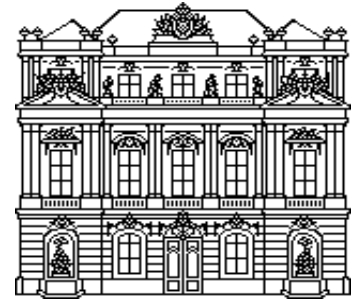# Polar Molecules: QIPC & Condensed Matter Physics

UNIVERSITY OF INNSBRUCK

AUSTRIAN ACADEMY OF SCIENCES

Peter Zoller

Innsbruck:

A. Micheli (PhD)
P. Rabl (PhD)
V. Steixner (PhD)

G. Brennen
H.P. Büchler
G. Pupillo

In collaboration with:

Harvard: J. Doyle, M Lukin
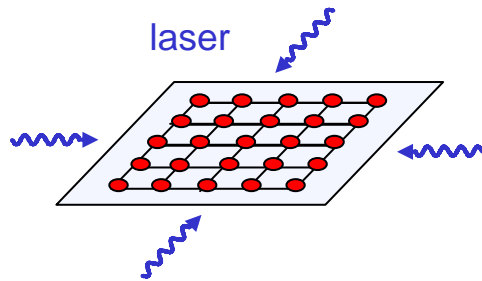Yale: D. DeMille, R. Schoelkopf
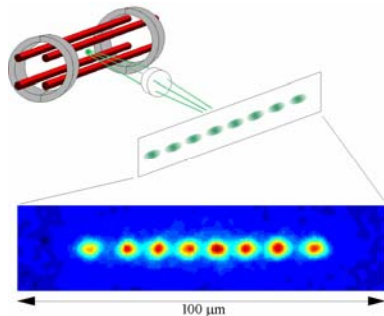
SFB
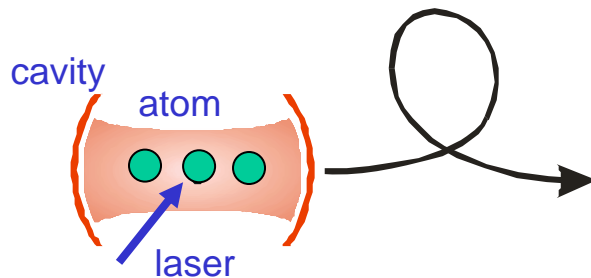*Coherent Control of Quantum Systems*

€U networks

Atoms & Ions

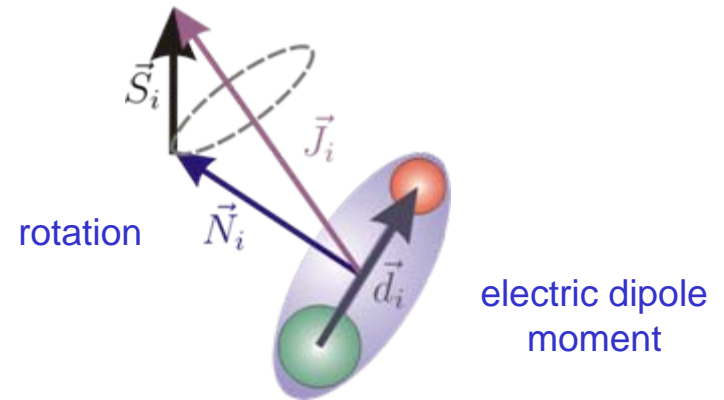- cold atoms in optical lattices

laser

- trapped ions / crystals of …

100 μm

- CQED

cavity
atom
laser

- atomic ensembles

Polar Molecules

$\vec{S}_i$

$\vec{J}_i$

rotation $\vec{N}_i$

$\vec{d}_i$

electric dipole moment

- single molecules / molecular ensembles
- coupling to optical & microwave fields
  - trapping / cooling
  - CQED (strong coupling)
  - spontaneous emission / engineered dissipation
- interfacing solid state / AMO & microwave / optical
  - strong coupling / dissipation
- superfluid – crystalline phases
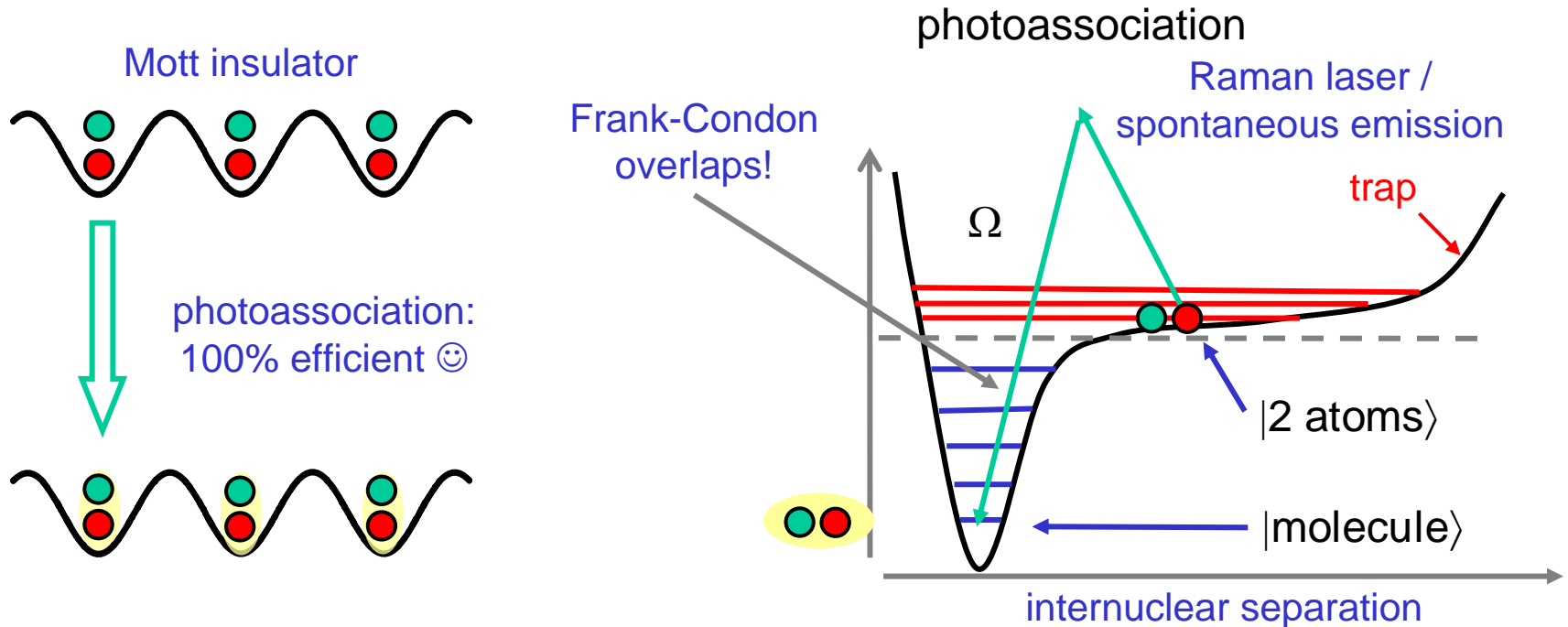 - cond mat / quantum info
  - dephasing

Polar Molecules I (basic features)

- rotational spectra of single molecules
  - … spins, and how to encode qubits
- dipole-dipole interactions

Polar Molecules

- We consider polar molecules *in electronic and vibrational ground state*
    - cooling and trapping techniques being developed
    - preparation via …
        - photoassociation (e.g. from two-species BEC) exp: all cold atom labs
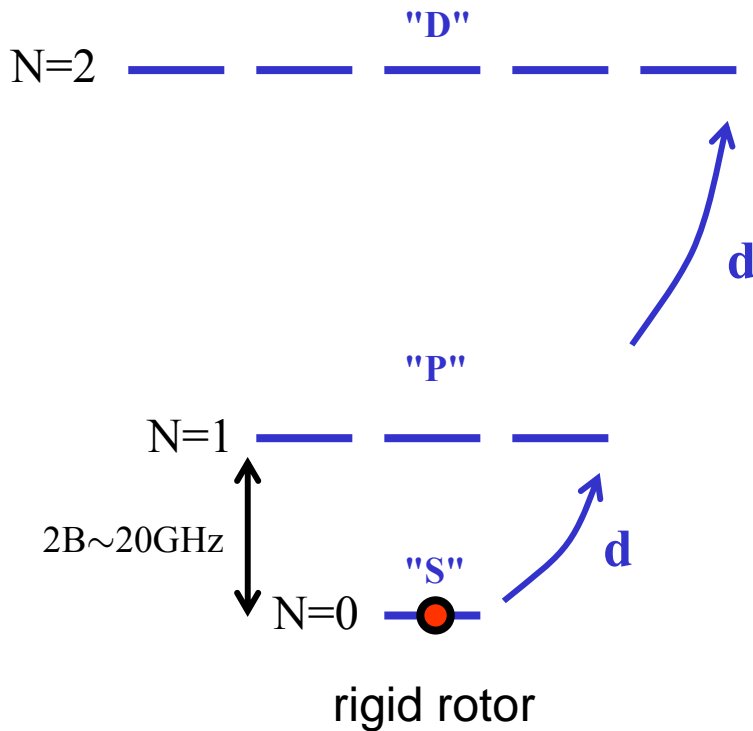   - buffer gas cooling exp: Demille, Doyle, Mejer, Rempe, Ye …

Mott insulator

photoassociation

Frank-Condon overlaps!

Raman laser / spontaneous emission

trap

Ω

photoassociation: 100% efficient ☺

|2 atoms⟩

|molecule⟩

internuclear separation

- Cold polar molecules will exist in the lab in a few years.

1a. Single Polar Molecule: rigid rotor

- single heteronuclear molecule

$$H = B\,\mathbf{N}^2$$

$|\,N, M_N\,\rangle$

$E_N = B\,N(N+1)$

N=2 — — — — — **"D"**

"P"

N=1 — — — —

$2B \sim 20\,\text{GHz}$

"S"

N=0 ●

d

d

rigid rotor

✓ dipole-moment $d \sim 10$ Debye ☺

✓ rotation $B \sim 10$ GHz ... anharmonic ☺

✓ essentially no spontaneous emission 10^{-3} Hz ☺
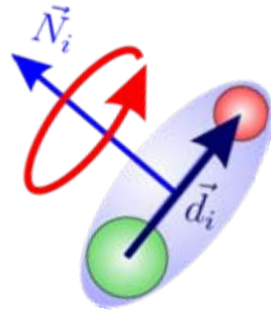black-body scatt.rate ~ 1 Hz ☺

i.e. excited states "useable" ☺

1b. Single Polar molecule: adding spin rotation coupling

- rigid rotor

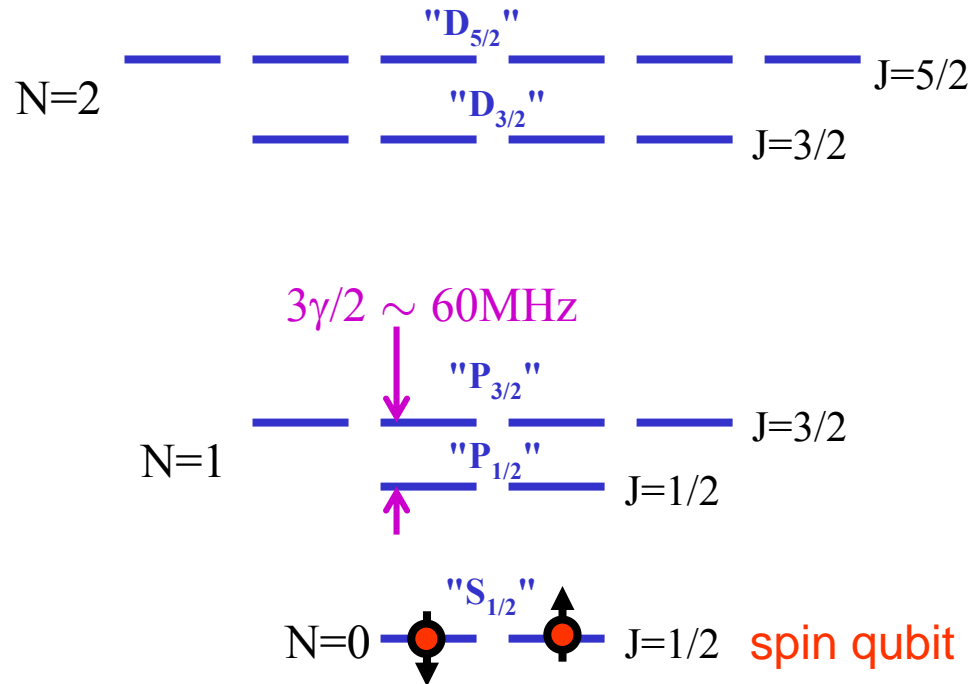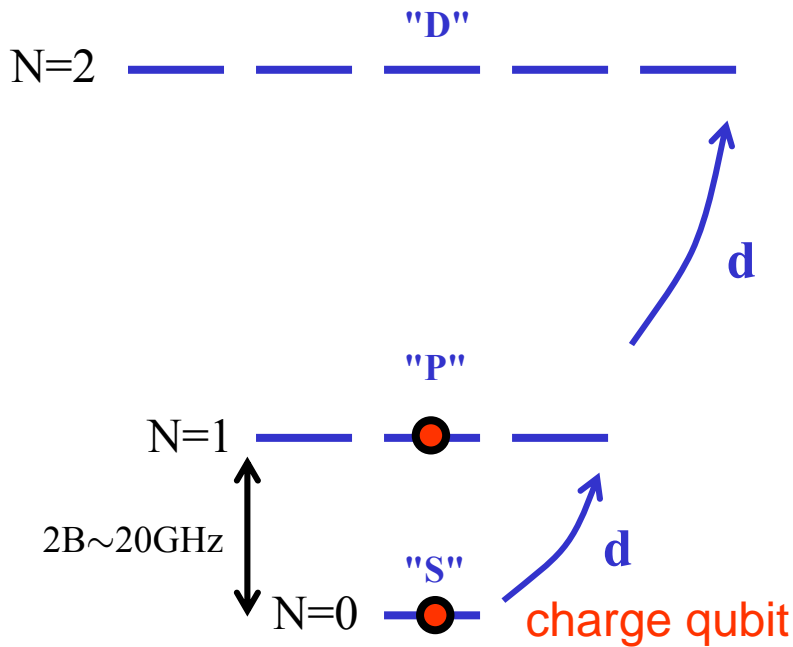$$H = B \, \mathbf{N}^2$$

