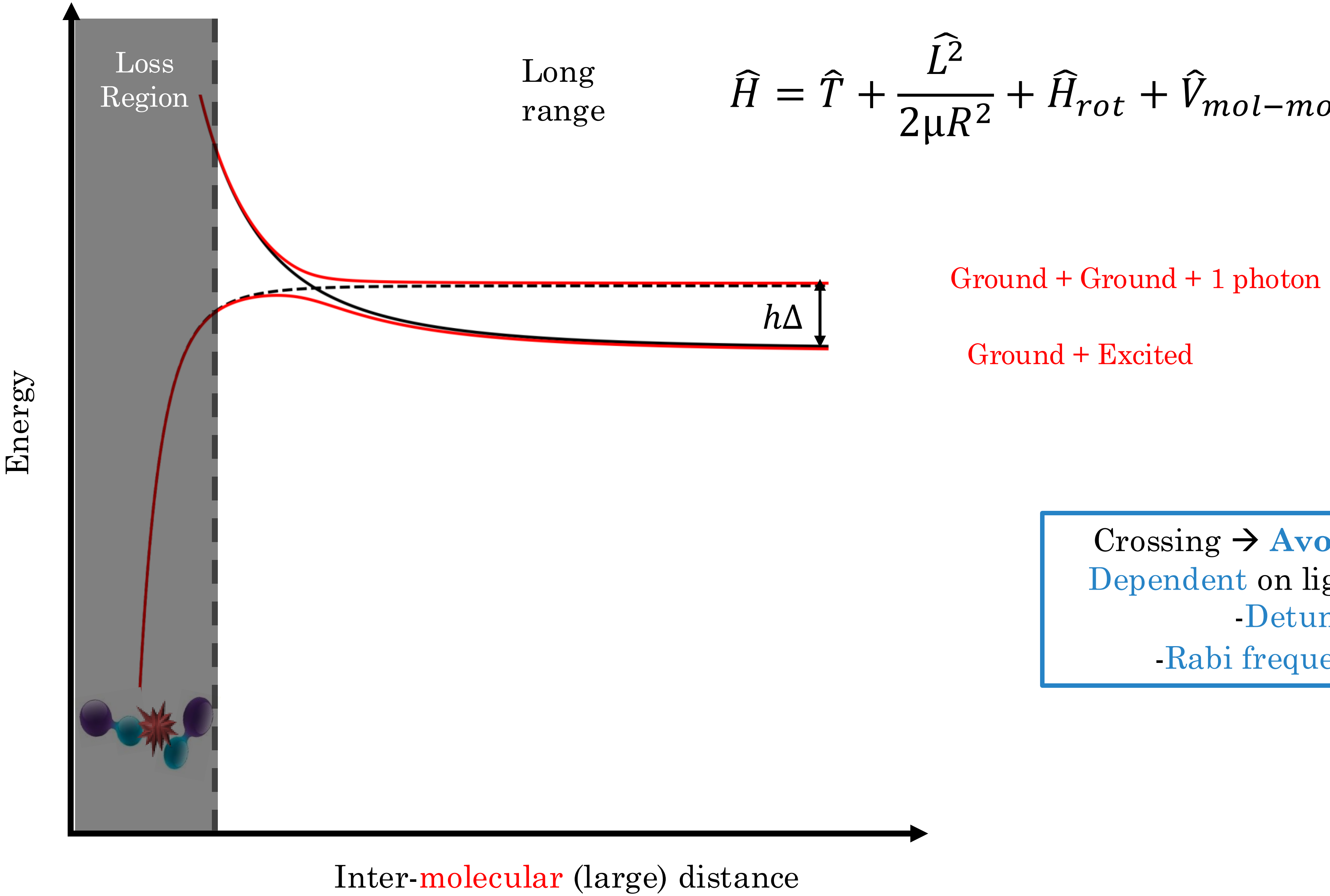
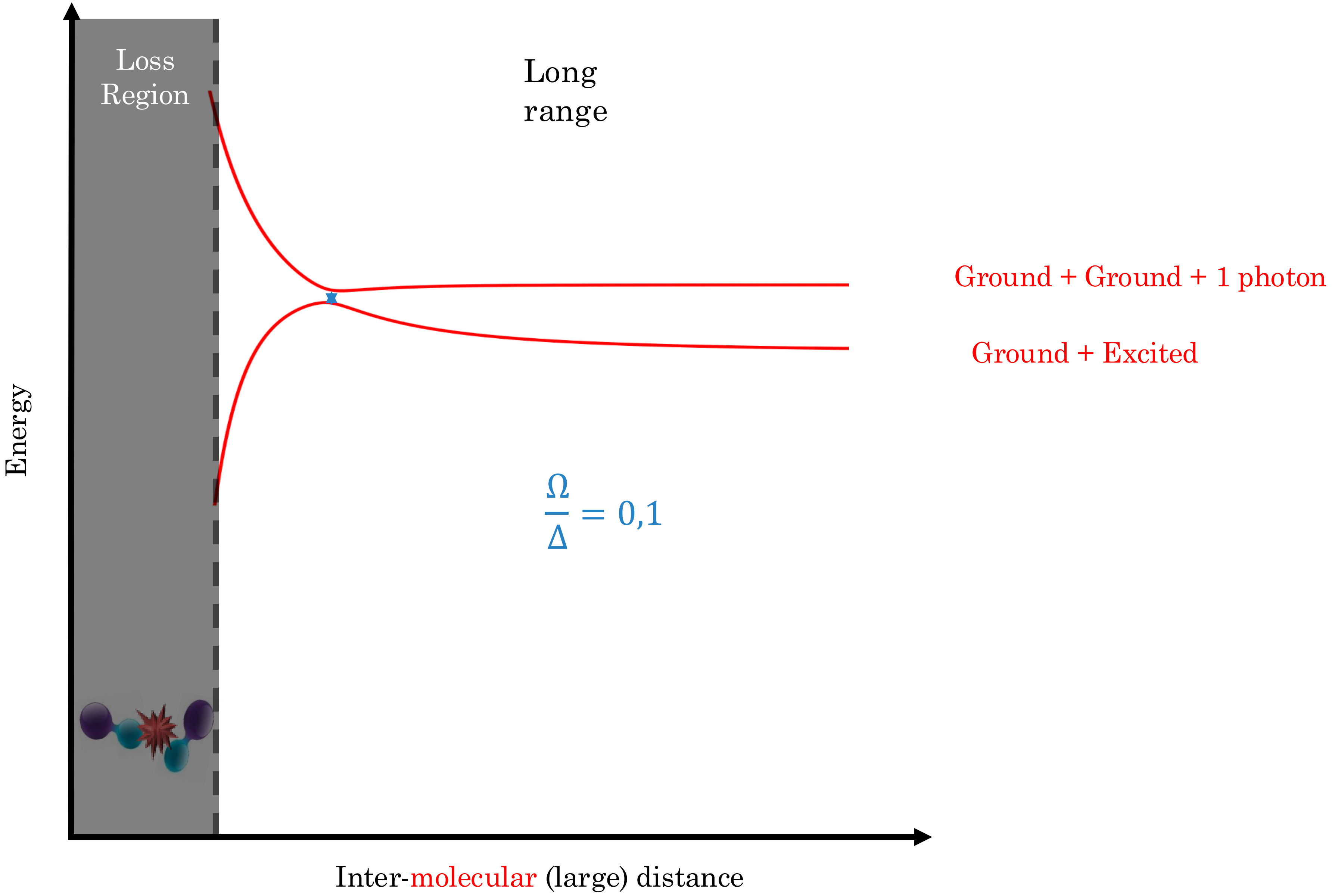
$$\downarrow$$

