Two-photon optical shielding of collisions between ultracold polar molecules

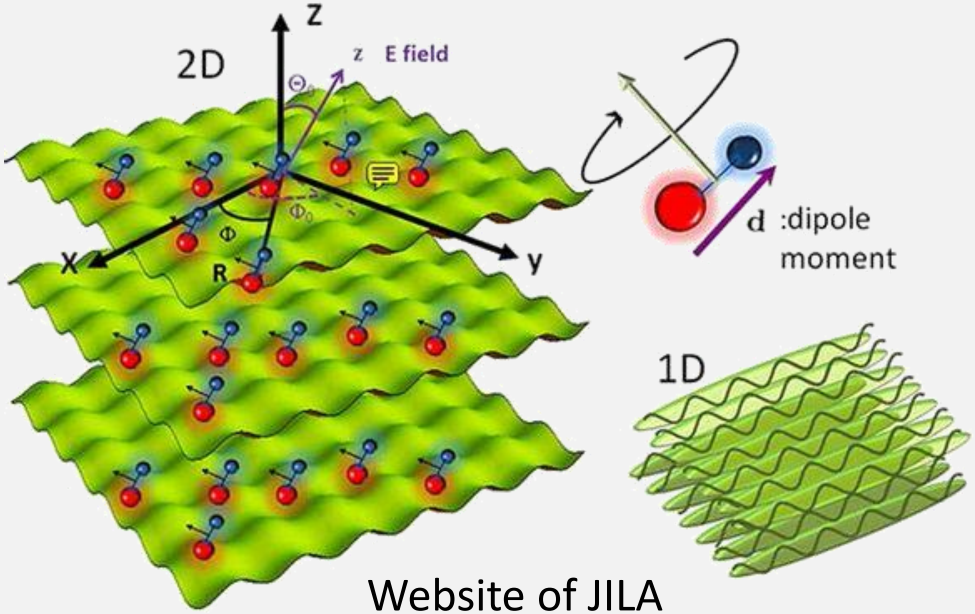
M. Lepers, C. Karam, R. Vexiau, N. Bouloufa-Maafa, O. Dulieu, M. M. z. A. Borgloh, S. Ospelkaus, L. Karpa

Congrès Général de la SFP, July 4, 2023

Ultracold polar molecules

- **Two alkalis**: KRb, RbCs, NaRb, **NaK**, NaCs
- In their **absolute ground state**

- **Anisotropic** and **long-range** interactions
- Quantum **simulation**, **ultracold chemistry** , …



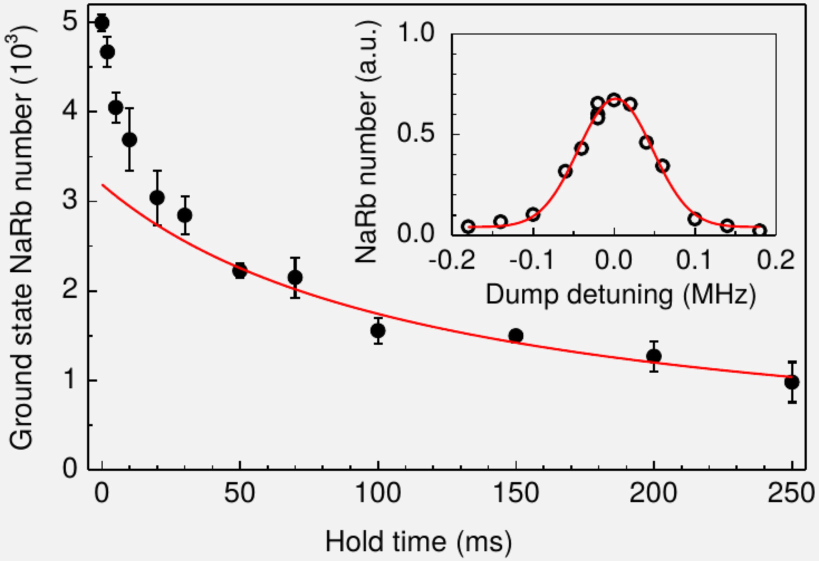
Website of JILA

BUT: limited lifetime

For all molecules !

KRb + KRb ⟶ $K_2 + Rb_2$ (exoenergetic)

NaK + NaK ⟶ $Na_2 + K_2$ (endoenergetic)



PRL **116**, 205303 (2016)

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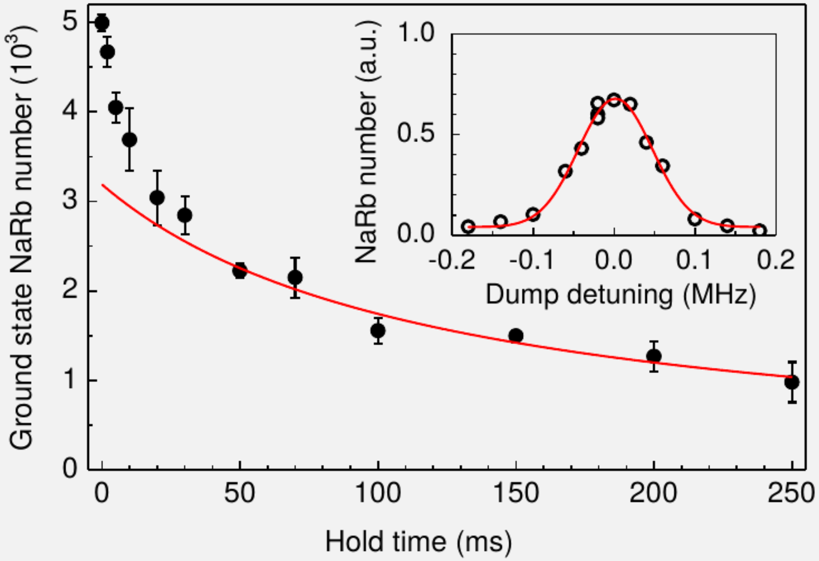
KRb + KRb $\longrightarrow$ **K_2 + Rb_2** (exothermic)

NaK + NaK $\longrightarrow$ **Na_2 + K_2** (endothermic)



PRL **116**, 205303 (2016)

What is the origin of losses ?

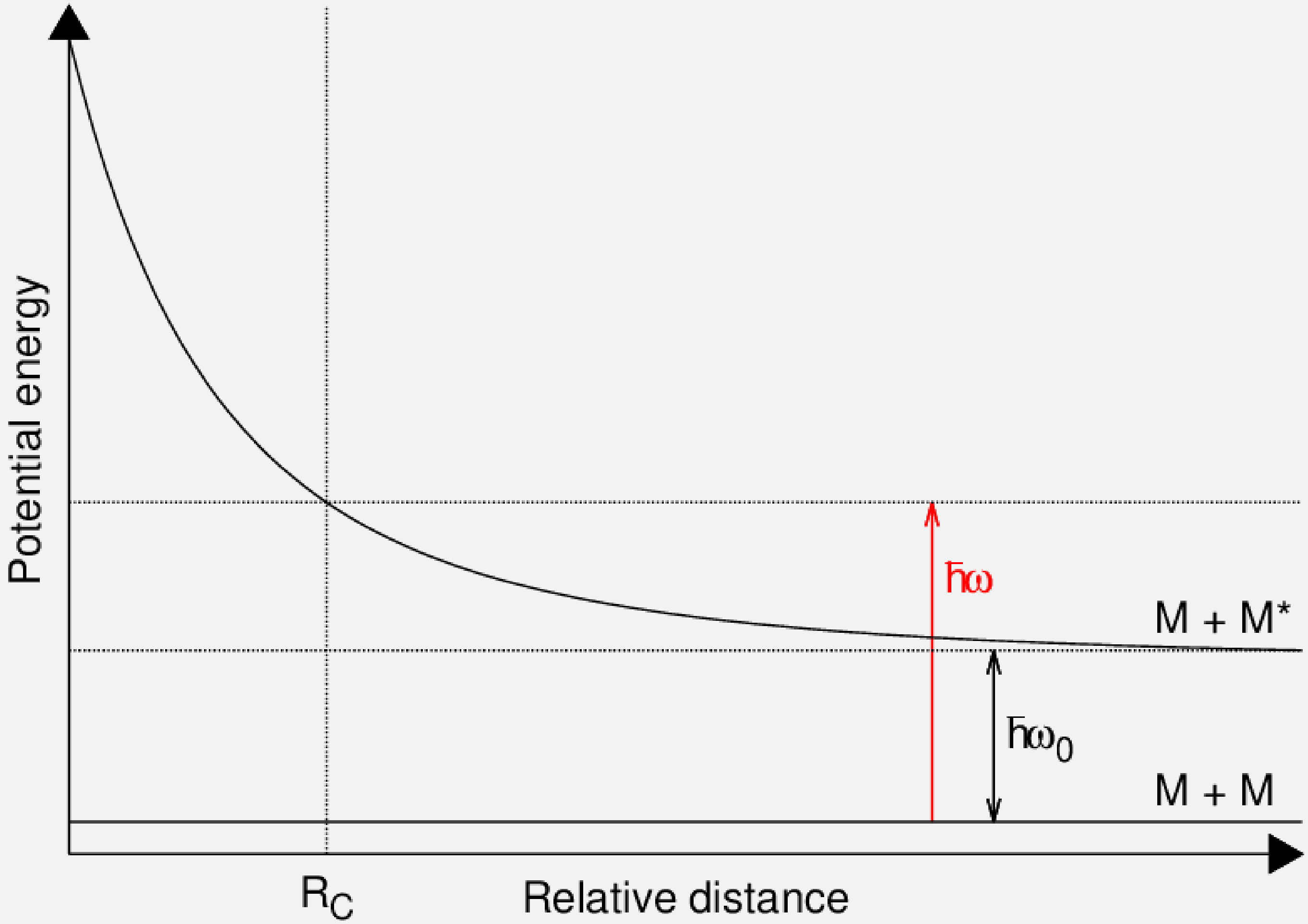
- **Sticky collisions ?**
- **Photoexcitation** of complex by **trapping light ?**

Idea: preventing molecules from getting close to each other and starting a reaction