$$| N, M_N \rangle$$

$$E_N = B \, N(N+1)$$

N=2 —— — — — **"D"**

d

N=1 —— ● —— **"P"**

$2B{\sim}20GHz$

"S"   **d**
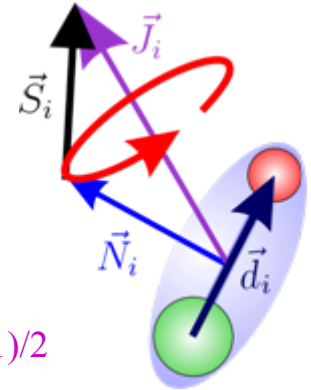
N=0 —— ● —— charge qubit

- adding spin-rotation coupling (S=1/2)

$$H = B \, \mathbf{N}^2 + \gamma \, \mathbf{N}\cdot\mathbf{S}$$

$$| N, J, M_J \rangle \quad (J=|N\pm 1/2|)$$

$$E_{N,J=N\pm\frac{1}{2}} = B \, N(N+1) + \begin{cases} +\gamma \, N/2 \\ -\gamma(N+1)/2 \end{cases}$$

N=2 —— — — — — **"D$_{5/2}$"** —— J=5/2

"D$_{3/2}$" —— J=3/2

$3\gamma/2 \sim 60MHz$

"P$_{3/2}$"

N=1 —— —— J=3/2

"P$_{1/2}$" —— J=1/2

"S$_{1/2}$"

N=0 —● —— ● —— J=1/2   spin qubit

- How to encode qubits?

``looks like an Alkali atom on GHz scale´´
(we adopt this below as our model molecule)

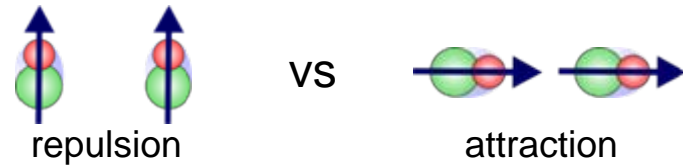# 2. Two Polar Molecules: dipole – dipole interaction

- interaction of two molecules

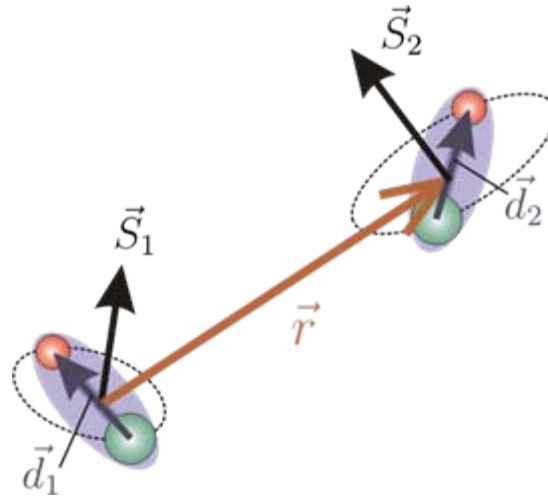$$V_{dd} = \frac{\vec{d}_1 \cdot \vec{d}_2 - 3\left(\vec{d}_1 \cdot \vec{e}_b\right)\left(\vec{e}_b \cdot \vec{d}_2\right)}{r^3}$$

features of dipole-dipole interaction

✓ long range $\sim 1/R^3$

✓ angular dependence

repulsion   vs   attraction
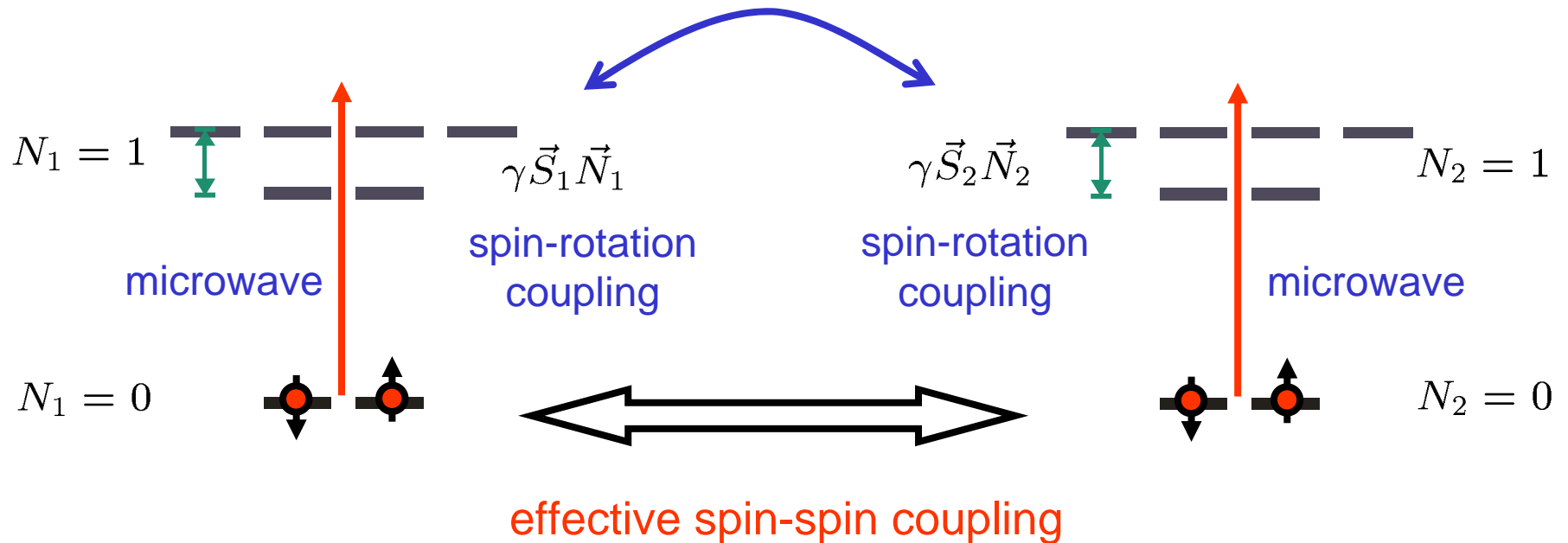
✓ strong! (temperature requirements)

… combining 1+2: engineering spin – spin interactions

dipole-dipole:
anisotropic + long range

$N_1 = 1$

$\gamma \vec{S}_1 \vec{N}_1$

$\gamma \vec{S}_2 \vec{N}_2$

$N_2 = 1$

microwave

spin-rotation
coupling

spin-rotation
coupling

microwave
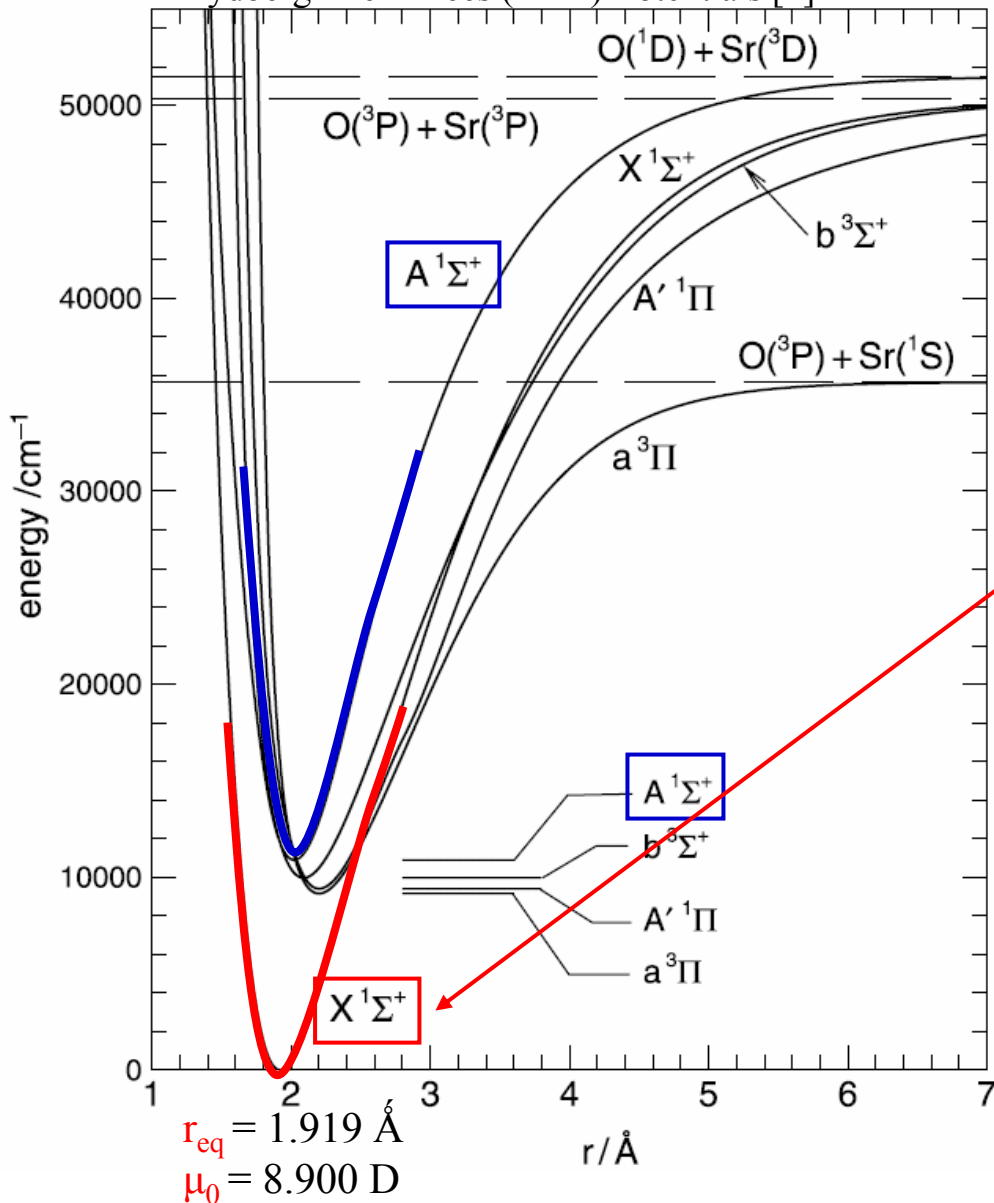
$N_1 = 0$

$N_2 = 0$

effective spin-spin coupling

Polar Molecules II (for the Brave)

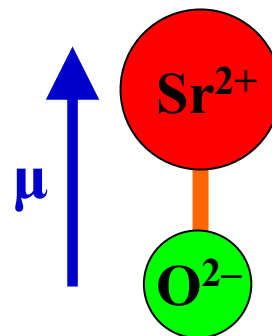- how real molecules look like
  - SrO
  - CaF

Polar Molecules I : Strontium Oxide (SrO)

Rydberg-Klein-Rees (RKR)-Potentials [1]

heteronuclear molecule with strong **persistent** dipole moment in electronic groundstate.