$$\boxed{-\vec{d}.\vec{E}(t)}$$



Crossing → **Avoided crossing**
Dependent on light parameters:
-Detuning Δ
-Rabi frequency $\Omega \propto \sqrt{I}$

How to shield: Dependency on light parameters



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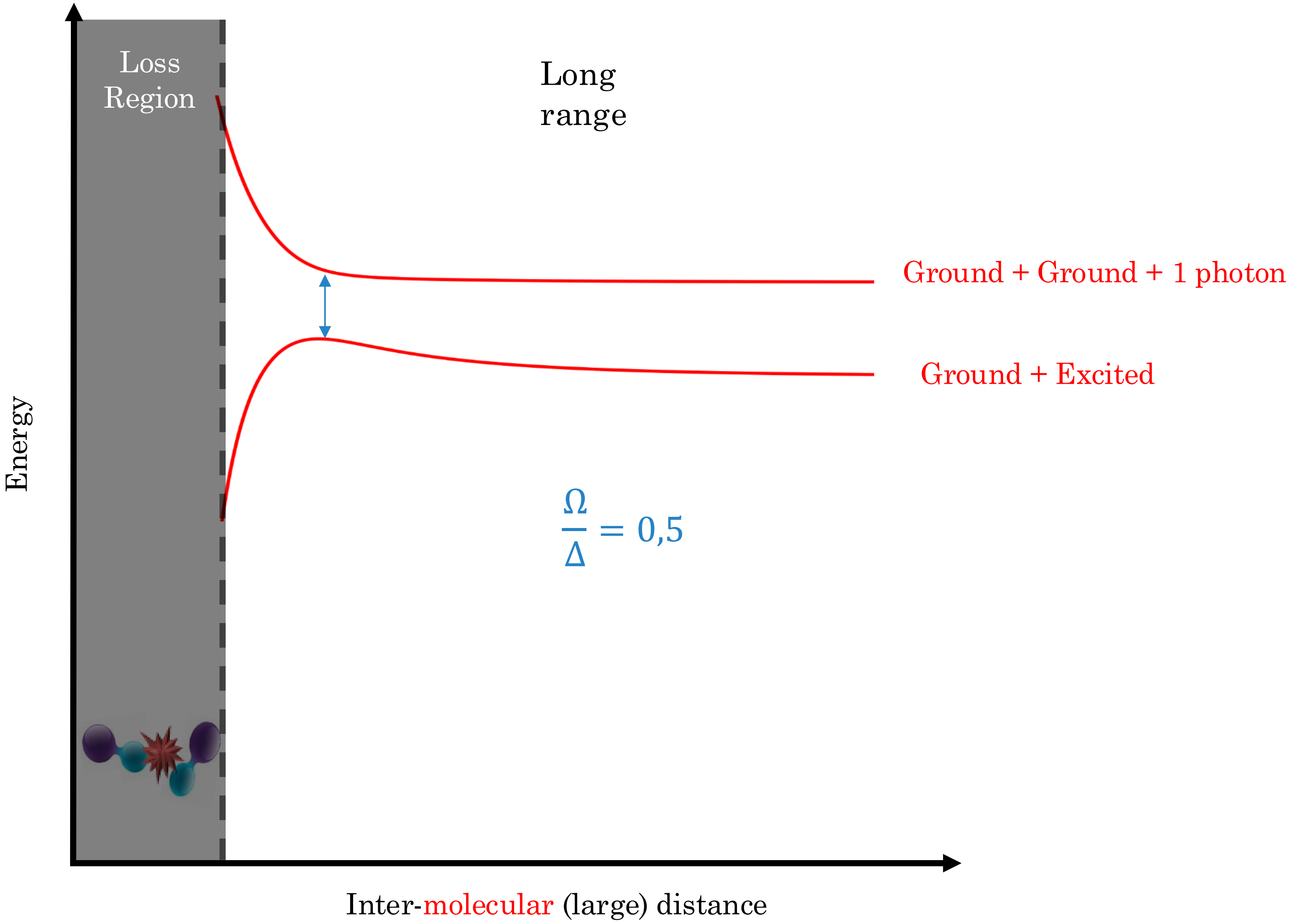