=> Shielding

Blue shielding : principle

= Engineer repulsive long-range interactions



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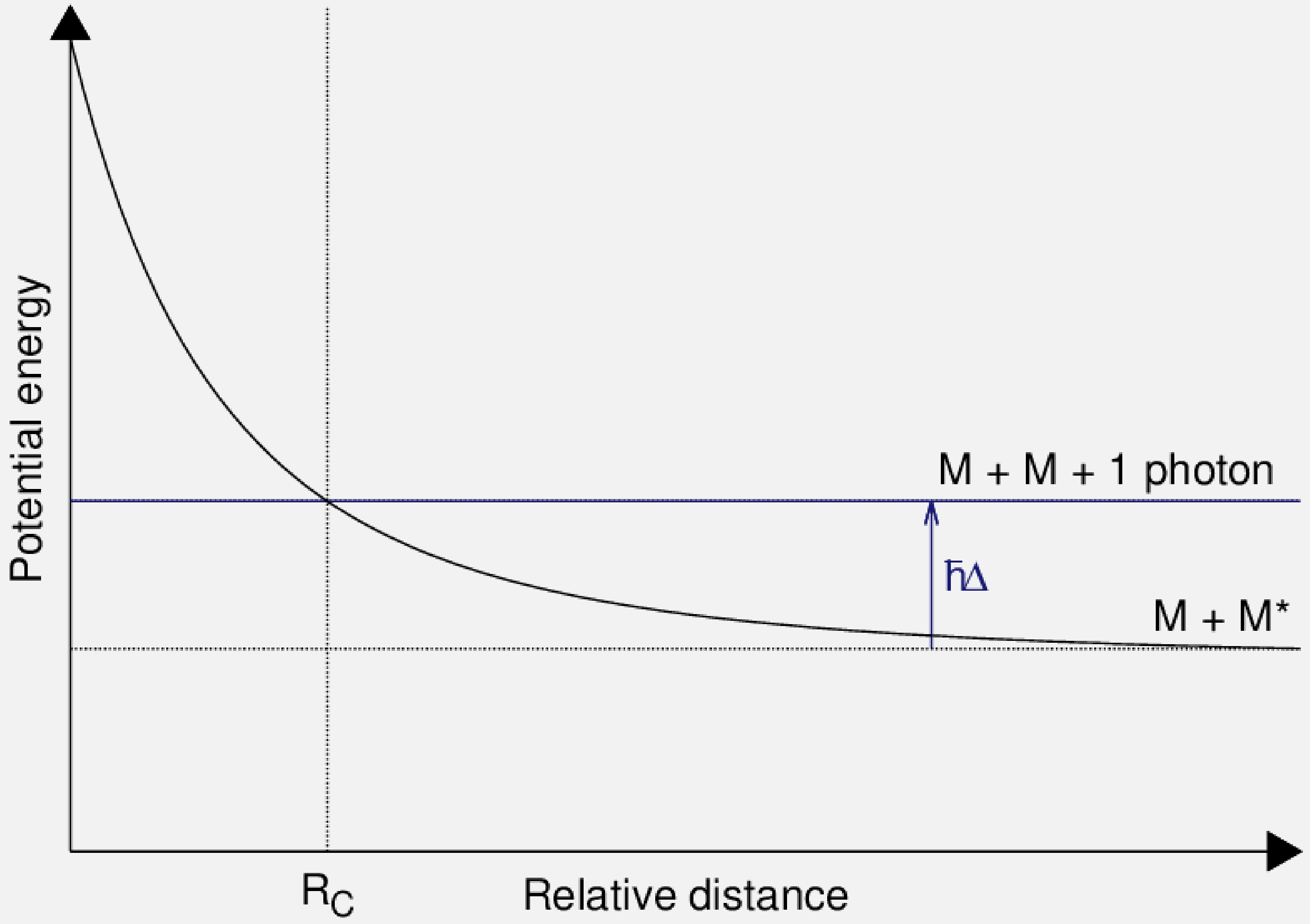

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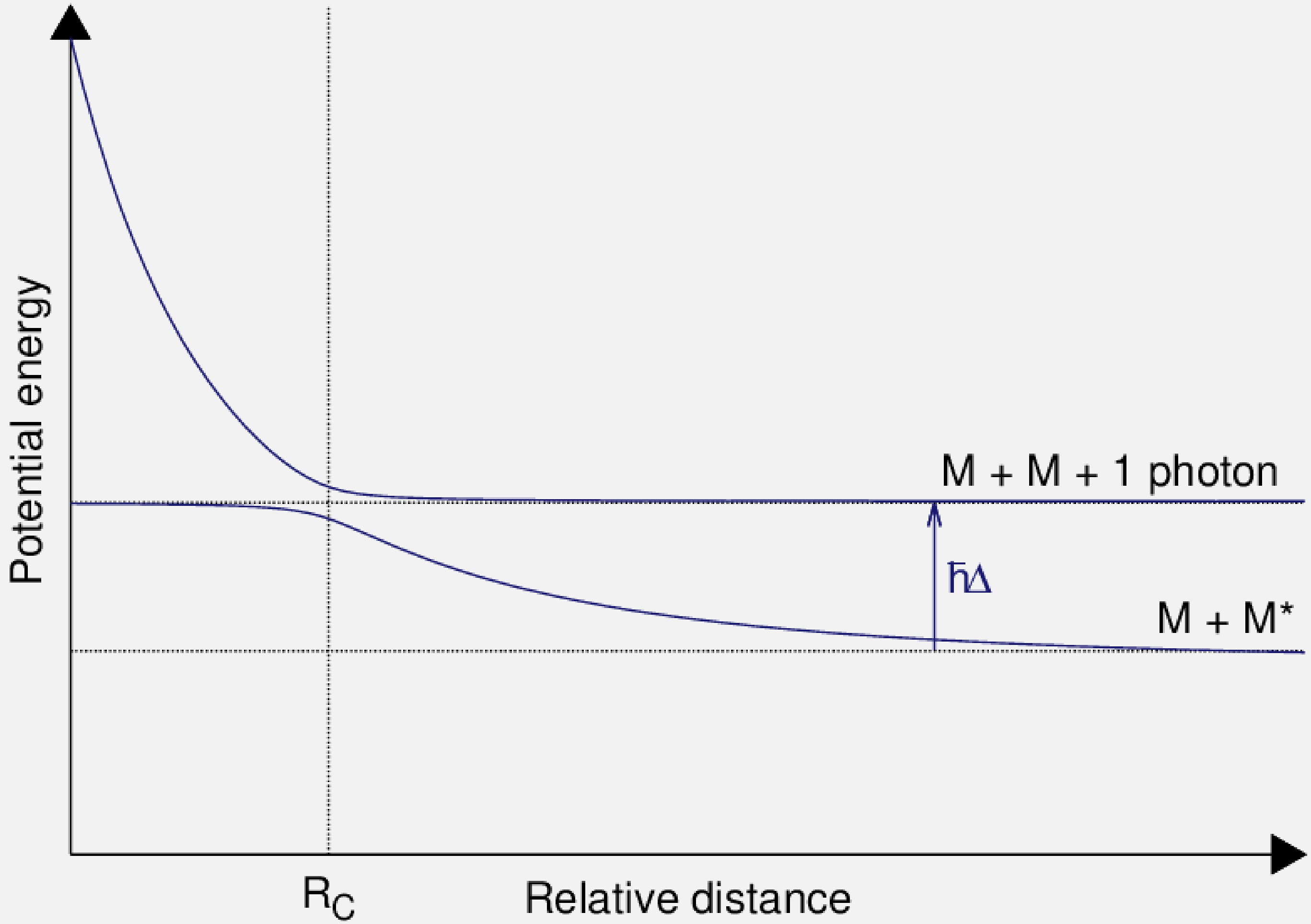

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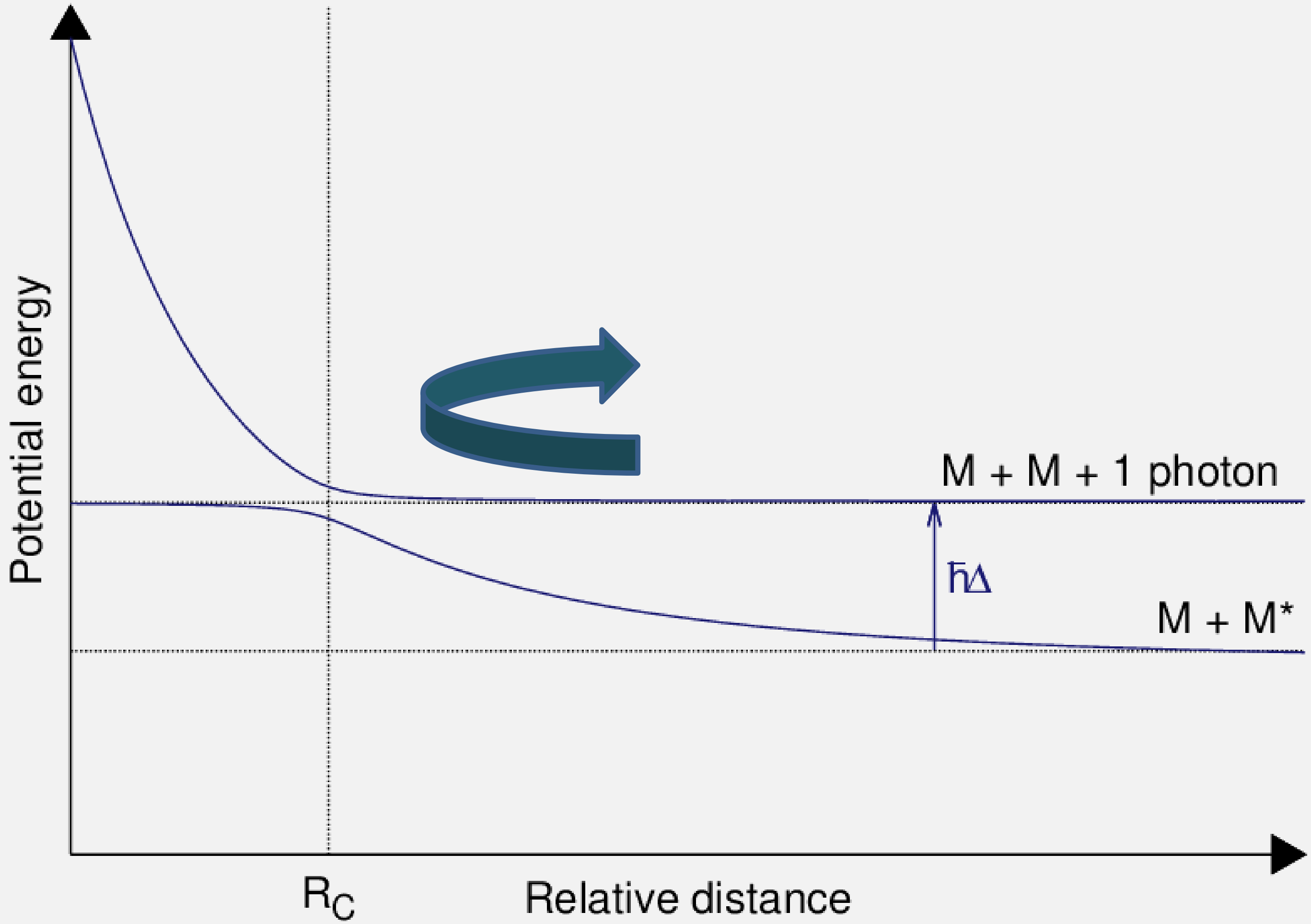
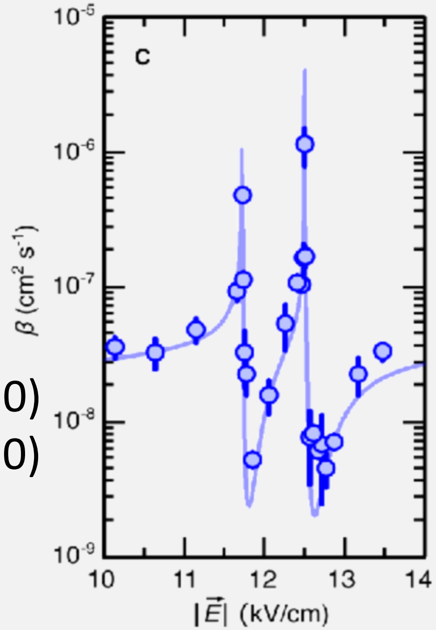


Engineering repulsive interactions using …

- **Static electric field**

Rotational-level mixing creates repulsive van der Waals interaction



JILA team: Science **370** , 1324 (2020)
Nature **588**, 289 (2020)

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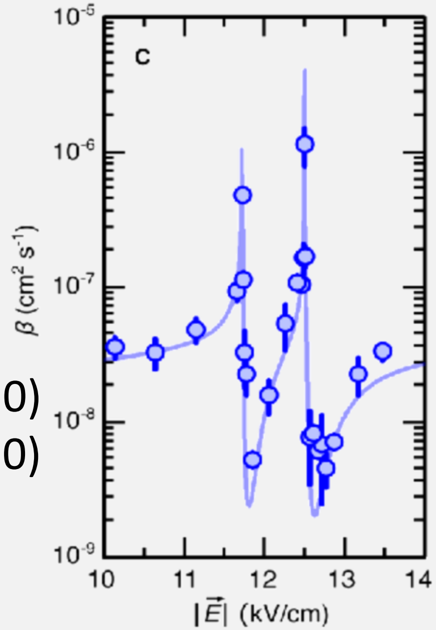