$Sr^{2+}O^{2-}$ ... ionic binding

$_{38}Sr$	[Kr]$5s^2$	
^{88}Sr	$I^p = 0^+$	(83%)
^{86}Sr	$I^p = 0^+$	(10%)
^{87}Sr	$I^p = \frac{3}{2}^+$	(7%)

$_8O$	$1s2s^2p^4$	
^{16}O	$I^p = 0^+$	(99.76%)
^{18}O	$I^p = 0^+$	(0.20%)

$X\ ^1\Sigma^+$ ... electronic groundstate:

S=0 ... closed shell $(..9\sigma^2\ 10\sigma^2\ 4\pi^4)$
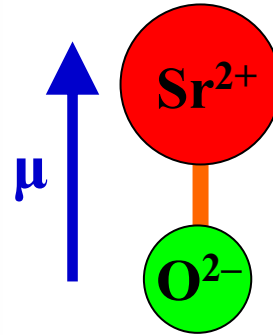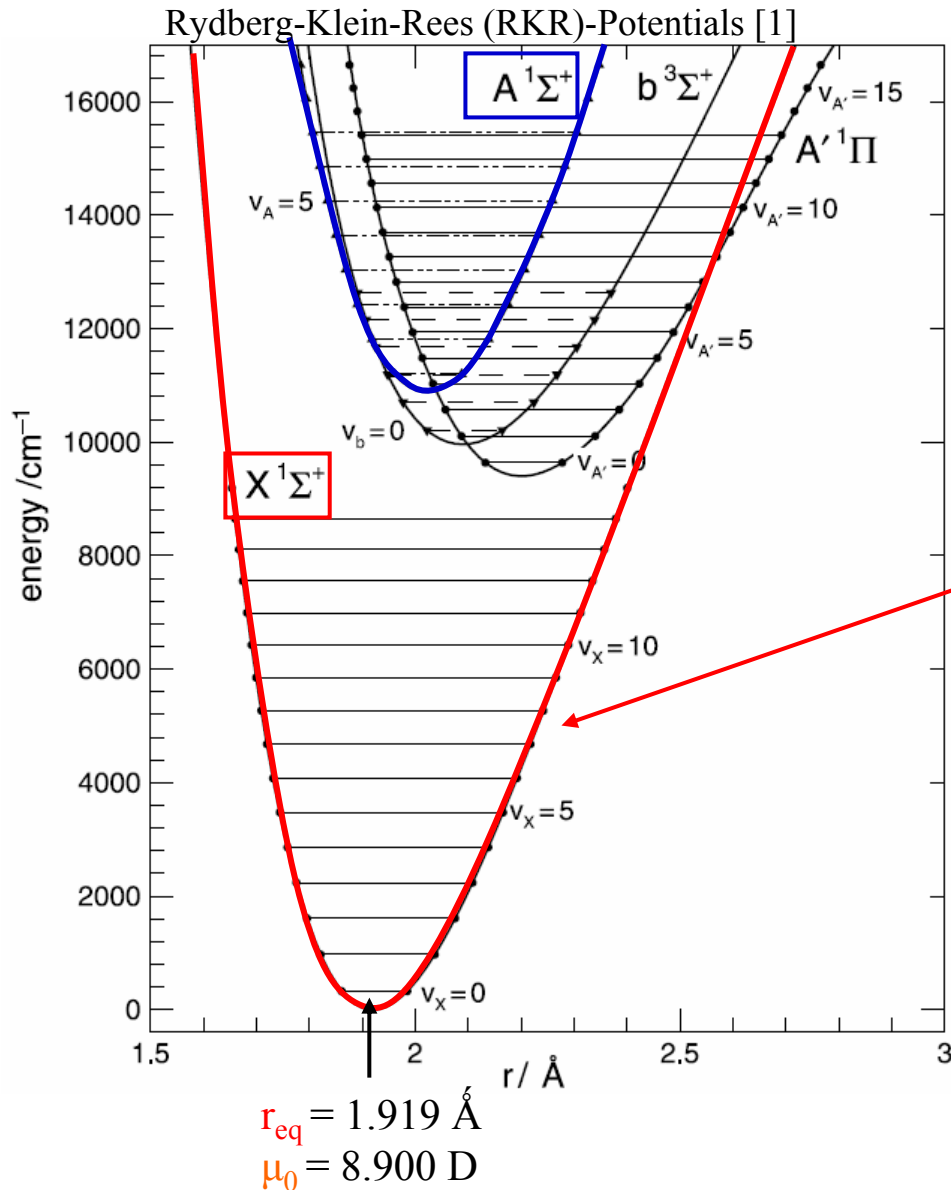
r_{eq} = 1.919 Å ... equilibirum distance
μ_0 = 8.900 D ... dipole-moment

ω_{eq} = 19.586 THz ... vibrational const.
B_{eq} = 10.145 GHz ... rotational $(h/8\pi^2 M_{red} r_{eq}^2)$

I=0 ... no nuclear momenta for ^{88}SrO, ^{86}SrO
[^{87}SrO has I=3/2 and thereby nuclear quadrupole momenta ... $eQq_n \approx -42MHz$]

r_{eq} = 1.919 Å
μ_0 = 8.900 D

Polar Molecules I : Strontium Oxide (SrO)

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$_8$O $1s2s^2p^4$

| ^{16}O | $I^p = 0^+$ | (99.76%) |
| ^{18}O | $I^p = 0^+$ | (0.20%) |

X $^1\Sigma^+$ ... el. groundstate:

S=0 ... closed shell ($..9\sigma^2 \, 10\sigma^2 \, 4\pi^4$)

r_{eq} = 1.919 Å ... equilibirum distance
μ_0 = 8.900 D ... dipole-moment

ω_{eq} = 19.586 THz ... vibrational const.
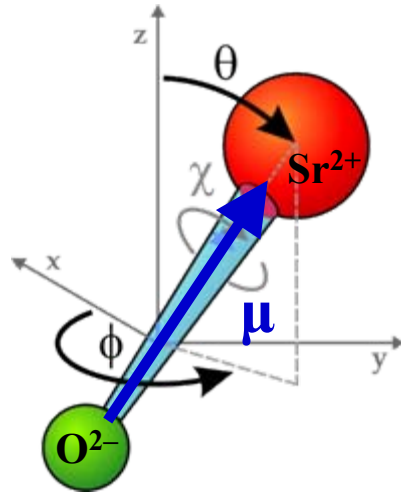B_{eq} = 10.145 GHz ... rotational *(h/8π²M$_{red}$r$_{eq}$²)*

I=0 ... no nuclear momenta for ^{88}SrO, ^{86}SrO
   *[^{87}SrO has I=3/2 and thereby nuclear
   quadrupol momenta ... $eQq_n \approx -42MHz$]*

r_{eq} = 1.919 Å
μ_0 = 8.900 D

SrO as an "ideal" rigid Rotor

^{88}SrO in vibrational gs X $^1\Sigma^+$ (υ=0):

[υ=1 at ω_{eq} = 19.586 THz, no fine/hfs: S=I=0]

• Rigid Rotor
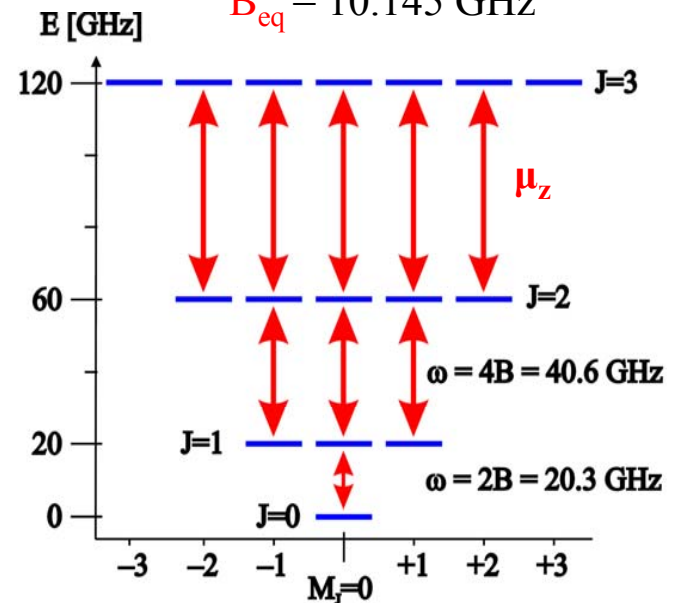
$$\hat{H} = \frac{\hat{\mathbf{J}}^2}{2\mathcal{I}}$$

$$E_{J,M_J} = BJ(J+1)$$

B_{eq} = 10.145 GHz

μ_z

$\omega = 4B = 40.6$ GHz

$\omega = 2B = 20.3$ GHz

• strong anharmonicity ~ J^2
• unpolarized
• electric dipole transitions
   $\Delta J = 1$ and $\Delta M_J = 0,1$
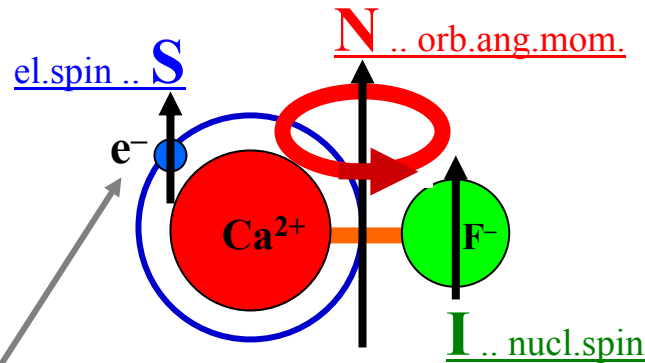
CaF – rotational, fine and hyperfine structure

_X_ $^2\Sigma_{1/2}$ ... el. groundstate:

S=½ ... from open (Ca-)shell
I=½ ... nuclear momentum (of F)

r_{eq} = 1.951 Å ... eq. distance
μ_0 = 3.077 D ... dipole-moment

el.spin .. **S**

N .. orb.ang.mom.

e⁻

Ca²⁺

F⁻

I .. nucl.spin

dipole moment

optical excitation
Alkali-like

talks to optical radation,
like an alkali atom

talks to microwave radation
(alkali atom: hyperfine / magnetic)
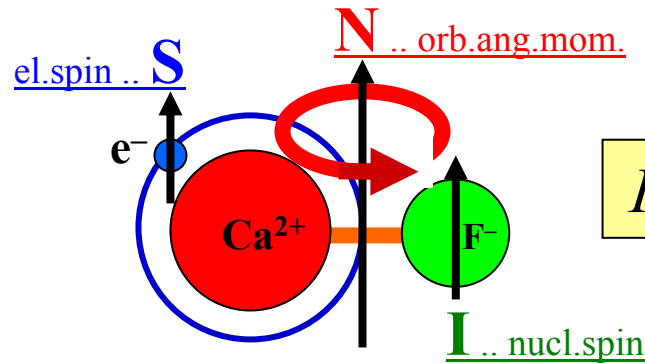
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<u>N</u> .. orb.ang.mom.

el.spin .. **S**

e⁻

Ca²⁺

F⁻

<u>I</u> .. nucl.spin

$$H_0 = H_{rot} + H_{spin\text{-}rot} + H_{hfs}$$

$$\boxed{H_{\textbf{rot}} = B\,\mathbf{N}^2}$$

B = 10.3 GHz

N=2

20.6 GHz

N=1

10.3 GHz

N=0

(rotor)

CaF – rotational, fine and hyperfine structure

optical excitation
Alkali-like

<u>*X* $^2\Sigma_{1/2}$ ... el. groundstate:</u>

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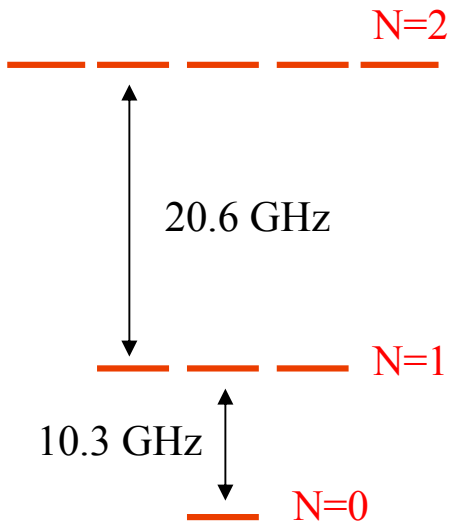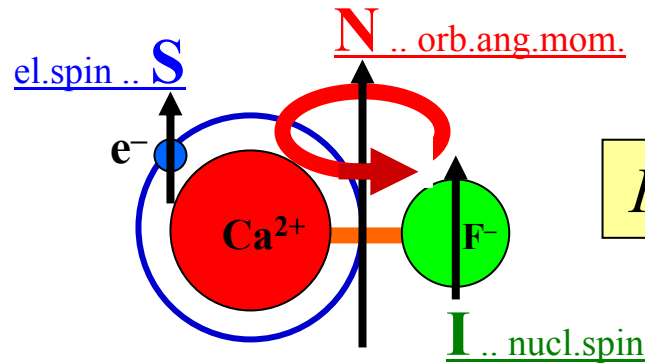I=½ ... nuclear momentum (of F)

r_{eq} = 1.951 Å ... eq. distance
μ_0 = 3.077 D ... dipole-moment

<u>**N** .. orb.ang.mom.</u>

el.spin .. **S**

e⁻

Ca²⁺

F⁻

I .. nucl.spin

$$H_0 = H_{rot} + H_{spin\text{-}rot} + H_{hfs}$$

$$H_{rot} = B\ \mathbf{N}^2$$

B = 10.3 GHz

$$H_{spin\text{-}rot} = \gamma\ \mathbf{N}\cdot\mathbf{S}$$

γ = 39.66 MHz

N=2 — — — — —   J=5/2 – – – – – – –   (N=2)