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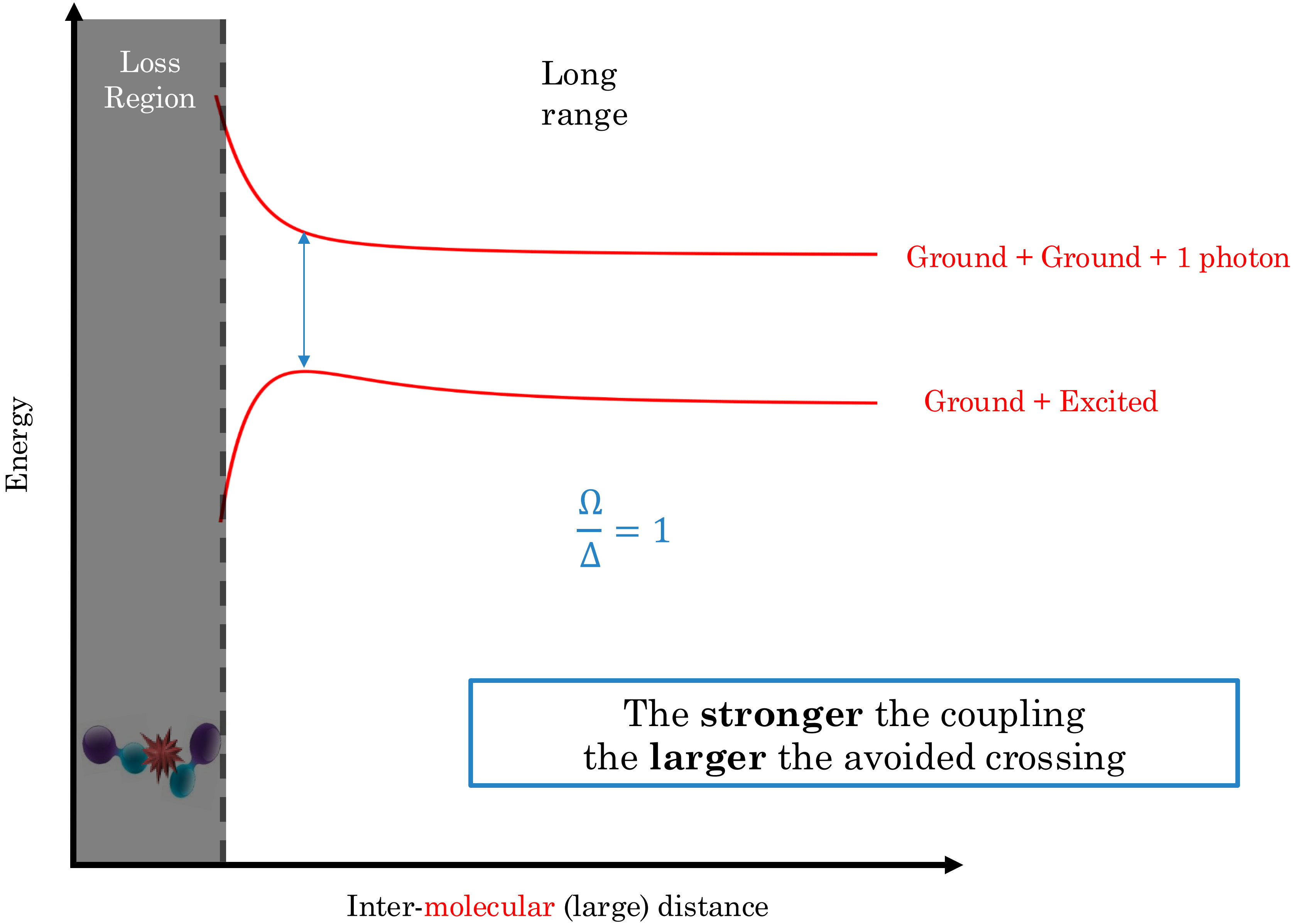
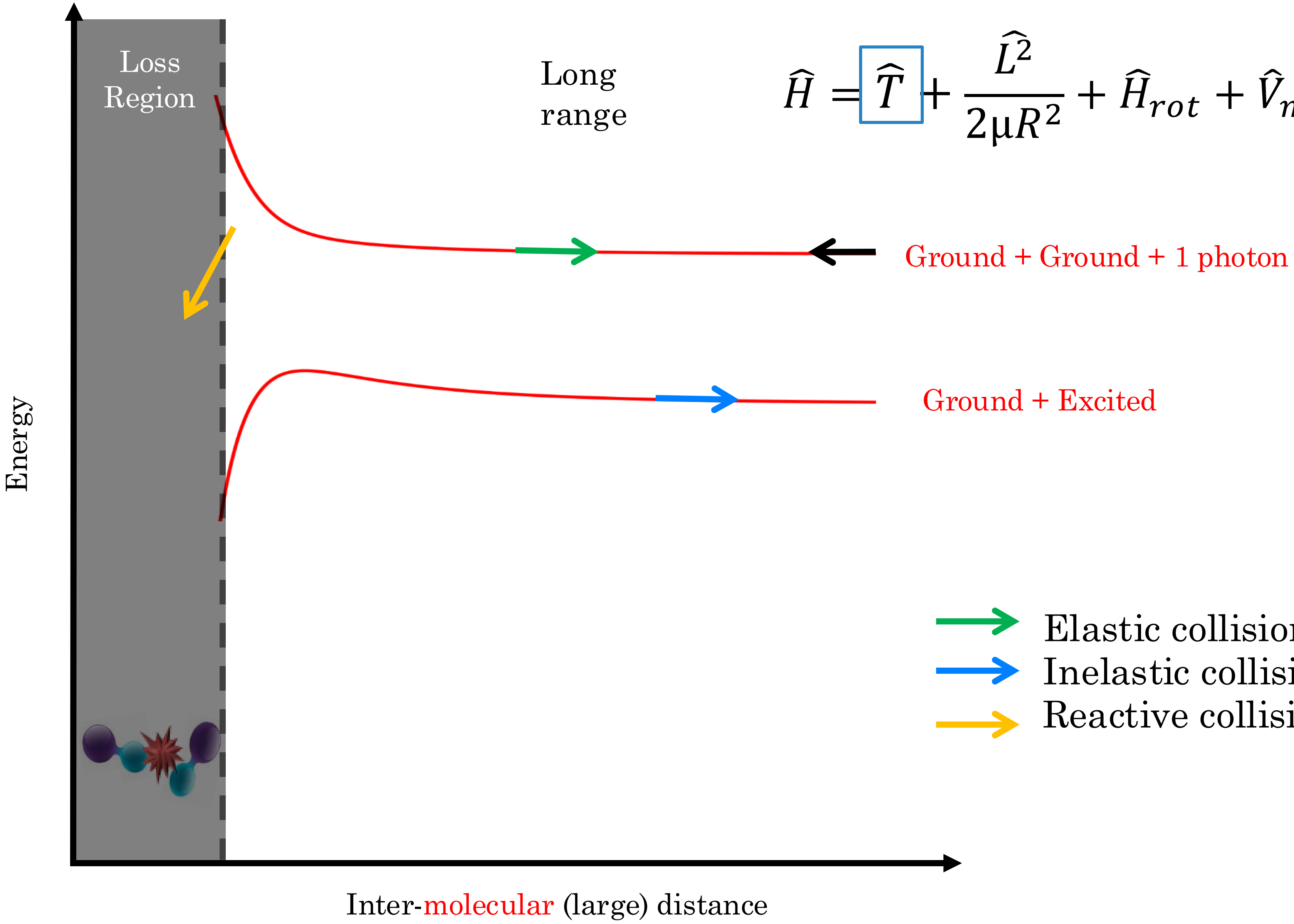