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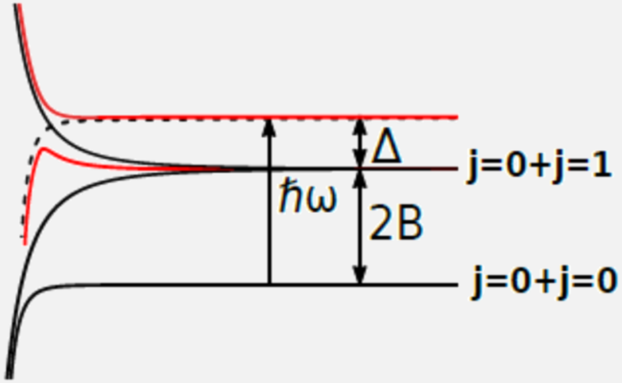
Nature **588**, 289 (2020)

- **Microwave (MW) field**

With respect to purely rotational transition



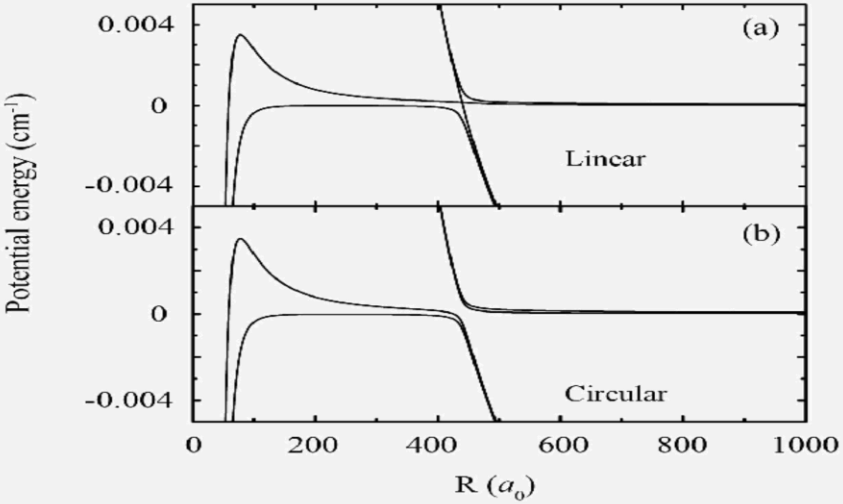
MIT/Harward: Science **373**, 779 (2021)
Garching: Nature **607**, 677 (2022)
Cornell: arxiv:2303.16845 (2023)

Engineering repulsive interactions using …

- **Optical field**

Frequency close to D_2 transition of alkali atoms

Deteriorated by spontaneous emission



Na_2: PRA **51**, 1446 (1995)

Engineering repulsive interactions using …

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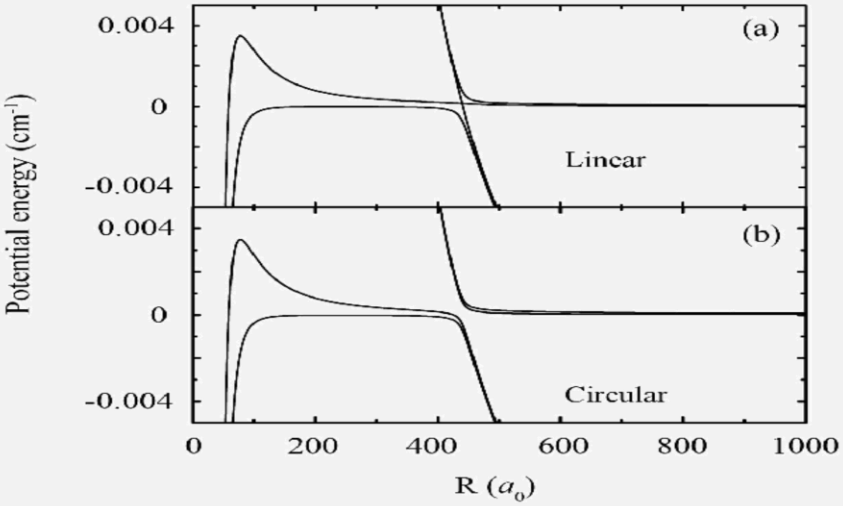


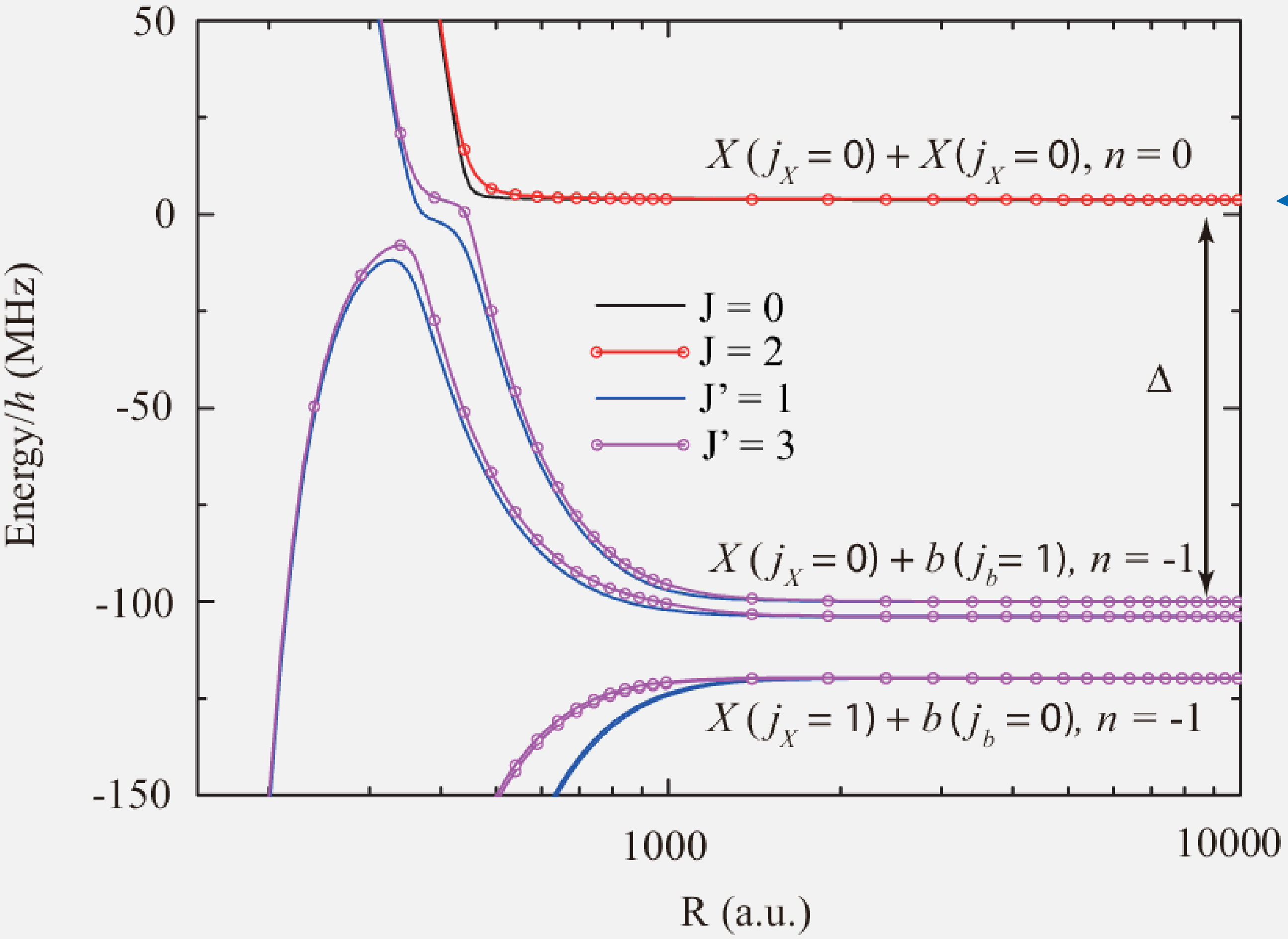
Na_2: PRA **51**, 1446 (1995)

Quasi-forbidden transitions in bialkali molecules

Transition $X^1\Sigma^+ (v_X = 0, j_X = 0) \longrightarrow b^3\Pi_{0+} (v_b = 0, j_b = 1)$

Theory for NaRb: PRL **125**, 153202 (2020)

X ⟶ b tranition in NaRb



Incoming channel

Δ = 100 MHz
Ω = 10 MHz
Linear pola.

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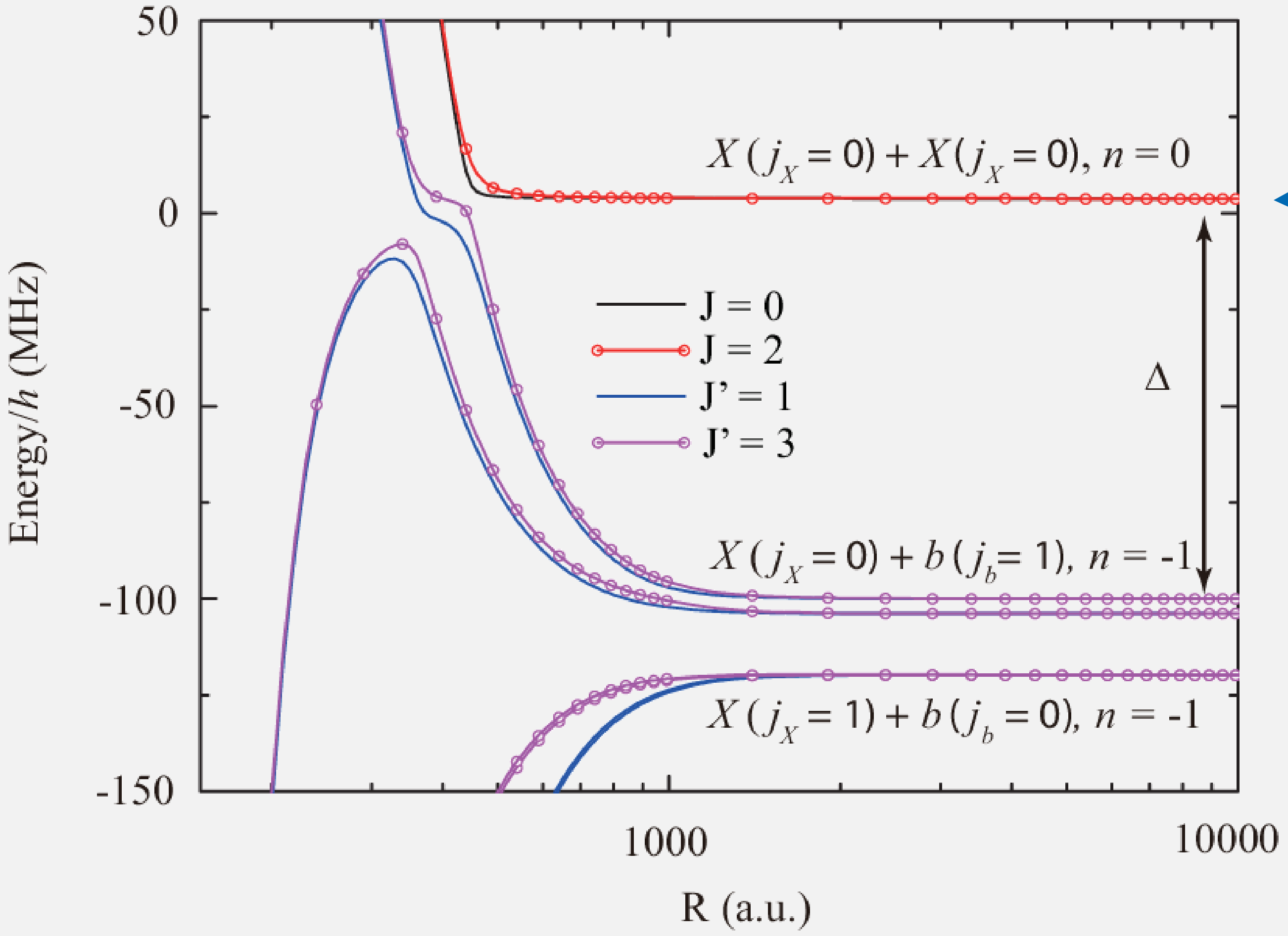