J=3/2 – – – – –

20.6 GHz

J = 3/2 – – – – –   (N=1)

J = ½ – –

N=1 — — — —

10.3 GHz

N=0 ————————   J=½   (N=0)

(rotor)

(**J = N + S**)

CaF – rotational, fine and hyperfine structure

$\underline{X\ {}^2\Sigma_{1/2}}$ ... el. groundstate:

S=½ ... from open (Ca-)shell
I=½ ... nuclear momentum (of F)

r_{eq} = 1.951 Å ... eq. distance
μ_0 = 3.077 D ... dipole-moment

<u>**N**</u> .. orb.ang.mom.
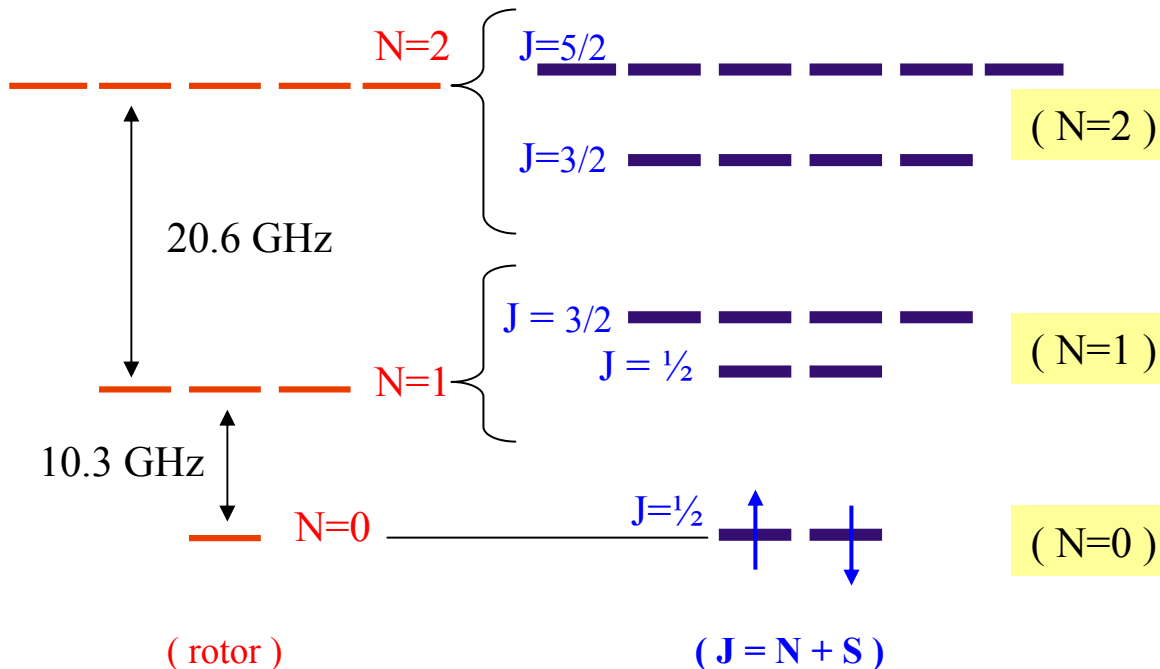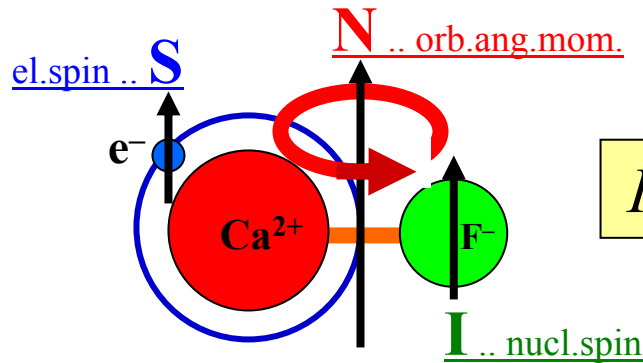
el.spin .. **S**

e⁻

Ca²⁺

F⁻

<u>**I**</u> .. nucl.spin

$$H_0 = H_{rot} + H_{spin\text{-}rot} + H_{hfs}$$

... S_ζ component along figure axis

$$H_{\mathbf{rot}} = B\ \mathbf{N}^2$$

B = 10.3 GHz

$$H_{\mathbf{spin\text{-}rot}} = \gamma\ \mathbf{N}{\cdot}\mathbf{S}$$

γ = 39.66 MHz

$$H_{\mathbf{hfs}} = (b\ \mathbf{S}{\cdot}\mathbf{I} + c\ S_\zeta I_\zeta) + C\ \mathbf{N}{\cdot}\mathbf{I}$$

b=109.18 MHz, c=40.12 MHz, C=30 KHz

N=2

J=5/2

J=3/2

(N=2)

F=3
F=2
F=1

F=2

20.6 GHz

J = 3/2

J = ½

(N=1)

F=2
F=1
F=0

F=1

N=1

10.3 GHz

N=0

J=½

(N=0)

F=1

F=0

125 MHz

(rotor)

(**J = N + S**)

(**F = N + S + I**)

Calcium Monofluoride (CaF): in external fields III

el.spin .. **S**

N .. orb.ang.mom.

e⁻

Ca²⁺ **F⁻**

I .. nucl.spin

Summary for Quantum
Optics Theorists

$|r\rangle$

$|q\rangle$

$|g\rangle$

two & three level systems

spontaneous emission from
excited states negligible

148 MHz

F=2
F=1
F=0

} N=1

F=1

20.6 GHz

microwave

F=1

123 MHz

} N=0

F=0

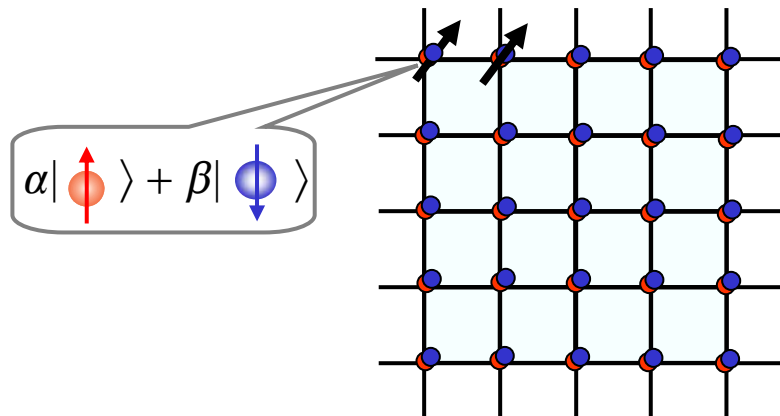−2   −1   0   +1   +2

M_F

clock transition

Sneak Preview:

What can we do with Polar Molecules?

- a few examples & ideas

1. Polar Molecules in Optical Lattices: "Spin Lattice Toolbox"

- Mott insulator state of molecules in optical lattices

$$\alpha| \uparrow \rangle + \beta| \downarrow \rangle$$

engineer spin models:

$$H = g \sum_{i \neq j} \sigma_\alpha^{(i)} A^{\alpha\beta}(\vec{x}_i, \vec{x}_j) \sigma_\beta^{(j)}$$

✓ range

✓ spin / space anisotropy

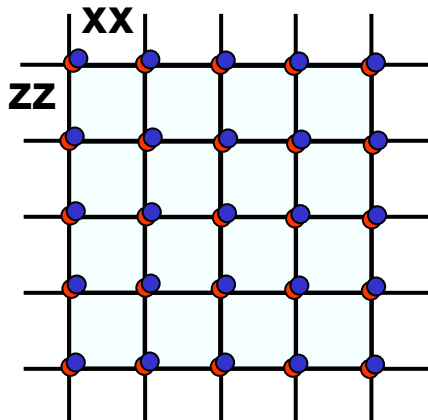dipoles: large offsite couplings!

- [… and Hubbard models with large offsite interactions]
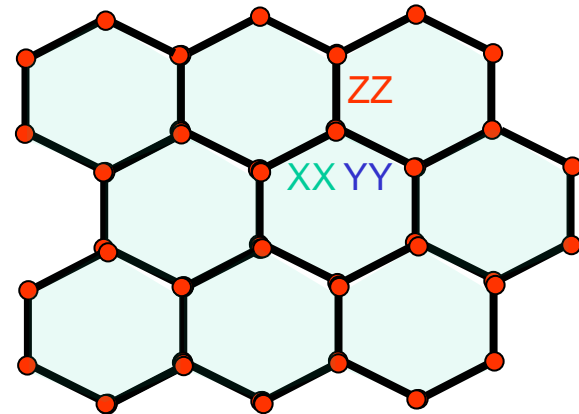
Examples …

- **Protected Quantum Memory**

  B.Douçot, M.V.Feigel'man, L.B.Ioffe,
  A.S.Ioselevich, Phys. Rev. B **71**, 024505 (2005).

- **Kitaev 2005**

  E.Dennis, A.Y.Kitaev, J.Preskill, JMP **43,** 4452;
  A.Y.Kitaev, cond-mat/0506438 (2005);

$$H_{\rm spin}^{(I)} = \sum_{i=1}^{\ell-1} \sum_{j=1}^{\ell-1} J(\sigma_{i,j}^z \sigma_{i,j+1}^z + \cos\zeta\, \sigma_{i,j}^x \sigma_{i+1,j}^x)$$
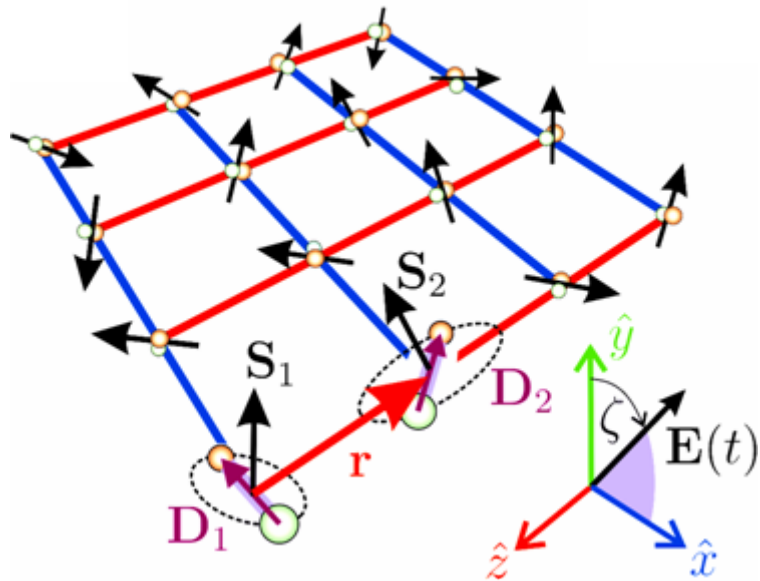
$$H_{\rm spin}^{(II)} = J_\perp \sum_{x-{\rm links}} \sigma_j^x \sigma_k^x + J_\perp \sum_{y-{\rm links}} \sigma_j^y \sigma_k^y + J_z \sum_{z-{\rm links}} \sigma_j^z$$

A. Micheli, G.K. Brennen, and P. Zoller, Nature Physics May 2006

Examples …

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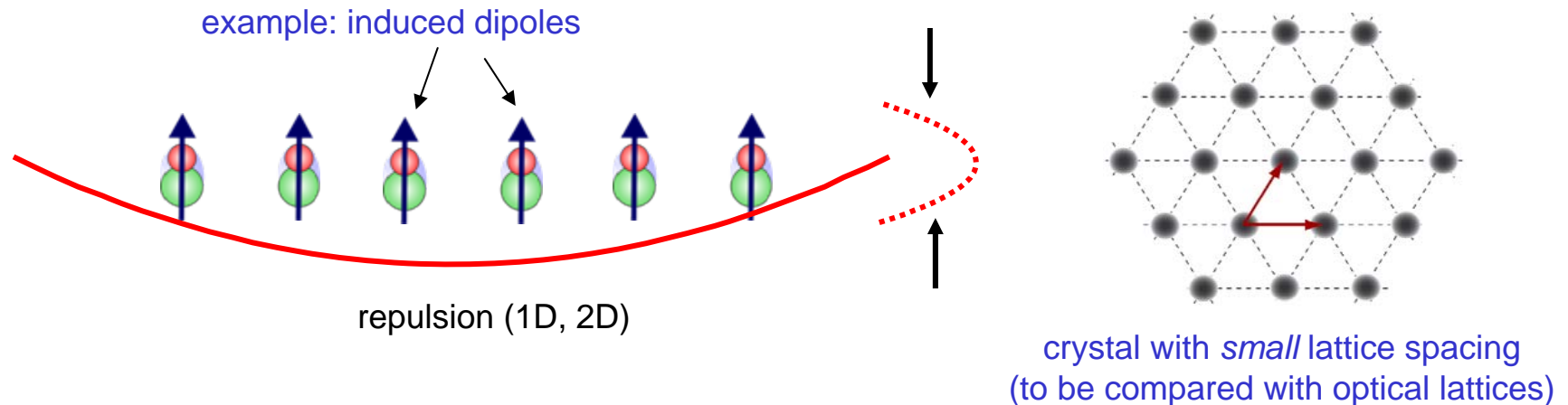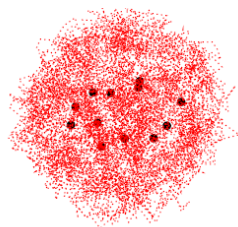

$$H_{\text{spin}}^{(\text{II})} = J_\perp \sum_{x-\text{links}} \sigma_j^x \sigma_k^x + J_\perp \sum_{y-\text{links}} \sigma_j^y \sigma_k^y + J_z \sum_{z-\text{links}} \sigma_j^z$$