$$\frac{\Omega}{\Delta} = 1$$

The **stronger** the coupling
the **larger** the avoided crossing

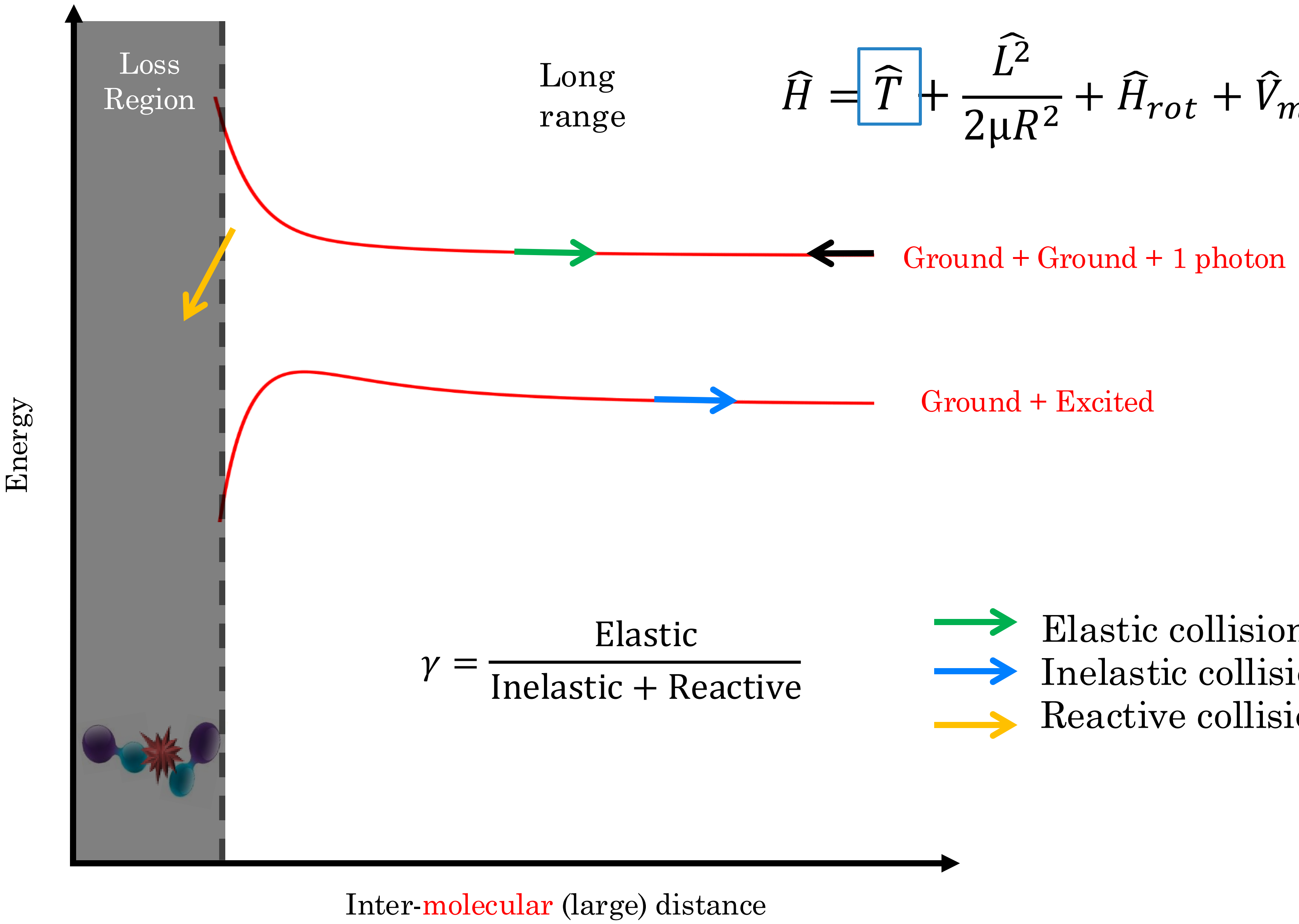
How to shield: Dynamics and collision rates

$$\hat{H} = \boxed{\hat{T}} + \frac{\hat{L}^2}{2\mu R^2} + \hat{H}_{rot} + \hat{V}_{mol-mol}(R) + \hat{H}_f + \hat{H}_I$$