Incoming channel

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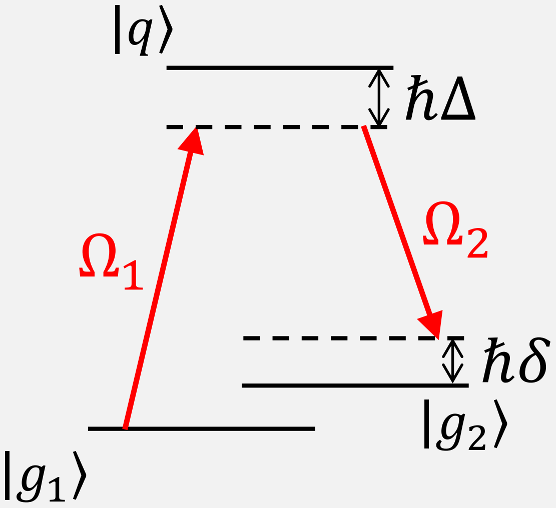
BUT : 1 molecule photon scattering

Idea: applying optical fields without spontaneous emission

=> 2-photon transition

2-photon transition (1 molecule)



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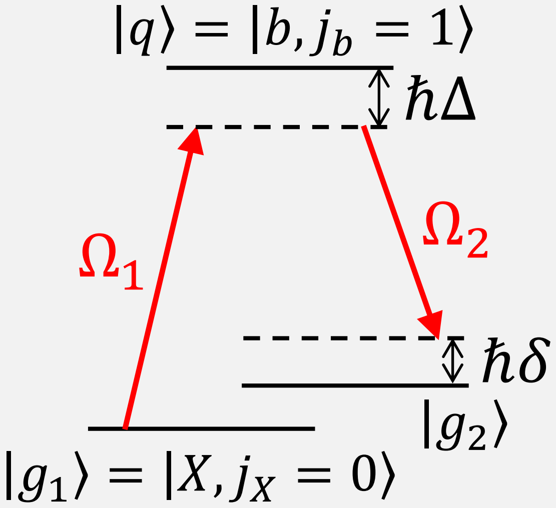

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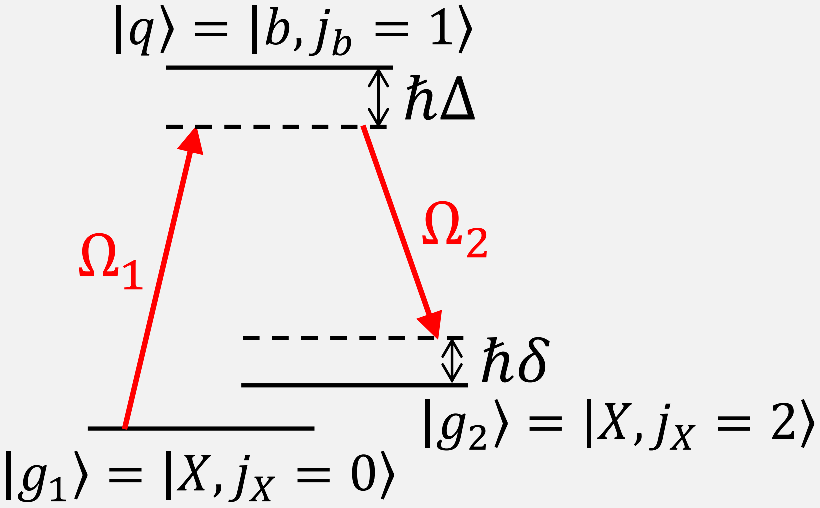

In dressed basis $\{|\tilde{g}_1\rangle, |\tilde{g}_2\rangle, |\tilde{q}\rangle\}$

$$H^I = \hbar \begin{pmatrix} 0 & 0 & \Omega_1/2 \\ 0 & \delta & \Omega_2/2 \\ \Omega_1/2 & \Omega_2/2 & \Delta \end{pmatrix}$$

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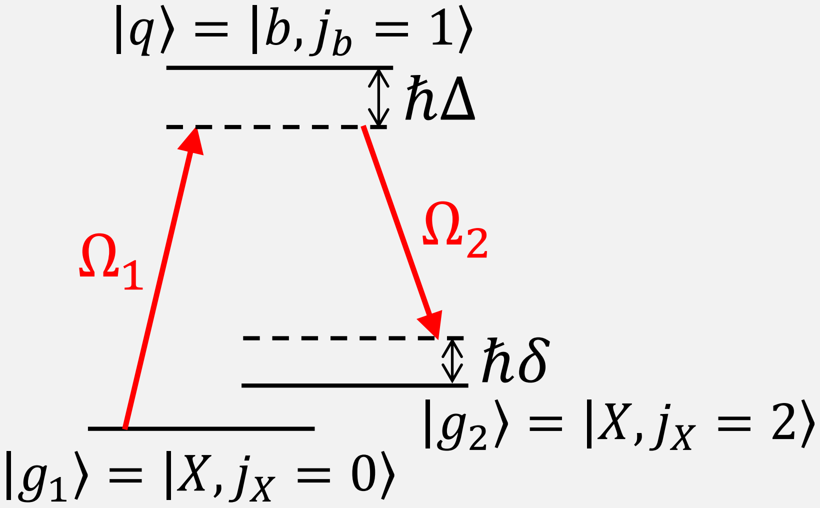

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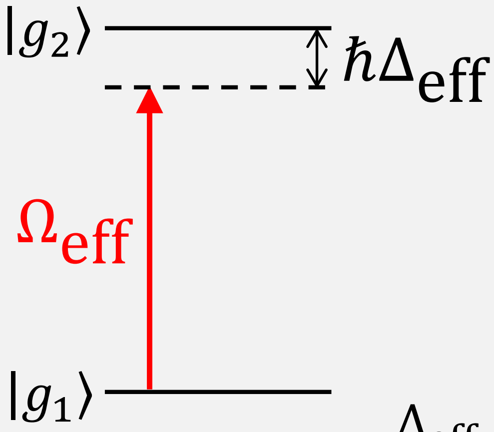
Adiabatic elimination of $|\tilde{q}\rangle$
$\Delta \gg \Omega_1, \Omega_2, \Gamma_q$

$\Downarrow$



$$H^I_{\text{eff}} = \hbar \begin{pmatrix} 0 & -\Omega_{\text{eff}}/2 \\ -\Omega_{\text{eff}}/2 & \Delta_{\text{eff}} \end{pmatrix}$$

$$\Delta_{\text{eff}} = \delta + \frac{\sqrt{\Omega_1^2 - \Omega_2^2}}{4\Delta} ; \Omega_{\text{eff}} = \frac{\Omega_1 \Omega_2}{2\Delta} \text{ with } \delta = 0$$

2 photons & 2 molecules

$$H = T + H_1 + H_2 + V(R) + H_f + H_{m-f}$$

Relative kinetic energy

Molecules 1 & 2

fields

molecule-moelcule

molecules-fields

$$T = T_R + \frac{\hbar^2 \mathbf{L}^2}{2\mu R^2}$$

$$H_i = B_0 \mathbf{J}_i^2$$

$V(R) = V_{\mathrm{dd}}(R) =$ dipole-dipole

$H_{m-f} =$ molec. 1 & 2

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$$H_{LR}(R) = \frac{\hbar^2 \mathbf{L}^2}{2\mu R^2} + B_0(\mathbf{J}_1^2 + \mathbf{J}_2^2) + V_{\rm dd}(R)$$

Diagonalization for each $R \gtrsim 50$ a.u.

$$V_{LR}(R);\ |\psi\rangle = \sum_m \chi_m(R)\,|m\rangle$$

$$\Omega_{\rm eff}(R) \propto \sum_{m,p} \chi_m(R)\chi_p(R)\,\langle m \| T^{(2)} \| p \rangle$$

Basis sets $\{|\boldsymbol{m}\rangle\}$

Lab-frame, fully uncoupled basis: $\lfloor j_i, m_i, j_k, m_k, \ell, m_\ell\rangle$

Basis sets $\{|m\rangle\}$

Lab-frame, fully uncoupled basis: $|j_i, m_i, j_k, m_k, \ell, m_\ell\rangle$

To better account for symmetries, fully-coupled basis

$$|[j_i, j_k], j_{ik}, \ell, J, M\rangle^{(\pm)}$$

- $\vec{\boldsymbol{J}} = \vec{\boldsymbol{J}}_{\boldsymbol{ik}} + \vec{\boldsymbol{L}} = (\vec{\boldsymbol{J}}_{\boldsymbol{1}} + \vec{\boldsymbol{J}}_{\boldsymbol{2}}) + \vec{\boldsymbol{L}}$ = total angular momentum of the complex (without HFS). *M* associated to its **z-projection**
- ℓ = partial wave
- $[j_i, j_k]$ = permutation
- $(\pm)$ = inversion of all coordinates

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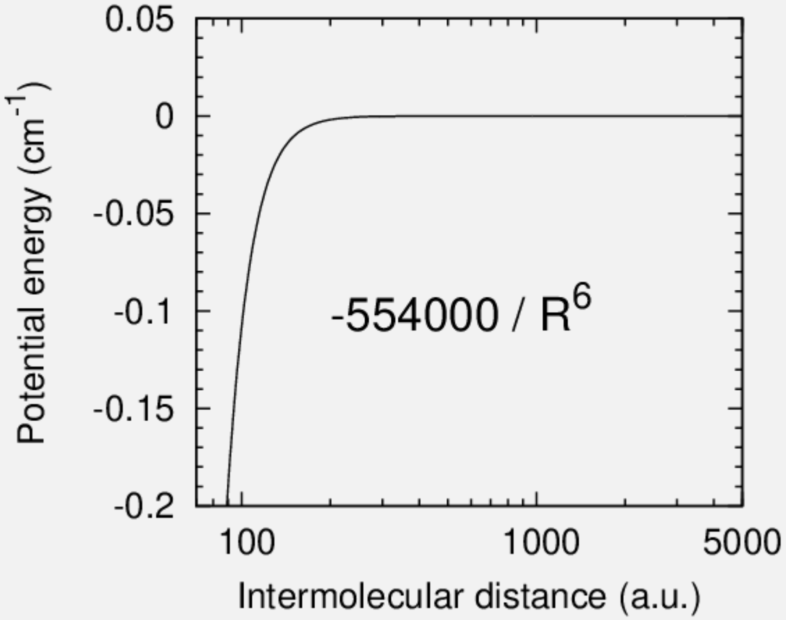
Initial collision channel = 2 ultracold ground-state bosonic molecules

$|m_1\rangle = |[0,0], 0,0,0,0\rangle^{(+)}$



Selection rules

Quant. nber	Dipole-dipole	Raman
$[\Delta j_i, \Delta j_k]$	$[\pm 1, \pm 1]$; $[\mp 1, \pm 1]$	$[0, \pm 2]$
$\Delta \ell$	$0 \text{ or } \pm 2^*$	0
ΔJ	0^*	$0 \text{ or } \pm 1 \text{ or } \pm 2^*$
ΔM	0	0, if = polarizations
parity	$\pm \leftrightarrow \pm \text{ or } \mp \leftrightarrow \mp$	$\pm \leftrightarrow \pm \text{ or } \mp \leftrightarrow \mp$