A. Micheli, G.K. Brennen, and P. Zoller, Nature Physics May 2006

2. Self-assembled Lattices / Crystals with Atoms / Molecules

- engineer *repulsive* interactions between atoms / molecules so that …

example: induced dipoles

repulsion (1D, 2D)

crystal with *small* lattice spacing
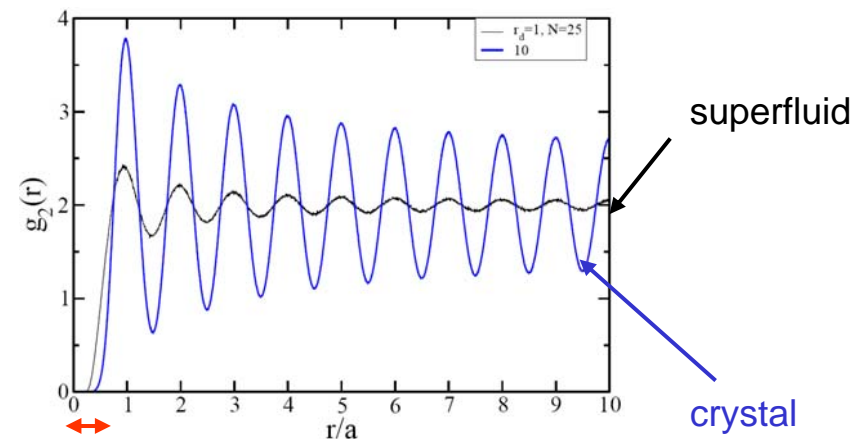(to be compared with optical lattices)

- Liquid – Crystal phase transitions:

superfluid

crystal: shell structure
(compare: ions)

superfluid

crystal

G. Pupillo, A.Micheli, V. Steixner, H.P. Büchler, and P. Zoller (Innsbruck)
N.V. Prokof'ev (Umass, Amherst) & Lukin group (Harvard); work in progress

3. Solid State / AMO Hybrid Quantum Processor

- solid state processor

- AMO quantum memory
  here: molecular ensembles

- interface to ‚flying' optical qubits

optical cavity

molecular ensemble

optical (flying) qubit

laser

superconducting *microwave* transmission line cavity (photon bus)

polar molecular ensemble (quantum memory)

Cooper Pair Box (superconducting qubit)

polar molecular ensemble (quantum memory)

circuit CQED: R. Schoelkopf, S. Girvin (Yale)

P. Rabl, D. DeMille, J. Doyle, M. Lukin, R. Schoelkopf and P. Zoller, preprint

- or: couple single molecules   Yale – Harvard – Innsbruck collaboration, draft
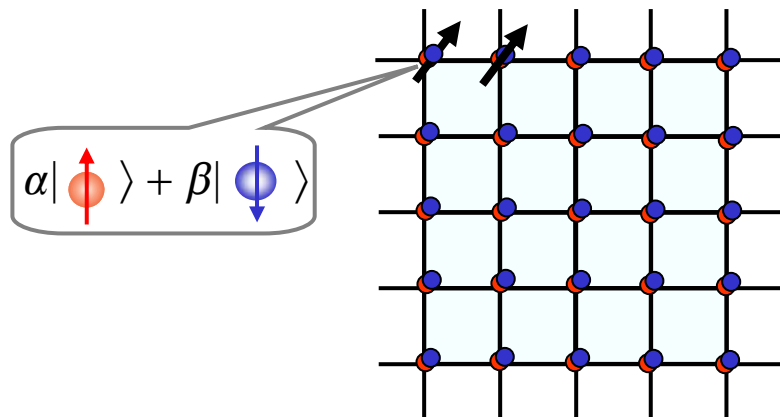
Spin Models with Optical Lattices

- quantum info relevance:
  - polar molecule realization of models for protected quantum memory (Ioffe, Feigelman et al.)
  - Kitaev model on honey comb lattice

A. Micheli, G.K. Brennen, and P. Zoller, Nature Physics May 2006

Polar Molecules in Optical Lattices: "Spin Lattice Toolbox"

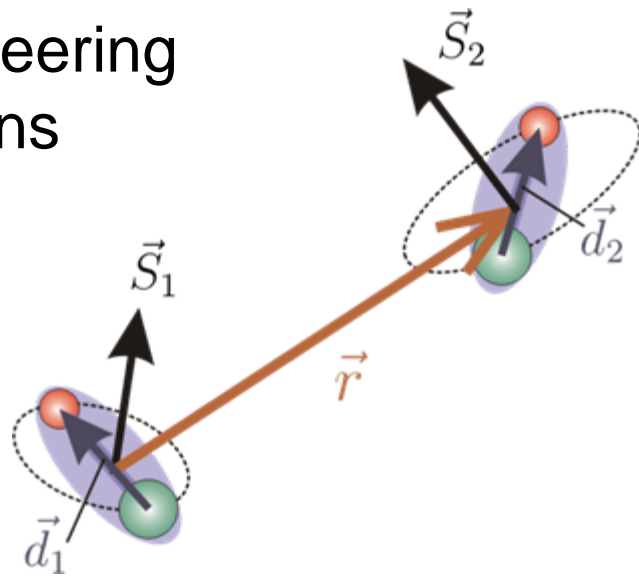- Mott insulator state of molecules in optical lattices

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✓ range

✓ spin / space anisotropy

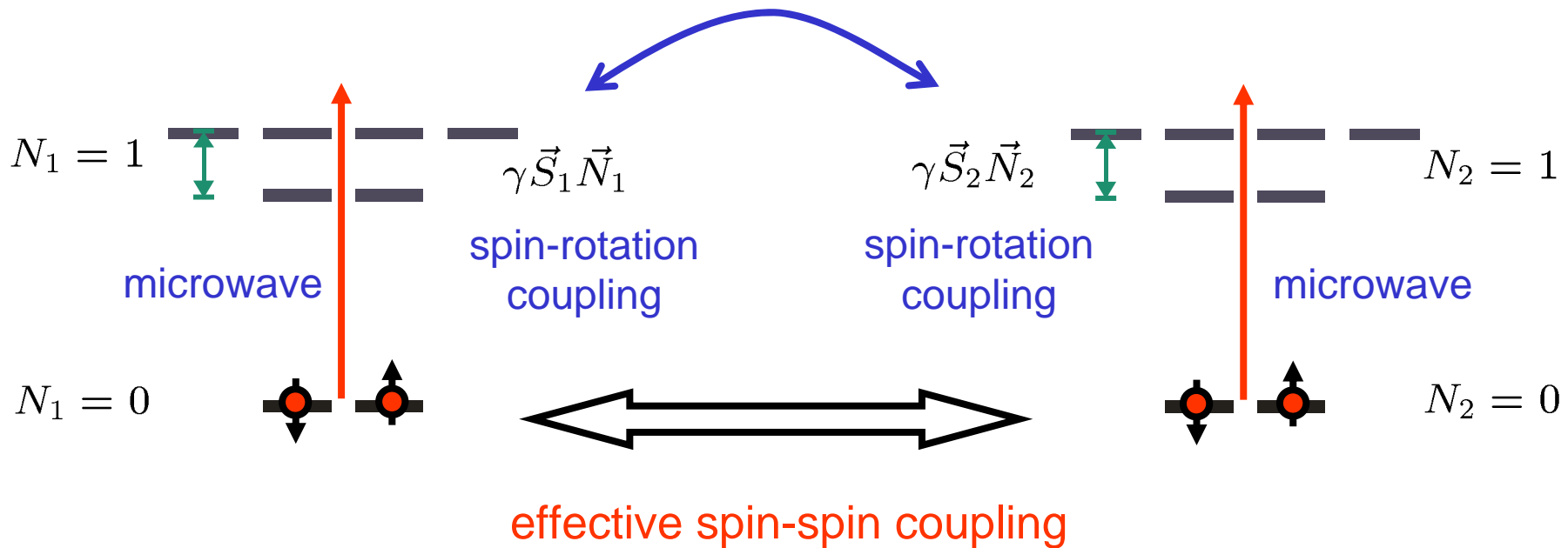dipoles: large offsite couplings!

- [… and Hubbard models with large offsite interactions]
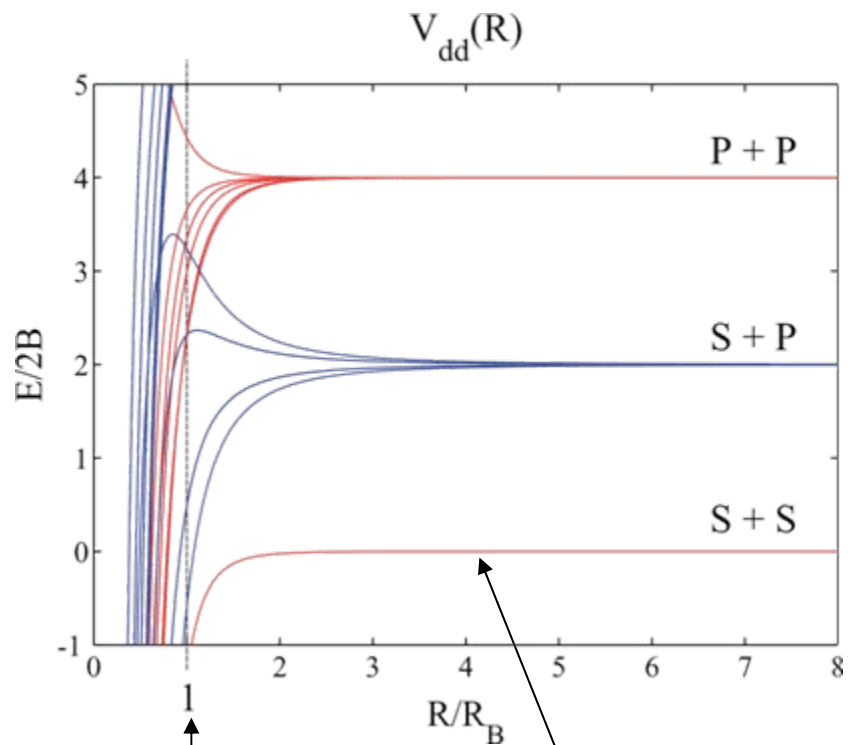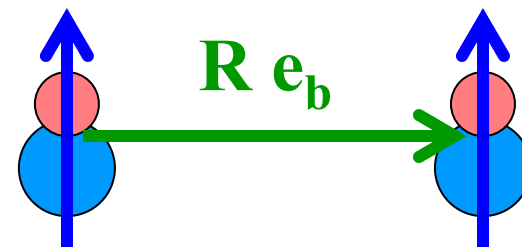
Basic idea of engineering spin-spin interactions