- → Elastic collisions
- → Inelastic collisions (heat ≡ T ↗)
- → Reactive collisions (loss ≡ n ↘)

How to shield: Dynamics and collision rates



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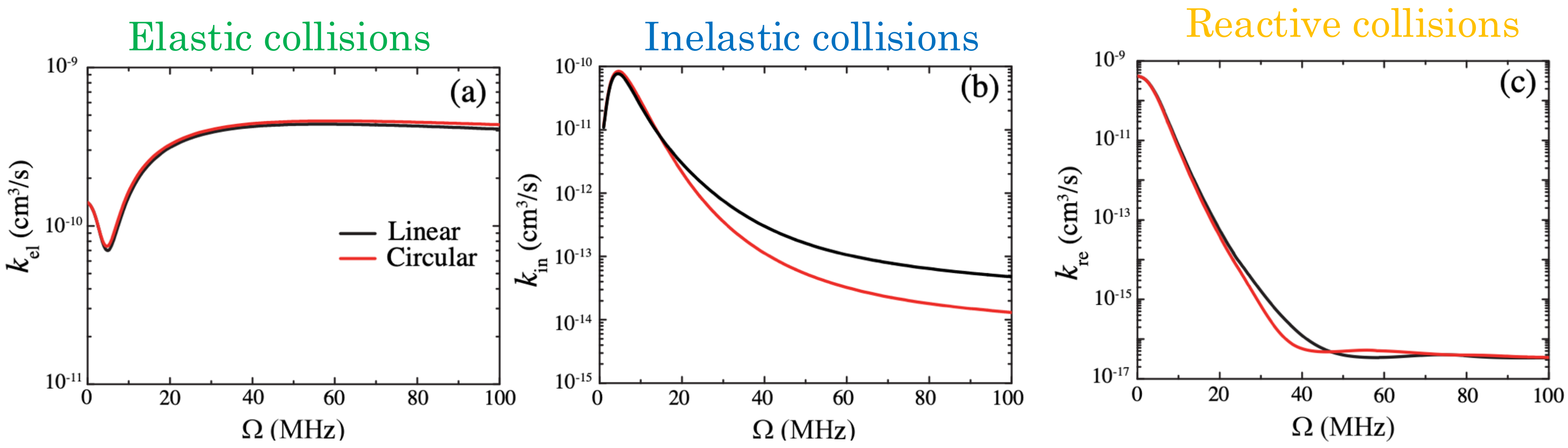
$$\gamma = \frac{\text{Elastic}}{\text{Inelastic} + \text{Reactive}}$$

→ Elastic collisions
→ Inelastic collisions (heat ≡ T ↗)
→ Reactive collisions (loss ≡ n ↘)