* $0 \nleftrightarrow 0$, and $1/2 \nleftrightarrow 1/2$ for ΔJ

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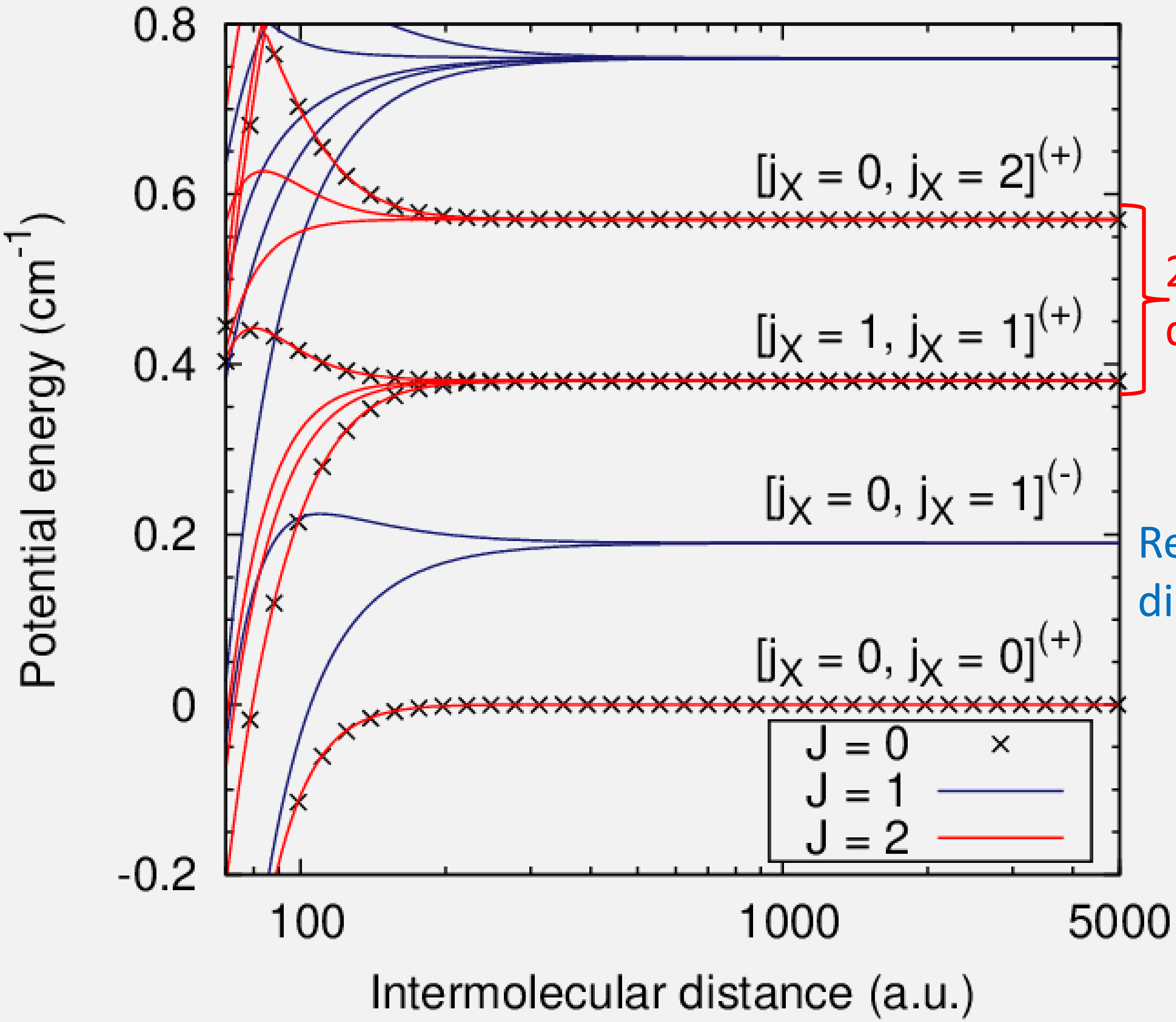
$$|m_1\rangle = |[j_X = 0, j_X = 0], j_{ik} = 0, \ell = 0, J = 0, M = 0\rangle^{(+)}$$

$$\begin{array}{l} |[1,1], 0 \text{ or } 2, 2, 2, 0\rangle^{(+)} \\ |[1,3], 0 \text{ or } 2, 2, 2, 0\rangle^{(+)} \end{array} \longleftrightarrow \boldsymbol{|m_2\rangle = |[0,2], 2, 0, 2, 0\rangle^{(+)}}$$

Results for NaK

Long-range potential energy curves

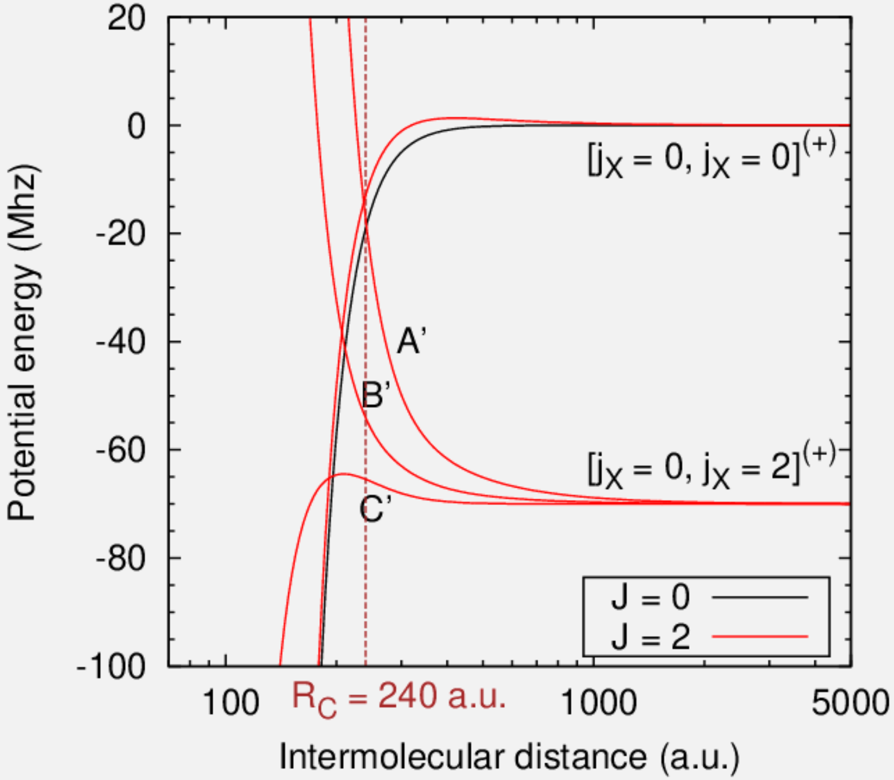


2nd-order dipole-dipole $\propto R^{-6}$

Resonant dipole-dipole $\propto R^{-3}$

Long-range potential energy curves

$\Delta_{\text{eff}} = 70$ MHz



A': $D + \frac{17{,}29}{2\mu R^2} + \frac{1{,}045\times 10^6}{R^6}$ (in a.u.)

B': $D + \frac{6{,}668}{2\mu R^2} + \frac{2{,}711\times 10^5}{R^6}$ (in a.u.)

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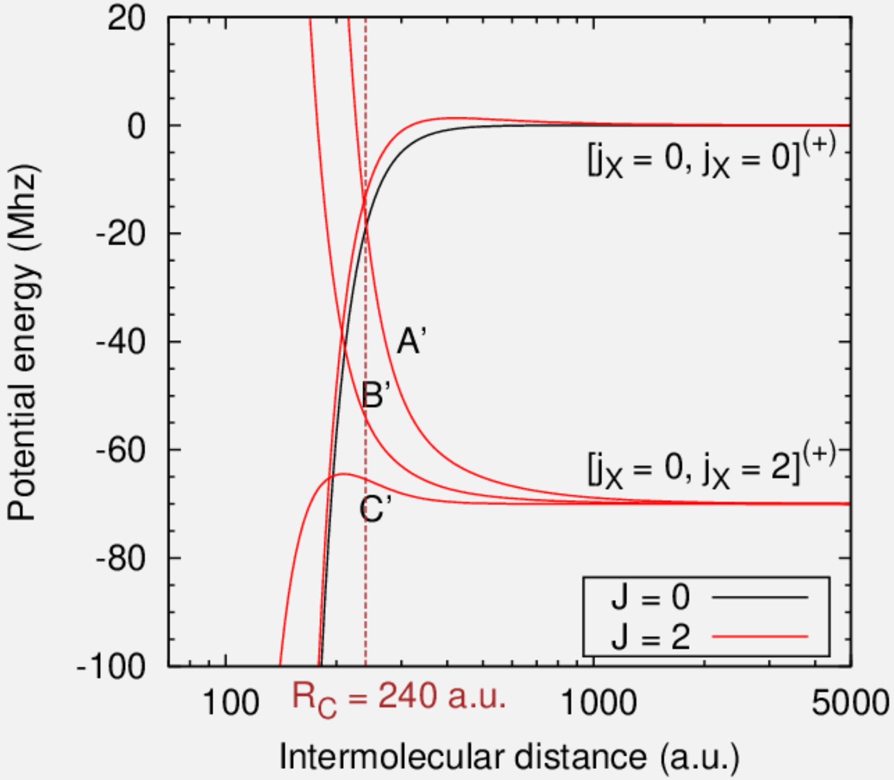

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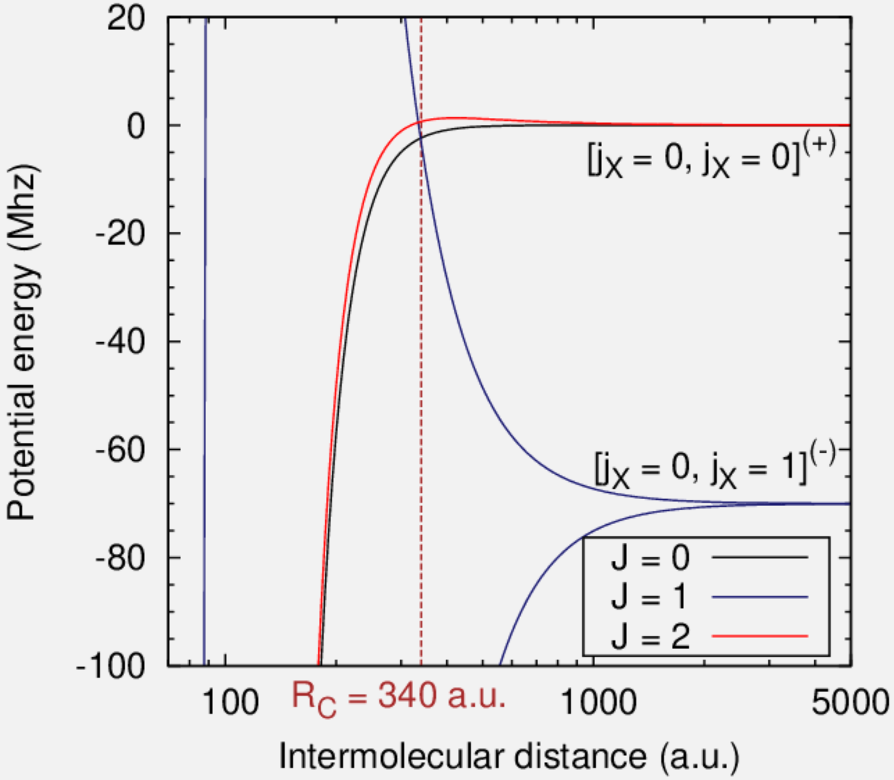