$\vec{S}_2$

$\vec{S}_1$

$\vec{d}_2$

$\vec{r}$

$\vec{d}_1$

dipole-dipole:
anisotropic + long range

$N_1 = 1$

$\gamma \vec{S}_1 \vec{N}_1$

$\gamma \vec{S}_2 \vec{N}_2$

$N_2 = 1$

microwave

spin-rotation coupling

spin-rotation coupling

microwave

$N_1 = 0$

$N_2 = 0$

effective spin-spin coupling

Adiabatic potentials for two (unpolarized) polar molecules
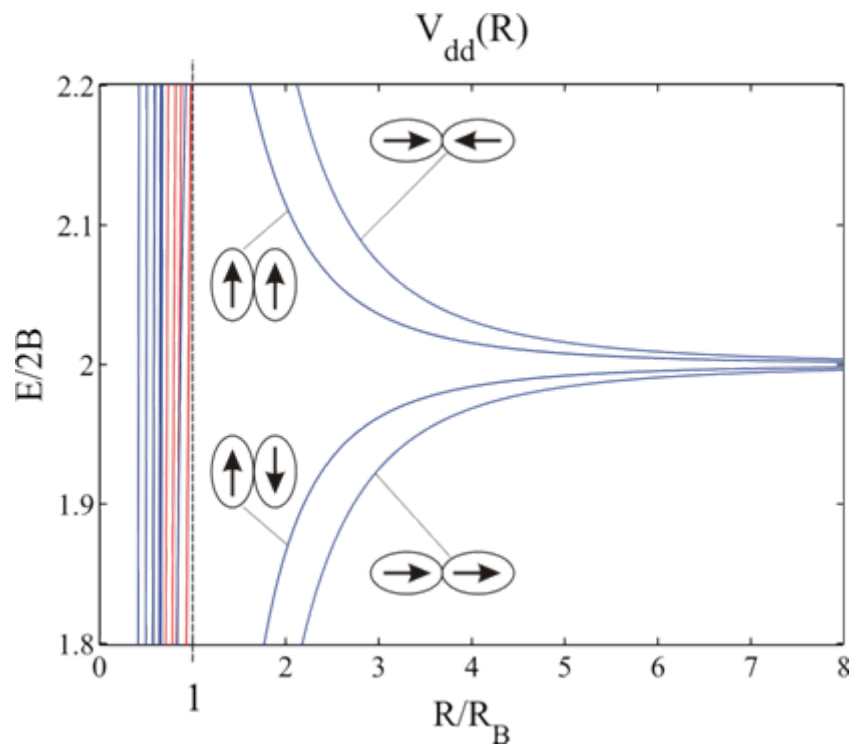

$$R\ e_b$$
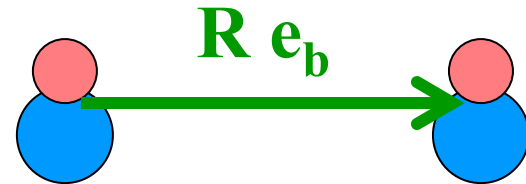
- **Rotor**

$$R_B = \left(\frac{\mu^2}{3B}\right)^{1/3}$$

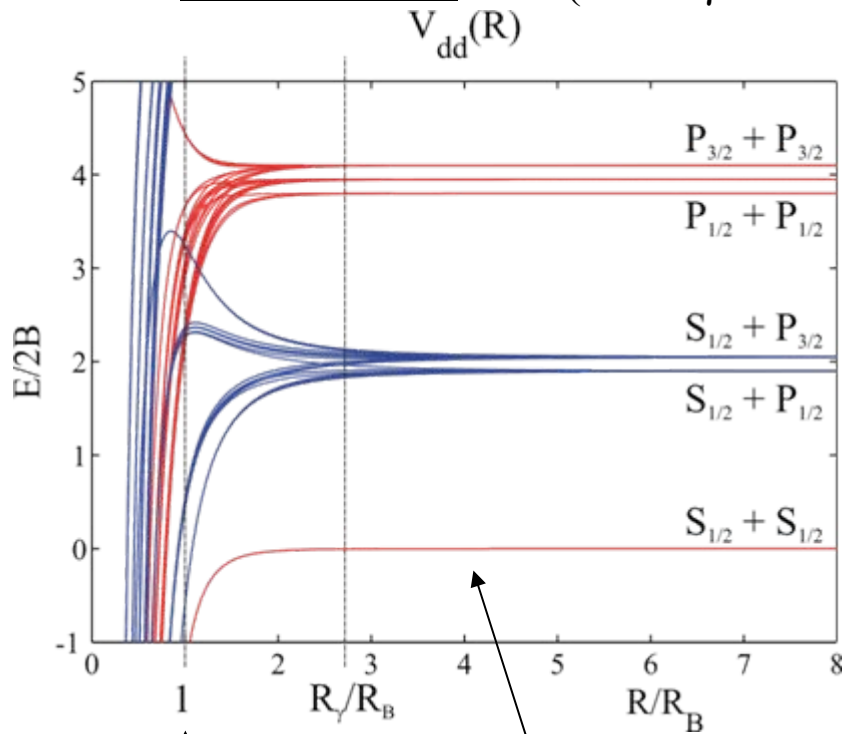$$V_{\text{eff}}(R) = -\frac{C_6}{R^6}$$

$$C_6 = \frac{\mu^4}{6B}$$

~ 30-60 nm

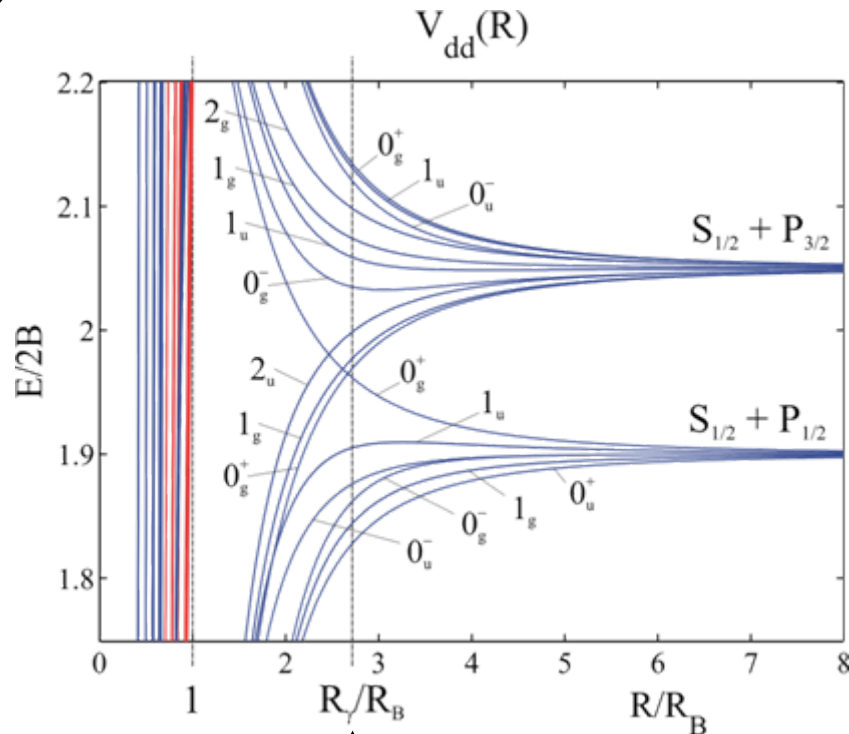# Adiabatic potentials for two (unpolarized) polar molecules

$R\ e_b$

- **Spin Rotation** (here: $\gamma/B = 1/10$)

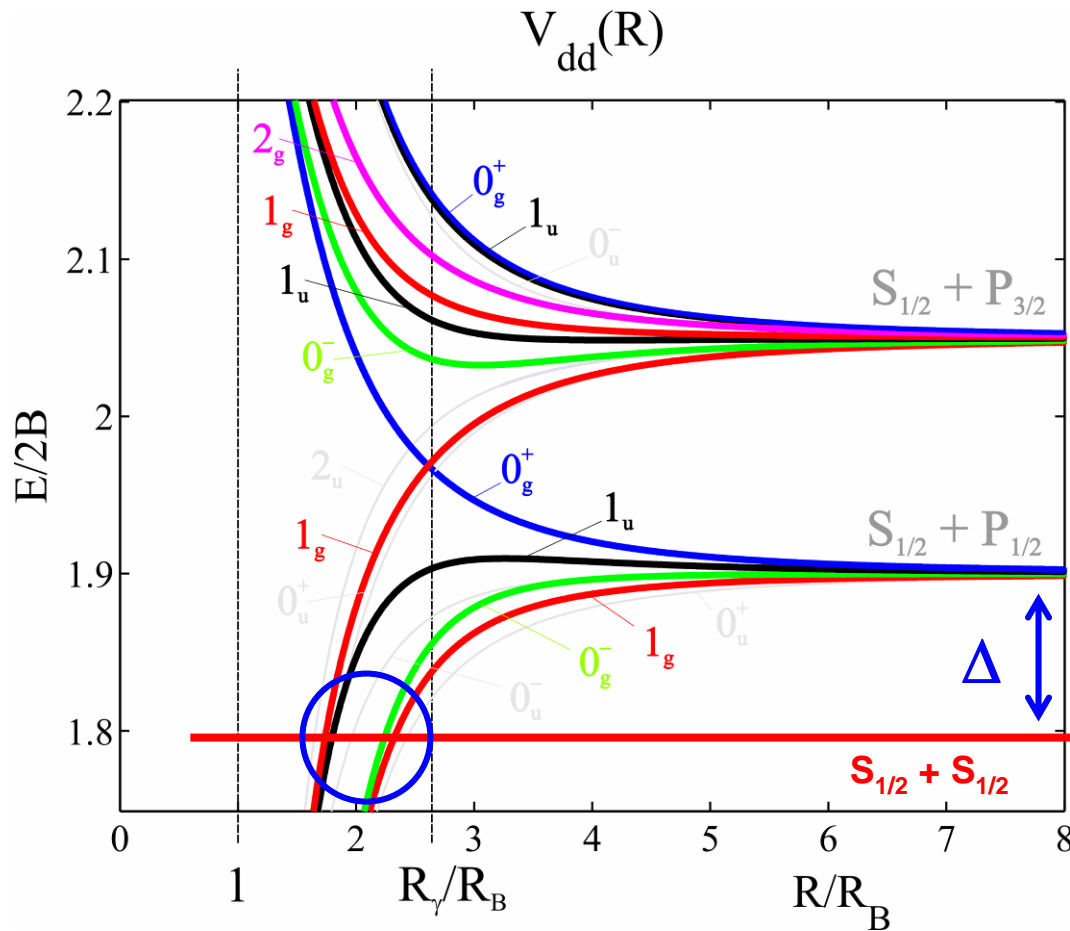$$R_B = \left(\frac{\mu^2}{3B}\right)^{1/3}$$

$$C_6 = \frac{\mu^4}{6B}$$

(here: $\gamma/B = 1/10$)

$$R_\gamma = \left(\frac{2\mu^2}{3\gamma}\right)^{1/3}$$ typical: ~300 nm

$$V_{\text{eff}}(R) = -\frac{C_6}{R^6}\left[1 + \left(\frac{\gamma}{2B}\right)^2\left(S_1 \cdot S_2 - S_1^b S_2^b\right) + O\left(\frac{\gamma}{2B}\right)^4\right]$$ ~small

Adiabatic potentials for two (unpolarized) polar molecules

- **Spin Rotation** (here: $\gamma/B = 1/10$)

Induced effective interactions:

$0_g^+ : + \mathbf{S}_1 \cdot \mathbf{S}_2 - 2\, S_1^c\, S_2^c$
$0_g^- : + \mathbf{S}_1 \cdot \mathbf{S}_2 - 2\, S_1^p\, S_2^p$
$1_g : + \mathbf{S}_1 \cdot \mathbf{S}_2 - 2\, S_1^b\, S_2^b$
$1_u : - \mathbf{S}_1 \cdot \mathbf{S}_2$
$2_g : + S_1^b\, S_2^b$
$0_u : 0$
$2_u : 0$

for $e_{body} = e_x$ and $e_{pol} = e_z$

$0_g^+ : +XX - YY + ZZ$
$0_g^- : +XX + YY - ZZ$
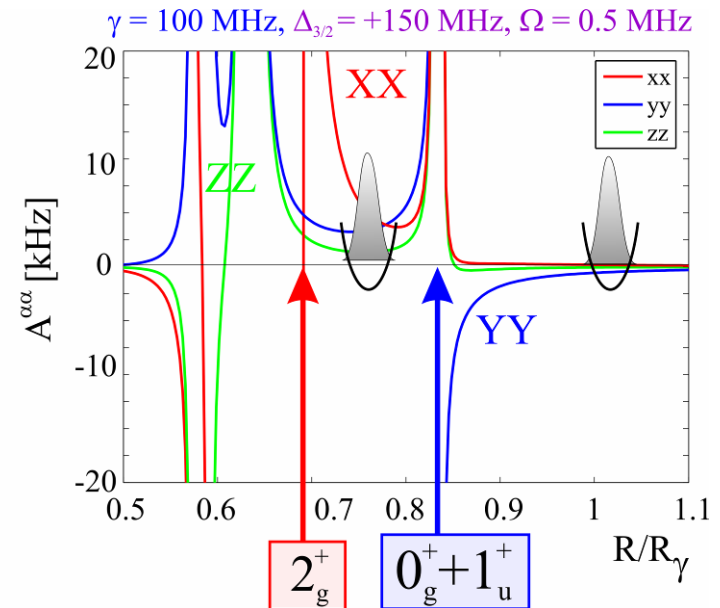$1_g : -XX + YY + ZZ$
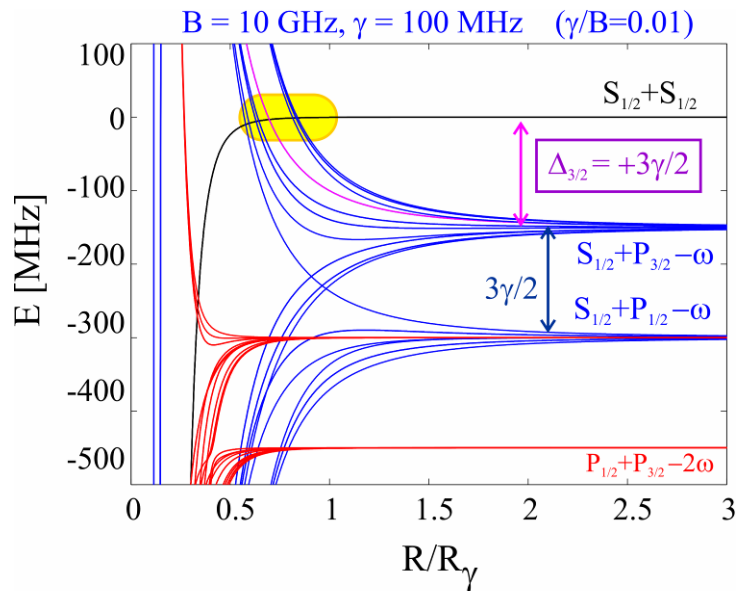$1_u : -XX - YY - ZZ$
$2_g : +XX$