Results: Optical shielding


Elastic collisions

Inelastic collisions

Reactive collisions

X.Tie *et al* PRL 125, 153202 **(2020)**

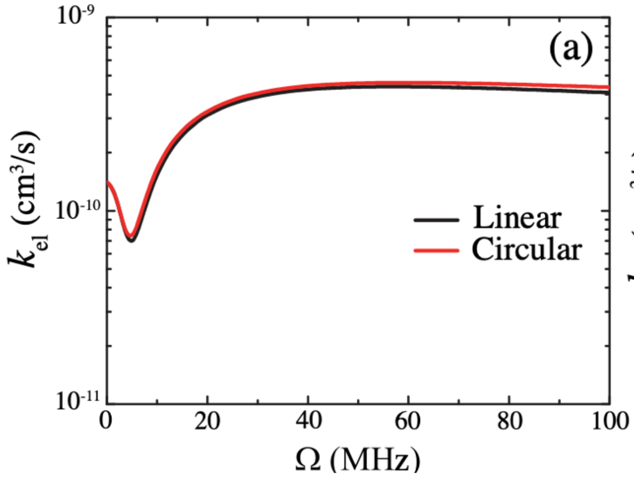
Results: Optical shielding

Higher elastic collision rate

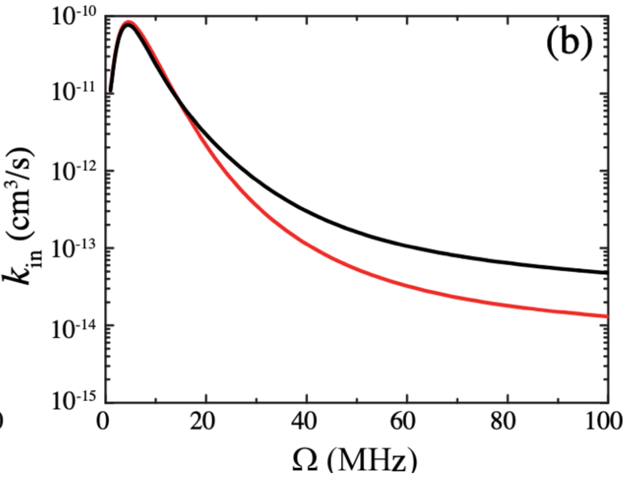
No additional heating

Low losses

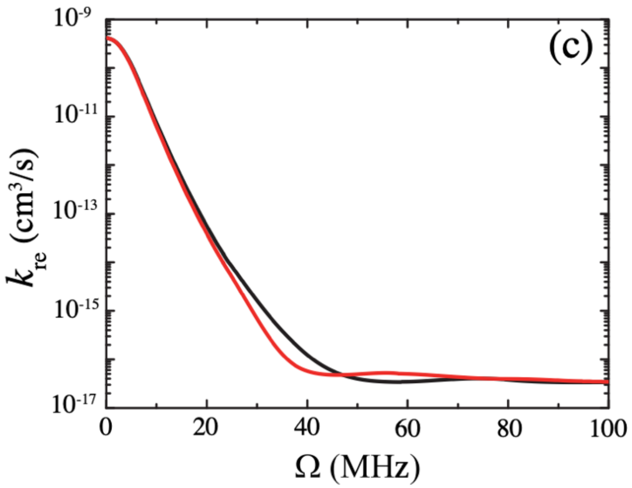
Elastic collisions



Inelastic collisions



Reactive collisions



X.Tie *et al* PRL 125, 153202 **(2020)**

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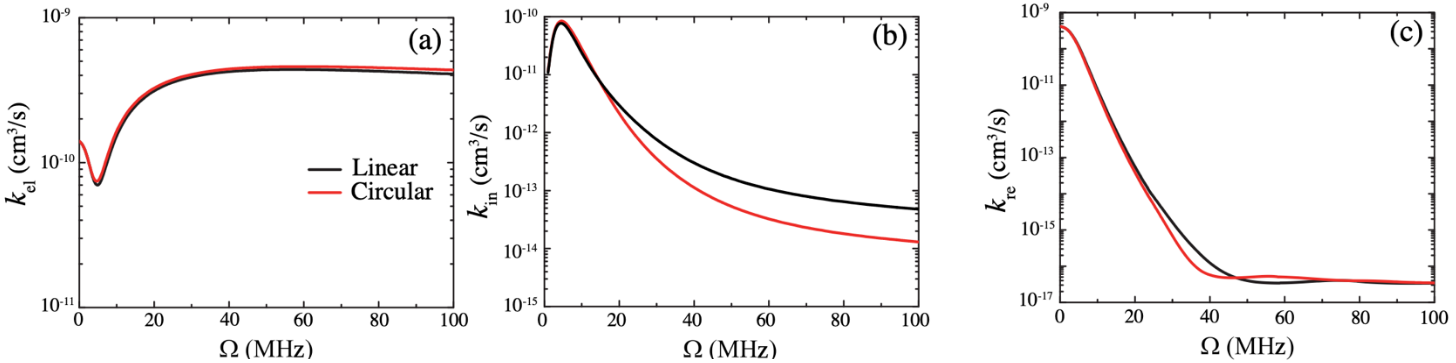
Inelastic collisions

Reactive collisions



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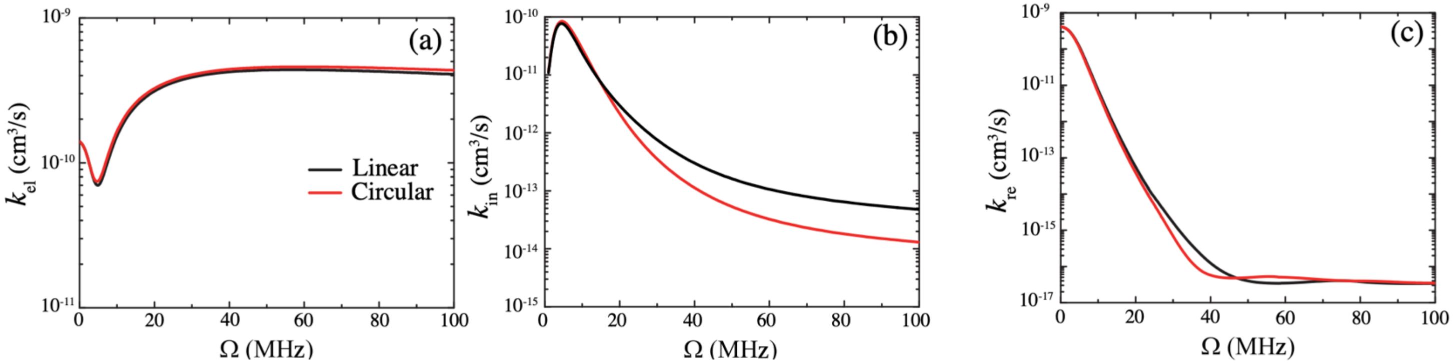
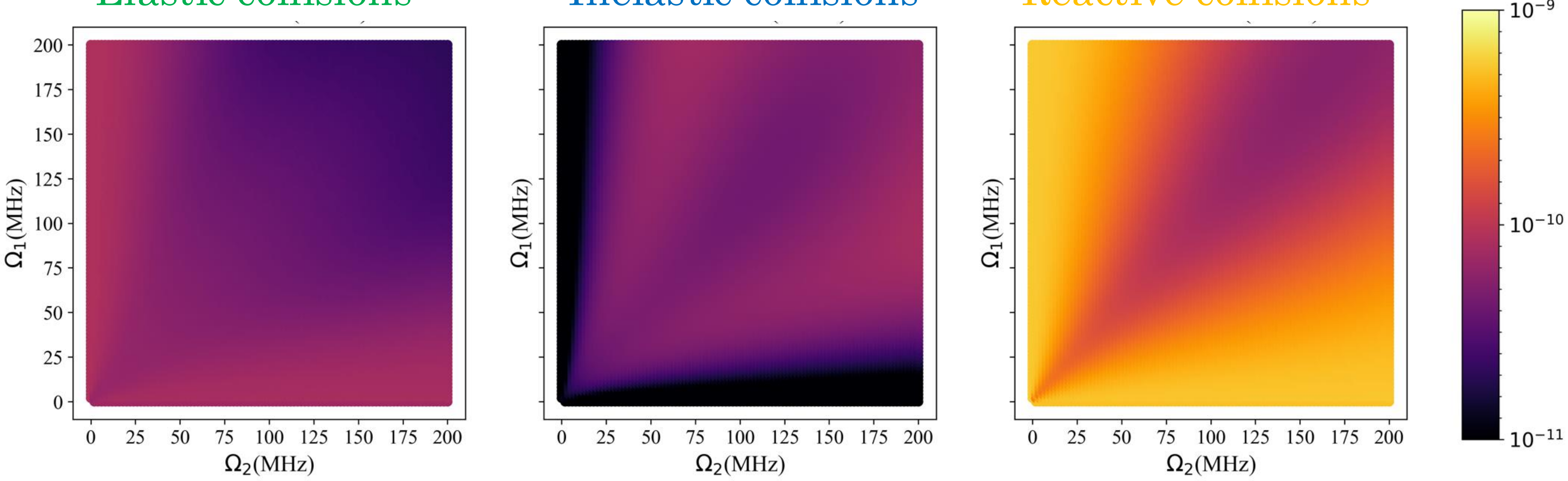