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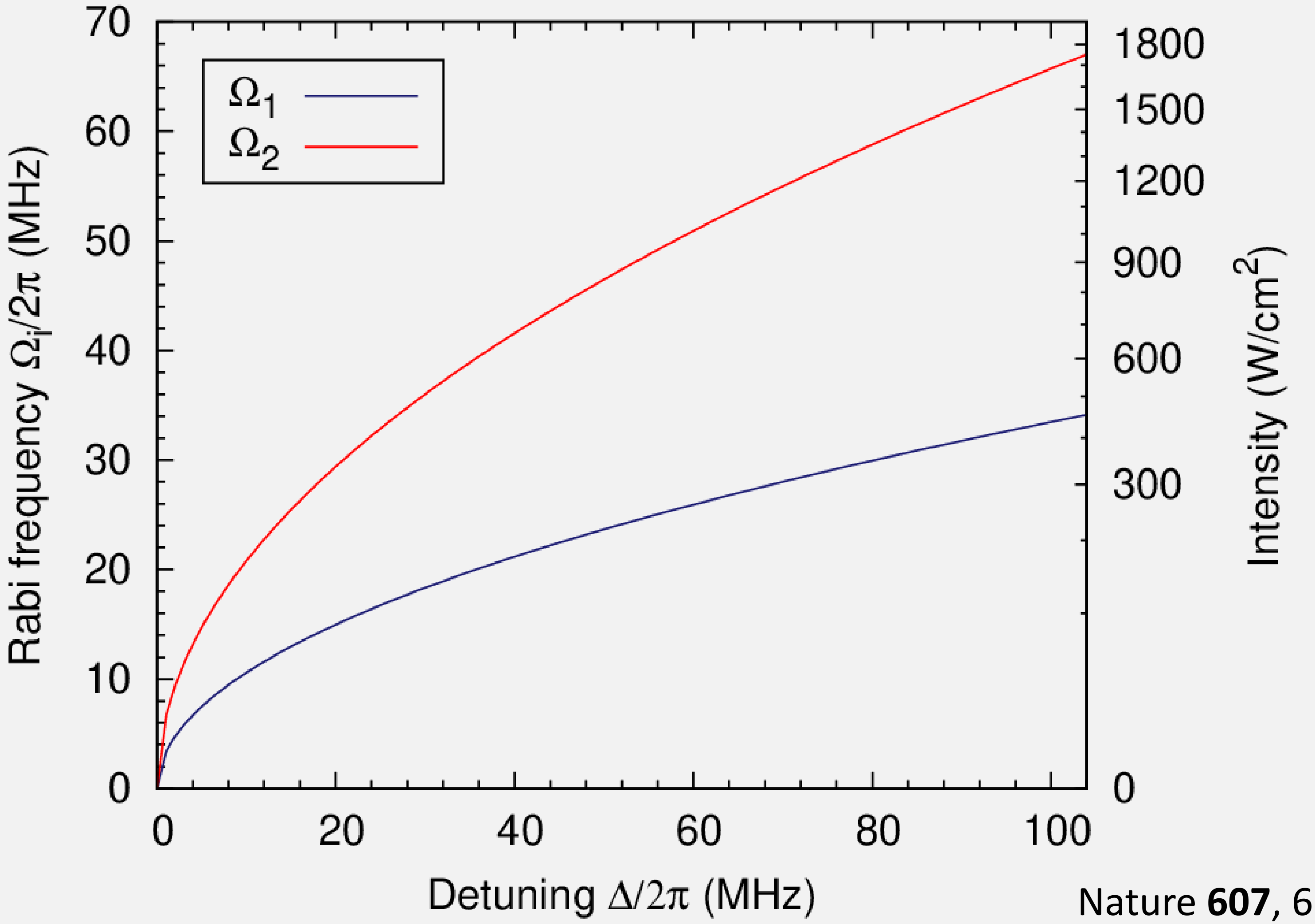
PRL **125**, 163402 (2018)

Eigenvector components at $\mathrm{R_C}$

Basis vector	**Potential curve**	$R_C = 240$ a.u. $\Delta_{\mathrm{eff}} = 70$ **MHz**	$R_C = 170$ a.u. $\Delta_{\mathrm{eff}} = 500$ **MHz**
$\vert m_1\rangle$ $= \vert[0,0],0,0,0,0\rangle^{(+)}$	$\vert g_1\rangle$ = init.	99.95 %	98.61 %
$\vert \boldsymbol{m_2}\rangle$ $= \vert[\mathbf{0},\mathbf{2}],\mathbf{2},\mathbf{0},\mathbf{2},\mathbf{0}\rangle^{(+)}$	$\vert g_2\rangle$ = A’	10.90 %	16.16 %
	$\vert g_2\rangle$ = B’	9.80 %	32.11 %
	$\vert g_2\rangle$ = C’	78.88 %	49.26 %

Physical parameters

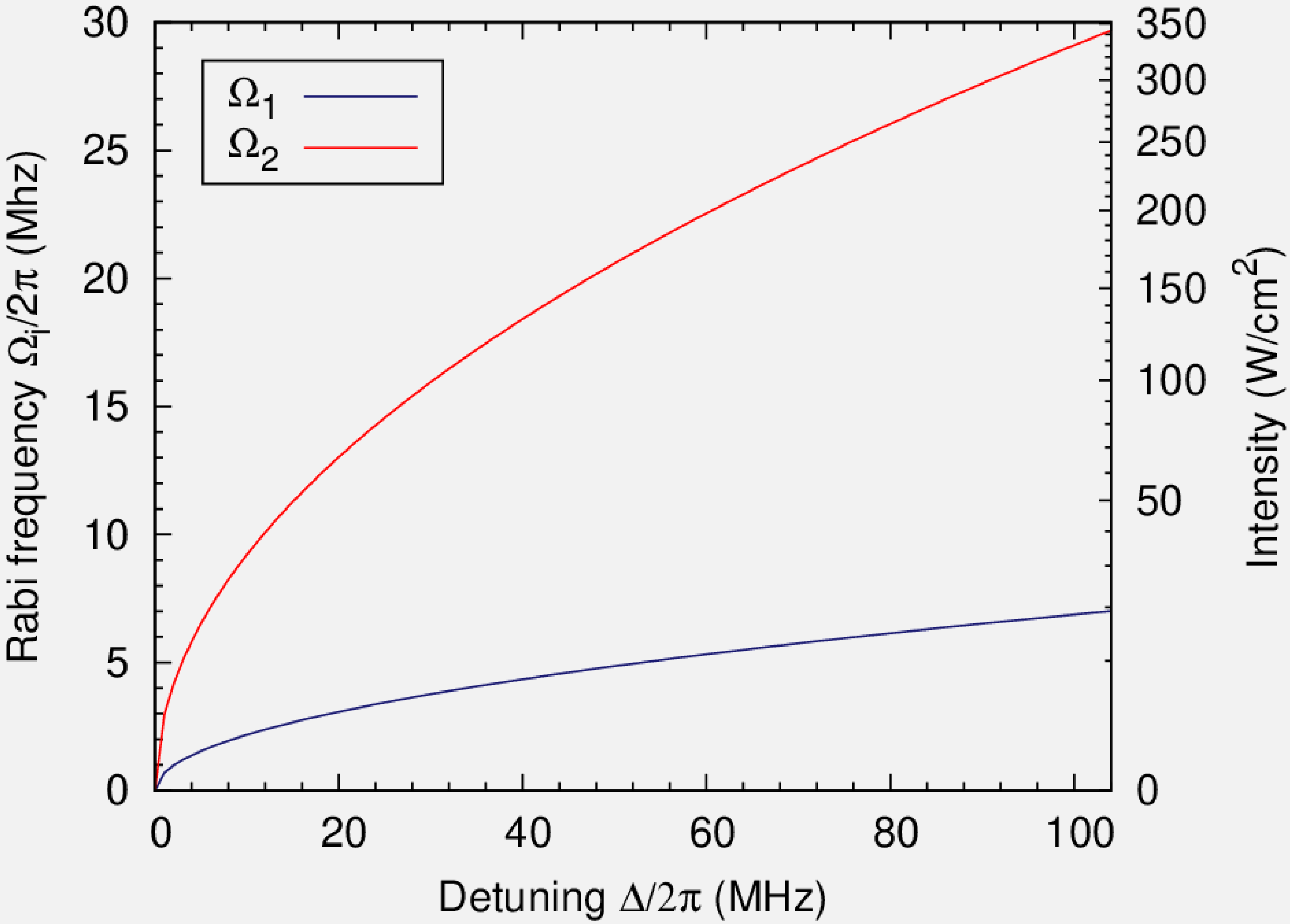
$\Delta_{\text{eff}} = 8$ MHz and $\Omega_{\text{eff}} = 11$ MHz



Nature **607**, 677 (2022)

Physical parameters

$\Delta_{\text{eff}} = 2$ MHz and $\Omega_{\text{eff}} = 1$ MHz



Conclusion

- Possibility of two-photon optical shielding
- No spontaneous emission or photon scattering
- « Mapping » on one-photon shielding
- Applicable to other molecules than NaK
- Possible use of other electronically excited states (A, B)
  - Full scattering calculations

Phys. Rev. Research, accepted (2023)

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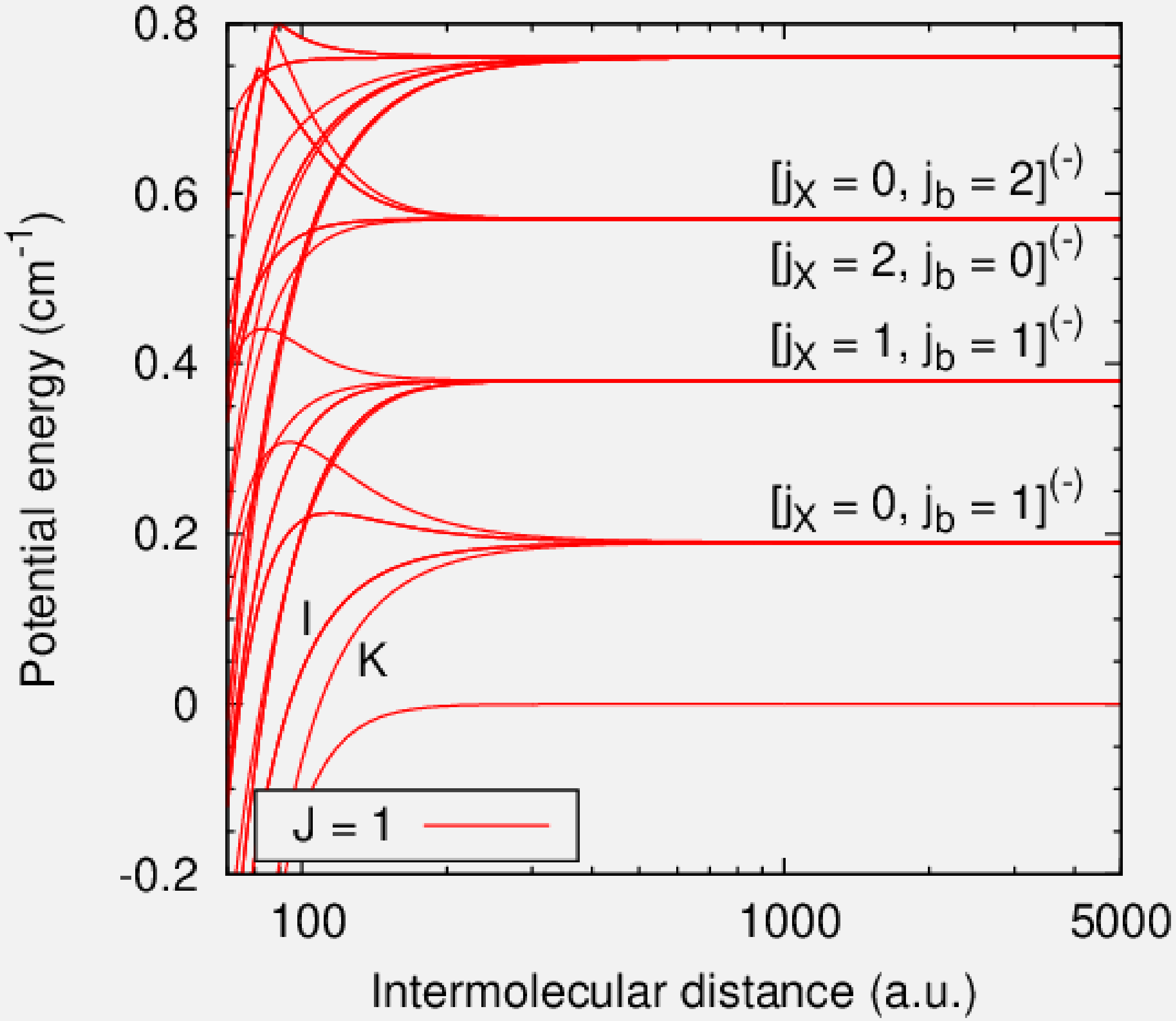
Thank you!