Feature 1. By tuning close to a resonance we can select a *specific spin texture*

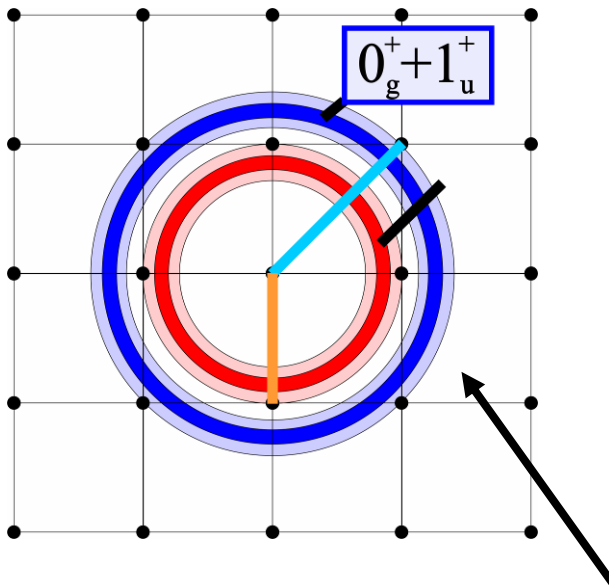# Example: "The Douçot-Feigel'man-Ioffe et al. Model"

$$H = J_x \sum_{i,j} \sigma_x^{(i,j)} \sigma_x^{(i,j+1)} + J_z \sum_{i,j} \sigma_z^{(i,j)} \sigma_z^{(i+1,j)}$$

- Consider blue detuning from S+P$_{3/2}$ transitions:

Example: "The Douçot-Feigel'man-Ioffe et al. Model"

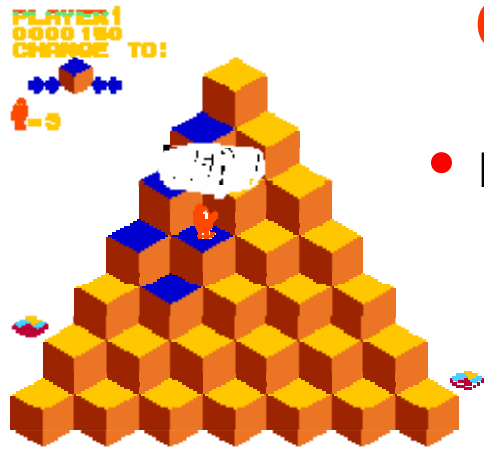- Model is simple in terms of long-range resonances …

$$0_g^+ + 1_u^+$$

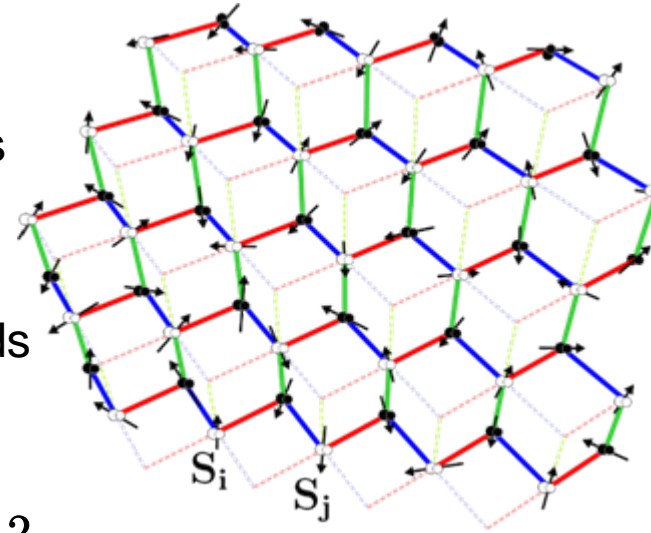Rem.: for a multifrequency field we can *add* the corresponding spin textures.

Feature 2. We can choose the *range* of the interaction for a given spin texture

Feature 3. for a *multifrequency* field spin textures are *additive*: toolbox
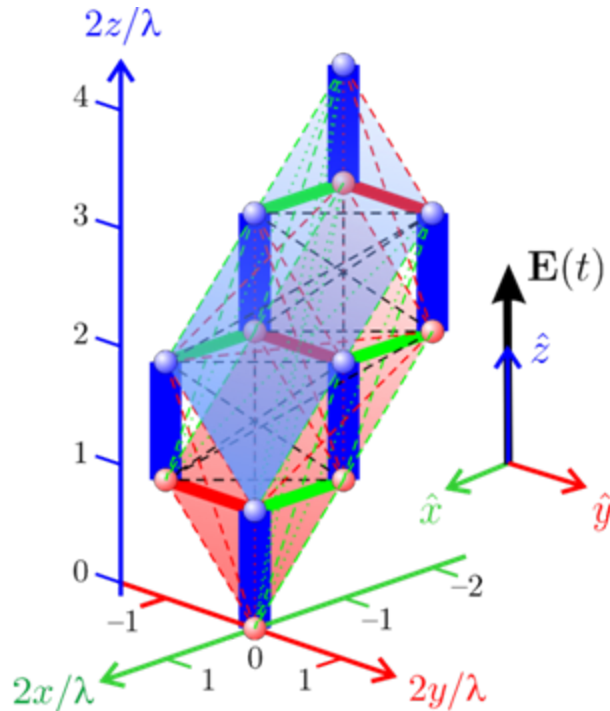
Construction in an optical lattice

- Implementation in *Q*bert lattice*:
  - Two staggered triangular lattices
  - Nearest neighbors give honeycombs
  - their edges form orthogonal triads

- Realization with 3 fields: (several possible choices) shown when all 3 being z polarized, resp. near 0_g, 1_g, 2_g

Spin pattern		Residual long range coupling strengths $|J_{lr}|$	
———	$\sigma^z\sigma^z$		
———	$\sigma^x\sigma^x$		
———	$\sigma^y\sigma^y$	– – –	$< 10^{-2}\,|J_z|$
———	Other	··········	$< 10^{-3}\,|J_z|$
$|J_\perp| = 0.4|J_z|$			

Operator fidelity (on a 4 spin configuration)

$$\sup\left[||H_{\mathrm{spin}} - H_{\mathrm{spin}}^{(\mathrm{II})}|\psi\rangle||_2;\ \langle\psi|\psi\rangle = 1\right] = 10^{-4}\,|J_z|$$
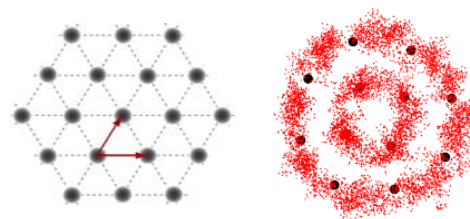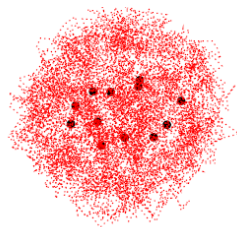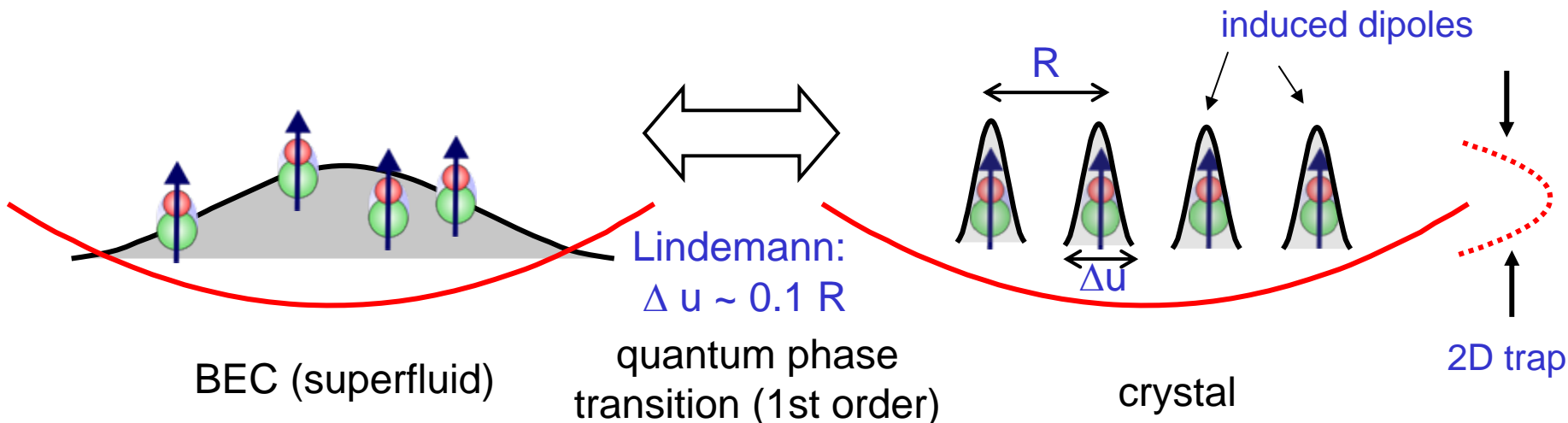
Crystals / self-assembled Lattices with Polar Molecules (and Atoms)

G. Pupillo, A.Micheli, V. Steixner, H.P. Büchler, and P. Zoller (Innbruck)
N.V. Prokof'ev (Umass, Amherst) & Lukin group (Harvard)

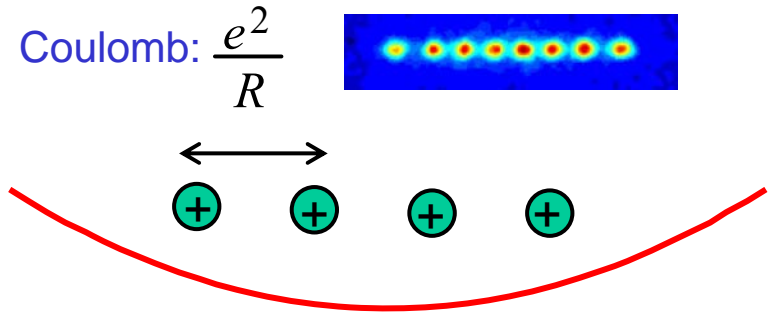# Crystalline Phases with Molecules (and Atoms)

- requirement: engineering *long range repulsive* interactions, e.g. $V \sim 1/r^\alpha$
  - e.g. (induced & aligned) electric dipole moments of molecules (or atoms) in 1D or 2D: $V \sim \mu^2 /r^3;$ or repulsive $+C_6/r^6$
- superfluid – crystal quantum phase transition (at T=0)

induced dipoles

R

Lindemann:
$\Delta u \sim 0.1\ R$

Δu

2D trap

BEC (superfluid)

quantum phase
transition (1st order)

crystal

self assembled lattice / shell structure

- how build, prepare, measure? properties? why? … from an AMO point of view
- Ref.:many papers on dipolar gases, and *non-AMO* crystals (e.g. Lozovik et al.)

Properties & Features …

Trapped Ions

Coulomb: $\dfrac{e^2}{R}$

- repulsive long range interaction
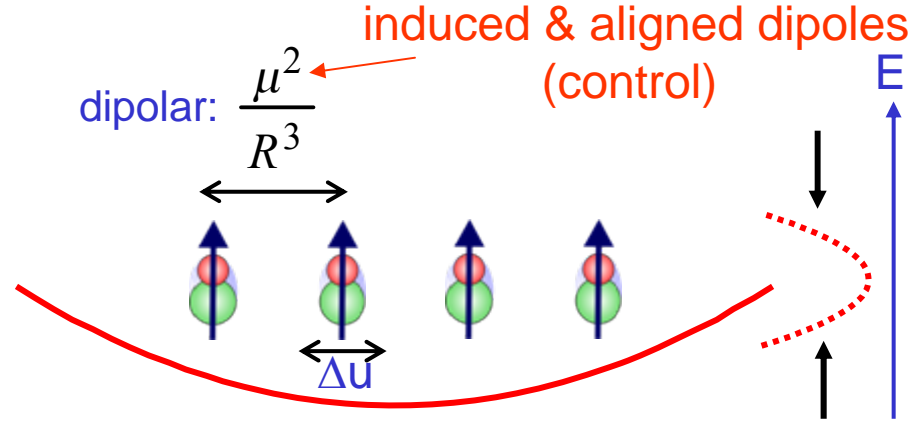
  Coulomb / 3D

- crystal forms for …

$$r_c = \frac{E_{\text{pot}}}{E_{\text{kin}}} \equiv \frac{e^2/R}{\hbar^2/mR^2} \sim R \gg 1$$

  Wigner crystal @ low density

- melting (?)