Results: Two photon optical shielding

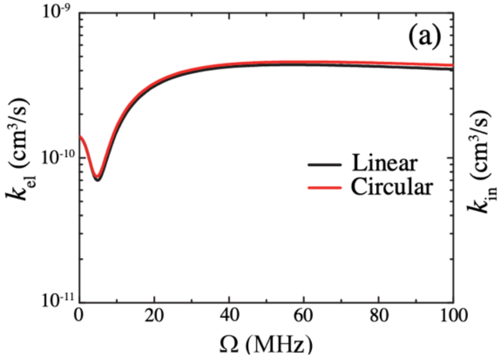
Elastic collisions Inelastic collisions Reactive collisions



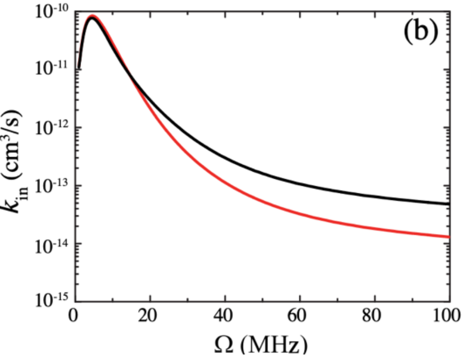
Results: Optical shielding

X.Tie *et al* PRL 125, 153202 **(2020)**

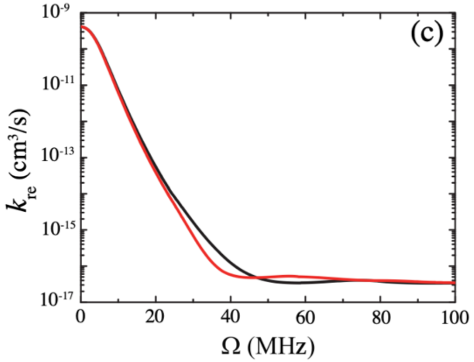
Elastic collisions



Inelastic collisions

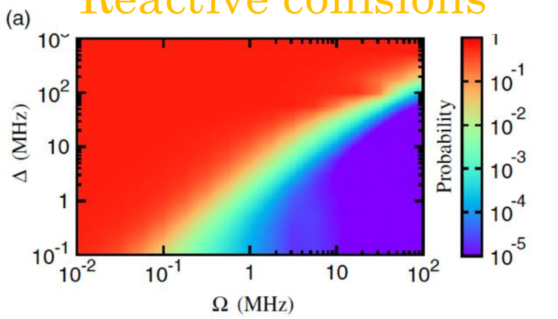


Reactive collisions

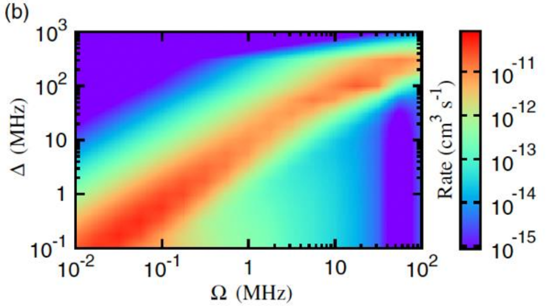


Results: Microwave shielding

Reactive collisions



Inelastic collisions



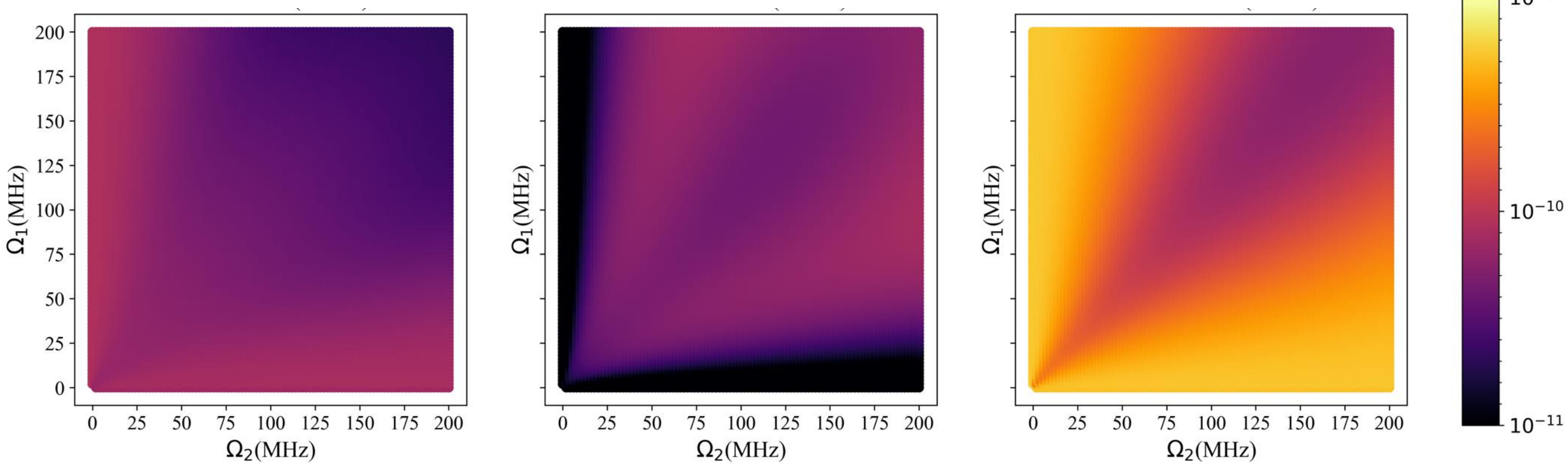
Karman *et al*
PRL 121, 163401 (**2018**)