Electronically-excited PECs



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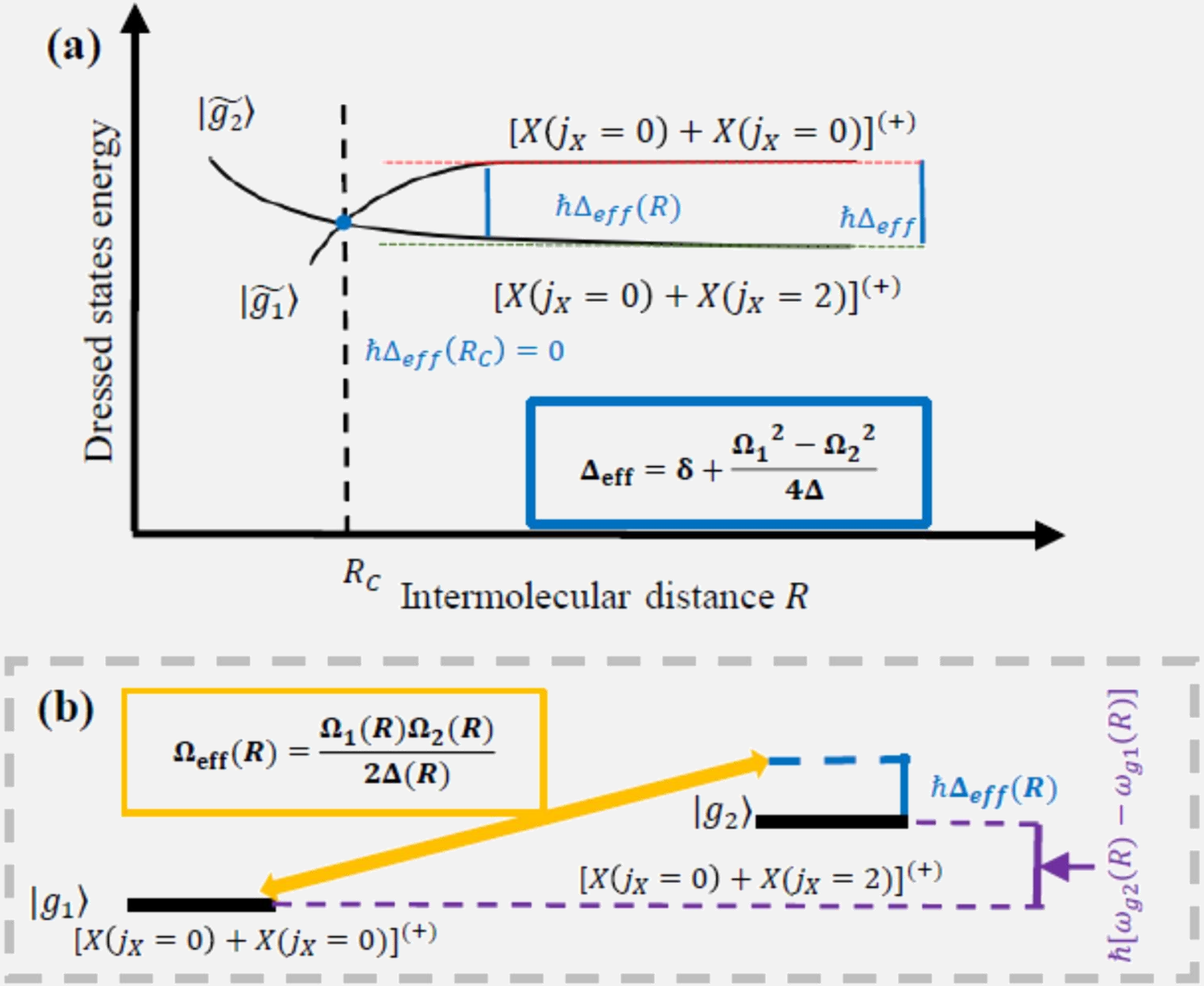
	$\lvert g_2\rangle =$ B’	9.80 %	32.11 %
	$\lvert g_2\rangle =$ C’	78.88 %	49.26 %
$\lvert[X,0,b,1],1,0,1,1\rangle^{(-)}$	$\lvert q\rangle =$ I	33.06 %	33.17 %
	$\lvert q\rangle =$ K	16.74 %	16.68 %

Scheme of energy levels

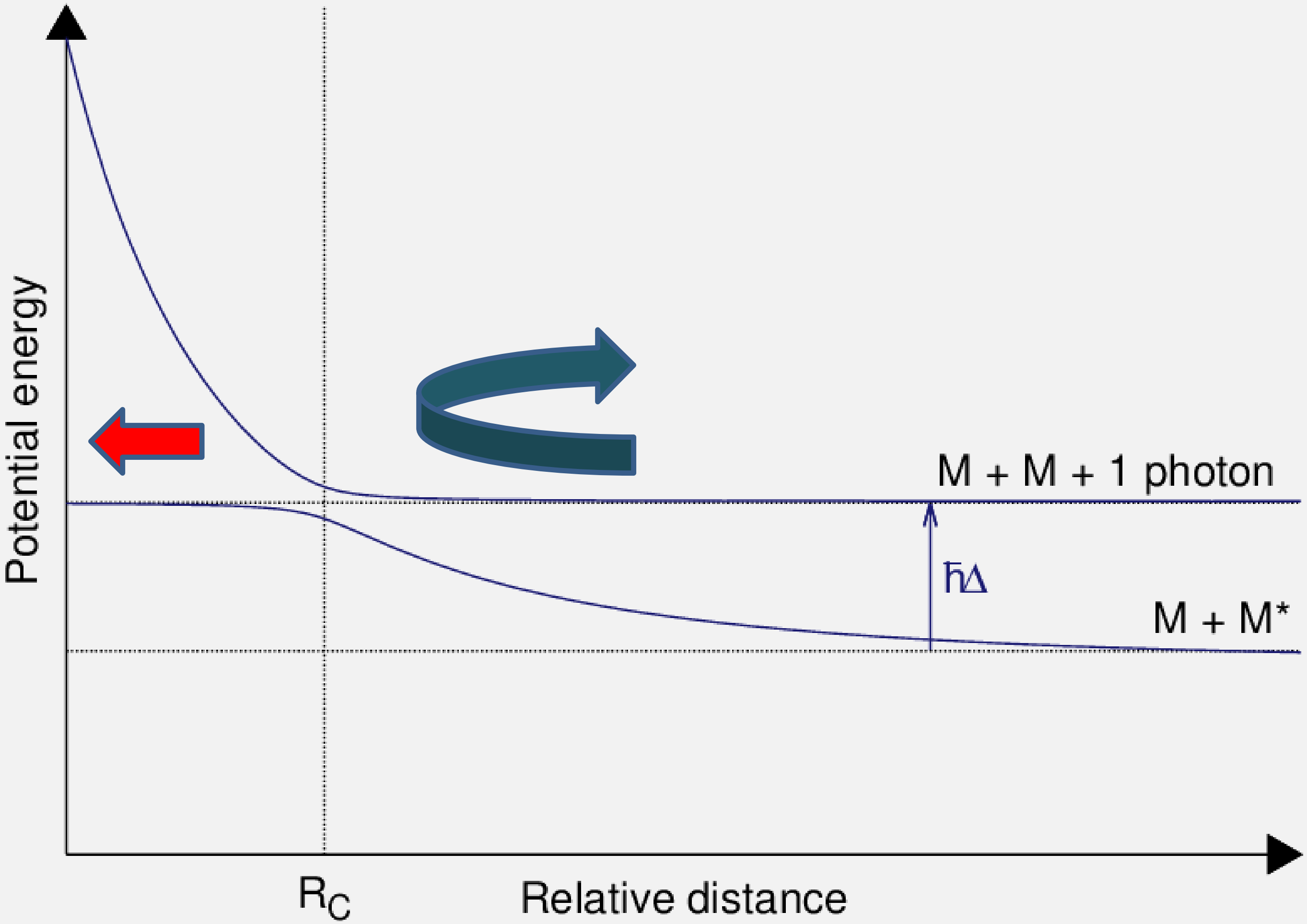


$$\Delta_{\text{eff}}=\delta+\frac{\Omega_1{}^2-\Omega_2{}^2}{4\Delta}$$

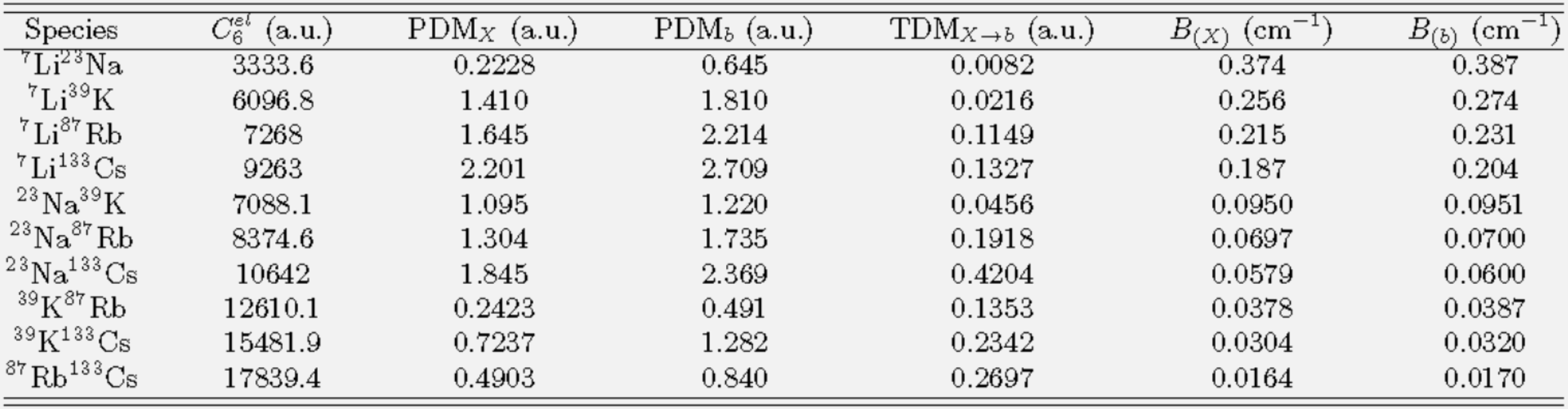
$$\Omega_{\text{eff}}(R)=\frac{\Omega_1(R)\Omega_2(R)}{2\Delta(R)}$$

Blue shielding : principle

= Engineer repulsive long-range interactions

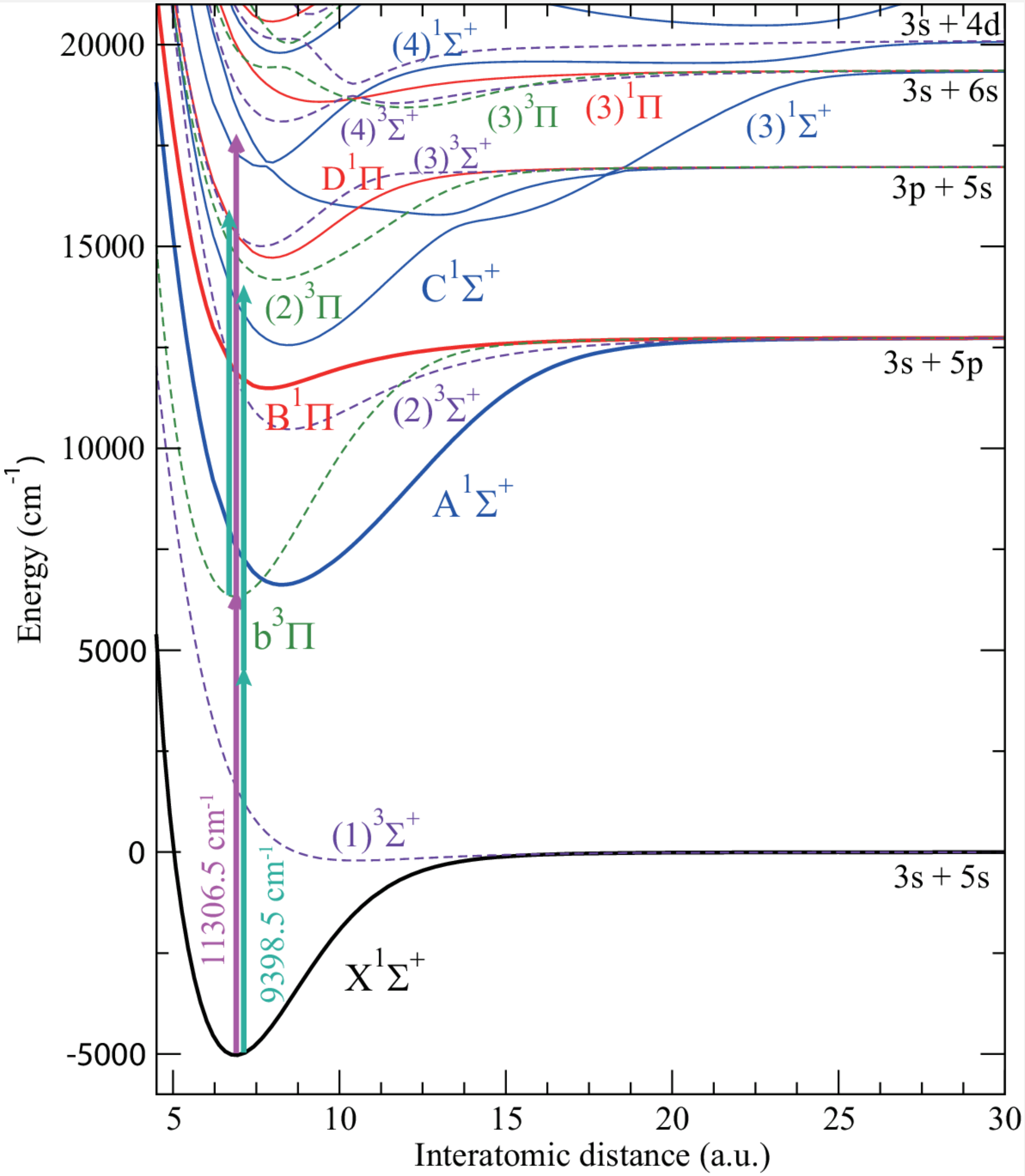


Molecular data

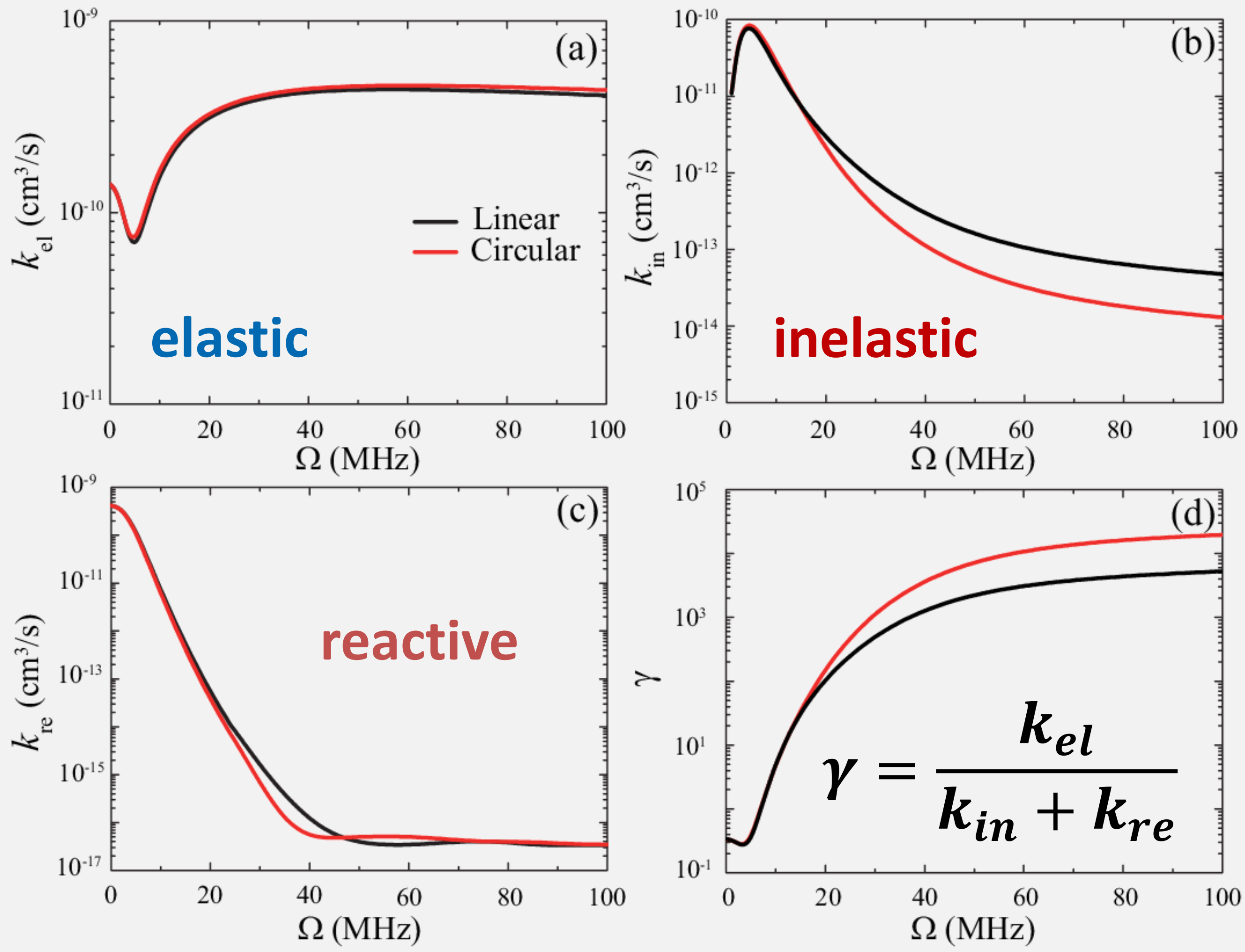


Species	C_6^{el} (a.u.)	PDM$_X$ (a.u.)	PDM$_b$ (a.u.)	TDM$_{X\to b}$ (a.u.)	$B_{(X)}$ (cm^{-1})	$B_{(b)}$ (cm^{-1})
^{7}Li^{23}Na	3333.6	0.2228	0.645	0.0082	0.374	0.387
^{7}Li^{39}K	6096.8	1.410	1.810	0.0216	0.256	0.274
^{7}Li^{87}Rb	7268	1.645	2.214	0.1149	0.215	0.231
^{7}Li^{133}Cs	9263	2.201	2.709	0.1327	0.187	0.204
^{23}Na^{39}K	7088.1	1.095	1.220	0.0456	0.0950	0.0951
^{23}Na^{87}Rb	8374.6	1.304	1.735	0.1918	0.0697	0.0700
^{23}Na^{133}Cs	10642	1.845	2.369	0.4204	0.0579	0.0600
^{39}K^{87}Rb	12610.1	0.2423	0.491	0.1353	0.0378	0.0387
^{39}K^{133}Cs	15481.9	0.7237	1.282	0.2342	0.0304	0.0320
^{87}Rb^{133}Cs	17839.4	0.4903	0.840	0.2697	0.0164	0.0170

NaRb with shielding and trapping lights



X ⟶ b tranition in NaRb



$$\gamma = \frac{k_{el}}{k_{in} + k_{re}}$$

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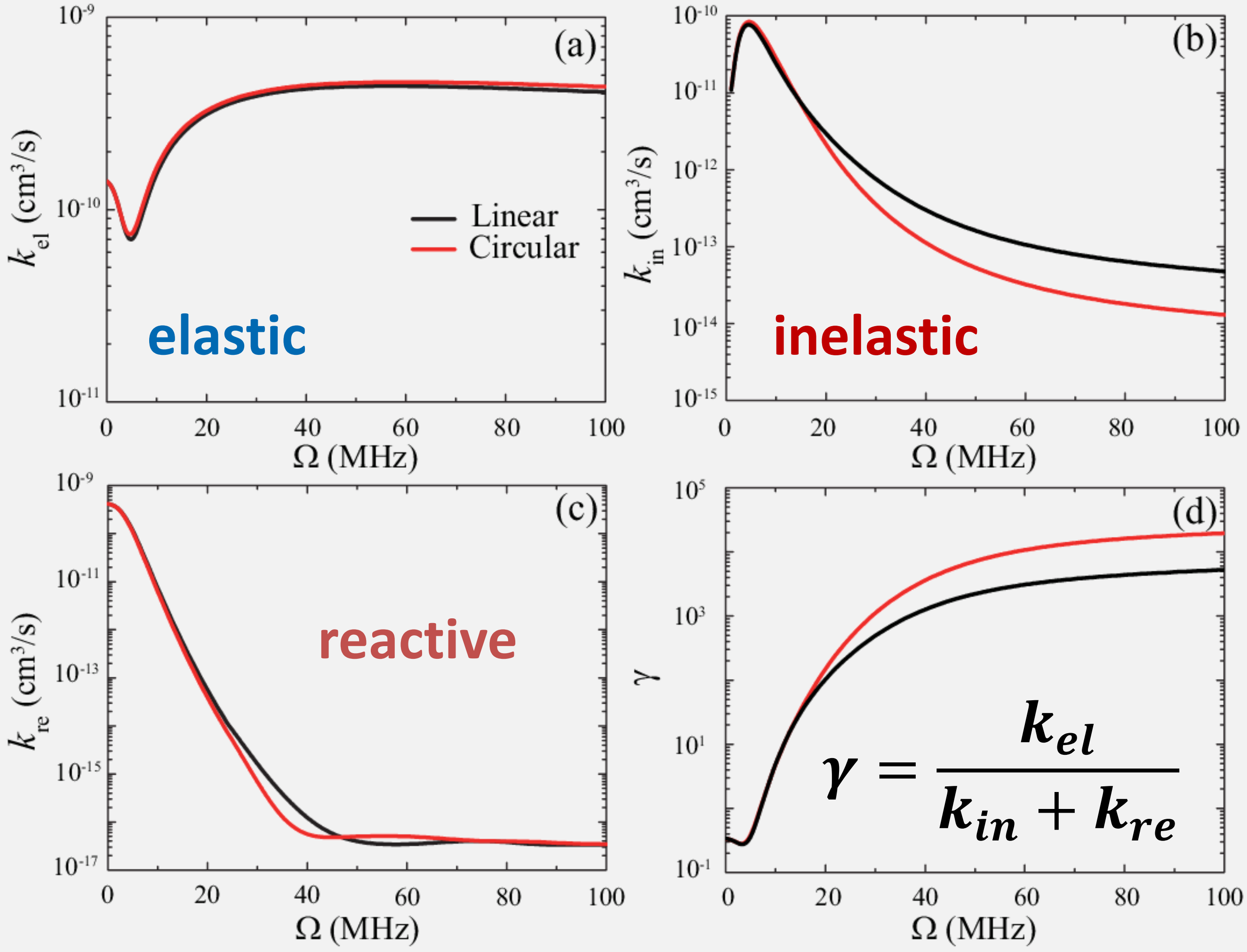

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BUT : 1 molecule photon scattering