Results: Two photon optical shielding

Elastic collisions

Inelastic collisions

Reactive collisions



First observation of ground state molecular BEC

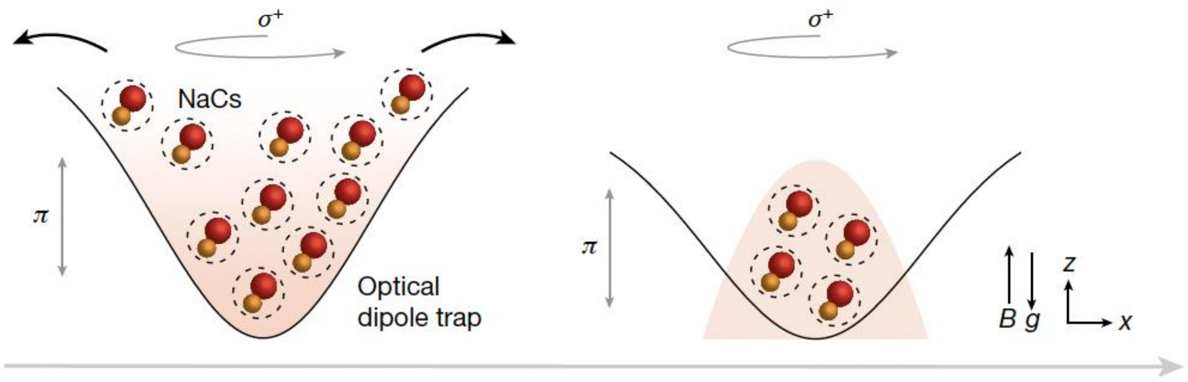
Bigagli *et al* Nature Phys. 19, 1579 **(2024)**

Step 1:
Density

Microwave Shielding ⟶ Stable & Good Density

Step 2:
Temperature

Evaporative cooling



First observation of ground state molecular BEC

Bigagli *et al* Nature Phys. 19, 1579 **(2024)**

Step 1:
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Microwave Shielding → Stable & Good Density

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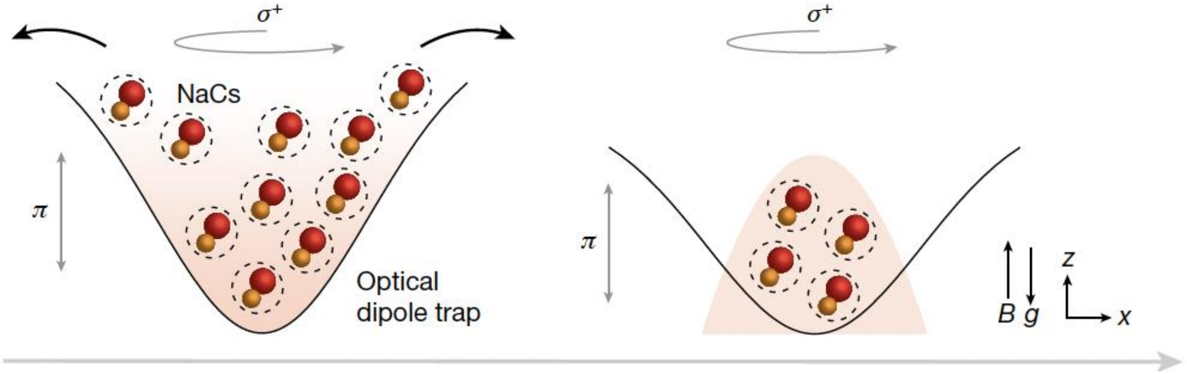
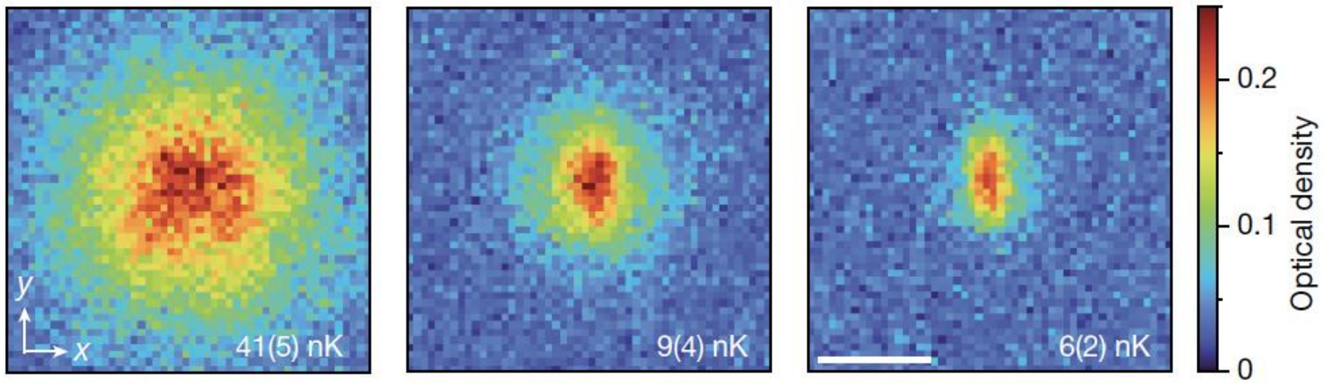


Step 3:



Thermal cloud

Condensed cloud

BEC

Conclusion

- Molecular BEC bring many of the theoretical predictions on dipolar gases into experimental reach.
- Ground state bi-alkali systems suffer from two-body losses.
- Shielding allows to engineer the inrteractions between the molecules to avoid losses.
- Shielding was experimentally proven to be efficient and lead to the formation of the first BEC of GD molecules.

Team and collaborators

LABORATOIRE Aimé Cotton

Théomol
Théorie des molécules froides : structure, dynamique, réactivité

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- Goulven Quéméner
- Nadia Bouloufa
- Olivier Dulieu

Thank you!