  ~~BEC~~

Trapped Polar Molecules

dipolar: $\dfrac{\mu^2}{R^3}$

induced & aligned dipoles (control)

E

Δu

repulsive in 1D & 2D trap

stability!?

$$r_d = \frac{\mu^2/R^3}{\hbar^2/mR^2} \sim \frac{1}{R} \gg 1$$

crystal @ high density
(high density ensembles / lattices)

$$\Delta u \sim 0.1\ldots0.2R$$
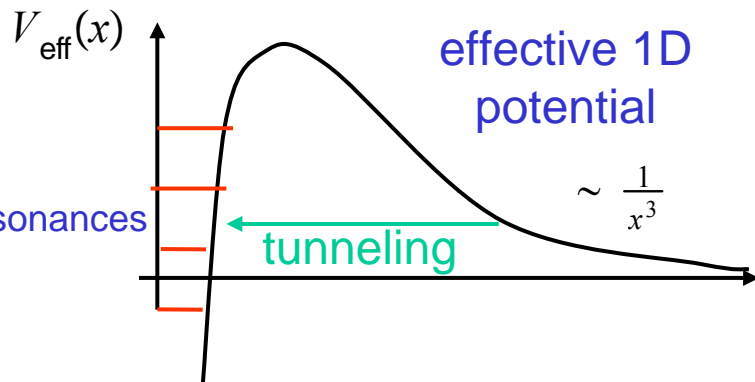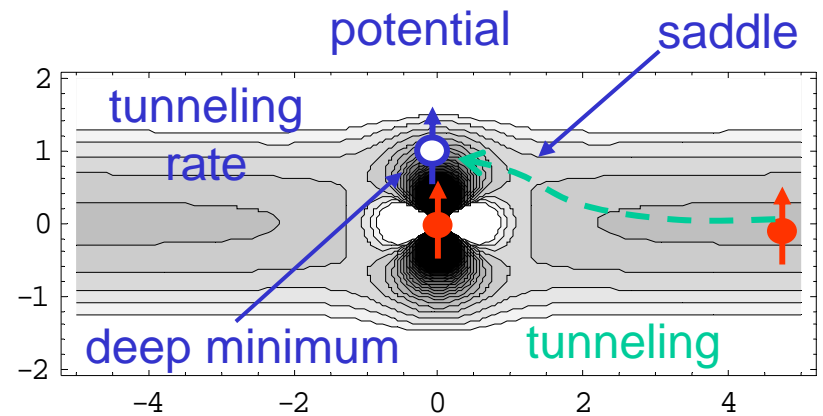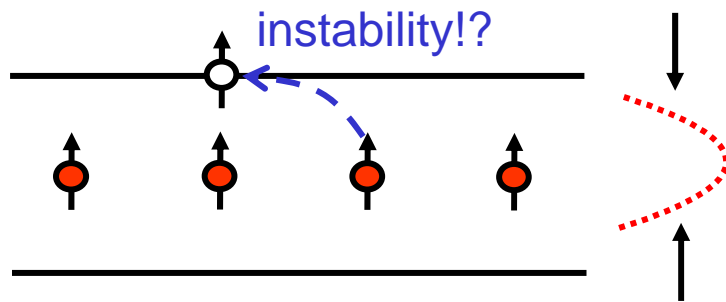
Lindemann: dipolar BEC

Hamiltonian

- Hamiltonian in 2D

$$H_{2D} = \sum_i \frac{\vec{p}_i^2}{2m} + V_T(\vec{r}_i) + \sum_{ij} V_d(\vec{r}_i - \vec{r}_j) + U_s(\vec{r}_i - \vec{r}_j)$$

"In-plane" confinement
(quadratic trap or box)

Dipole-dipole
int.: $\sim \mu^2/r^3$

Short-range

- tunneling / instability

instability!?

potential

saddle

tunneling
rate

deep minimum

tunneling

$V_{\text{eff}}(x)$

effective 1D
potential

$\sim \frac{1}{x^3}$

resonances

tunneling

$$\Gamma = \omega_p e^{-(r_d \frac{a}{a_\perp})^{2/5}}$$

$a/a_\perp > 10$

$r_d > 15$

oscillation
frequency

$\omega = (\mu^2/mR^5)^{1/2}$

Sketch of Phase Diagram

Homogeneous system in 2D

superfluid density

Superfluid

- ● N=25
- ◆ N=50
- ■ N=100

crystal

Transition point: $r_d \equiv Dm(2\pi)^2 / h^2 a \approx 13 - 15$

2D *angle-averaged* **pair correlation function**

N=100

— r_d=9.68
— r_d=19.36

Liqu

Crystal

— r_d=1, N=25
— 10

1D (r_d~0.1, **Lozovik et al.**)

Dipoles in 2D Harmonic Trap

$$H = \sum_{i=1}^{N} \left[\frac{\mathbf{p_i^2}}{2m} + \frac{m\omega^2 \mathbf{r_i^2}}{2} \right] + \sum_{i<j} \frac{D}{|\mathbf{r_i} - \mathbf{r_j}|^3}$$

$$r_0 = (D/m\omega^2)^{1/5} \quad l_{ho}^2 = \hbar/(m\omega)$$
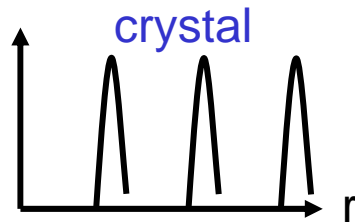
$$r_s = r_0/l_{ho}$$

- New Features:
  - Shell structure
    - Indication of two-stage Crystal/SF melting transition
    - Orientational melting
  - Radial melting
  - "Magic Numbers"

Why?

Self-Assembled Crystals

Optical Lattices

- interest *per se*
  - quantum phase / transitions
- high-density stable ensembles

- Mott insulator

crystal

r

laser

atoms / molecules

switch off (destructive) short range interactions / collisions

- lattices parameters / dynamics
  - small lattice constants ☺
  - phonons

- …
  - spacing: laser wave length λ

superradiance

Potential future applications … ⟷ optical lattices

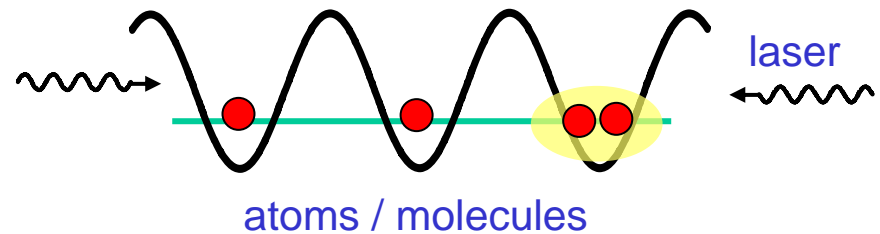- spin lattice toolbox
  - add spin degree of freedom & engineer spin dependent interactions
- Hubbard models with the new lattices

small lattice constants ☺

2. particles moving in this lattice will see a honey comb lattice= Hubbard model

1. dipoles generate
a 2D periodic lattice
= trap

shallow trap: quasi-free   strong trap: tight binding

- bi- and multilayer systems

SF/C Transition point in homogeneous systems:

$$r_d^{\ c} \equiv Dm(2\pi)^2 / h^2 a$$

$$= d^2 m\pi / h^2 a\varepsilon_0 \approx 13-15$$

Polar molecules

Examples: CaF, CaCl, SrO

Obtain a crystal for
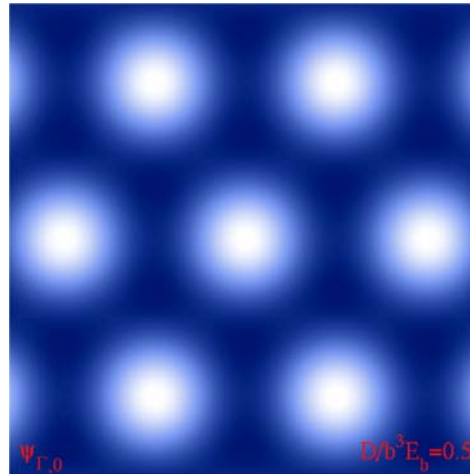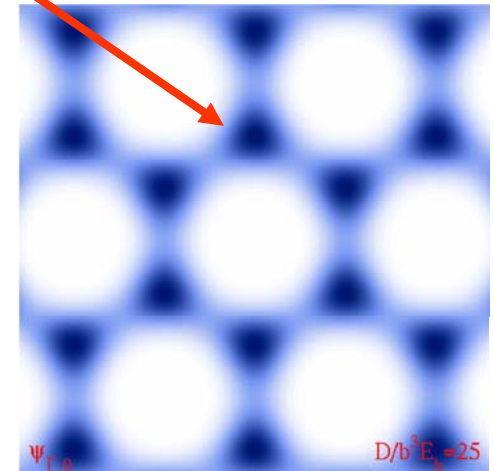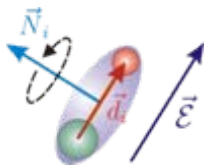
$$a < a_c = \frac{d^2 m\pi}{(h^2\varepsilon_0)r_d^{\ c}}$$

Requirements on confinement in z-direction:
$E_{int}(a_c) < \hbar\,\omega_z$

	CaF	CaCl	SrO
m[amu]	59	75 77	103
d[D]	3.07(7)	4.265(3)	8.9
a_c[nm]	568.5	1326.5 1361.9	8180.3
E_int(a_c)[KHz]	7.9	1.14 1.05	0.021

Ok! (at a_c[nm] row)
Ok! (at $E_{int}(a_c)$ row)

Preparation from optical lattice: $E_{int}(\lambda/2) < \hbar\,\omega_z$
$r_d(\lambda/2) > r_d^{\ c}$

For CaF: $\lambda/2 \sim 250$ nm,
$E_{int}(\lambda/2) \sim 93$ KHz
$r_d \sim 34 > r_d^{\ c}$

Rydberg states

Example: Hydrogen Stark states:

• **Stark splitting:** $\Delta E_{nqm} = \frac{3}{2}ea_0\mathcal{E}nq$

with $q = n-1,\ldots-(n-1)$

• **effective dipole moment:**
$\mu_{eff} = \frac{3}{2}ea_0\frac{\Omega^2}{4\Delta^2}nq \propto n^2$
use $q = n-1$

• **Condition for crystal formation:**
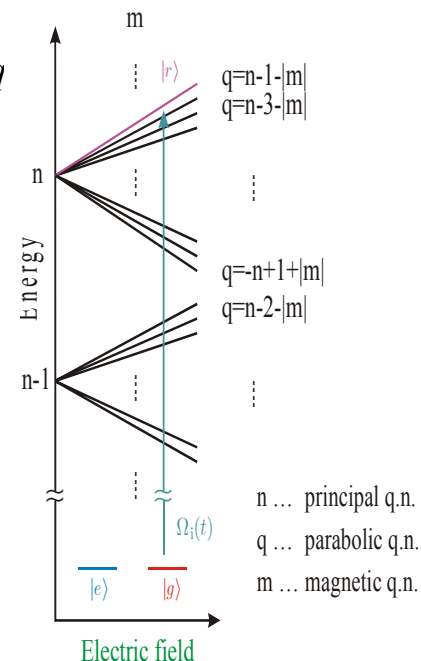
$$a_{Ryd} \ll a < a_c$$

$a_{Ryd} = \frac{3}{2}a_0 n^2$
(size of Rydberg state)

$a_c \equiv \frac{\mu_{eff}^2 m}{4\pi\varepsilon_0 h^2 \gamma_c}$

(Kin. energy = pot. Energy)

• **Example: (values for Hydrogen)**

	$n=20, \frac{\Omega}{\Delta}=0.2$	$n=20, \frac{\Omega}{\Delta}=0.1$
$a_{Ryd}\ [nm]$	31	31
$a_{c,2D}\ [nm]$	209	13
$a_{c,1D}\ [nm]$	3.1×10^4	2×10^3
$\mu_{eff}\ [D]$	14.5	3.6
$\tau\ [\mu s]$	50	200
$E_{int}(a_c)$	3.4 MHz	0.9 GHz

m

|r⟩ q=n-1-|m|
      q=n-3-|m|

n

q=-n+1+|m|
q=n-2-|m|

n-1

$\Omega_i(t)$

|e⟩ |g⟩

Energy

Electric field

n … principal q.n.
q … parabolic q.n.
m … magnetic q.n.

Example: Rb⁸⁷ Crystal for
$a < a_c \sim 1.1\ \mu m$

Preparation from optical lattice:
$\lambda/2 = a = 390$ nm,
$E_{int}(\lambda/2) \sim 33$ KHz
$r_d \sim 44 \rightarrow$ **Ok!**

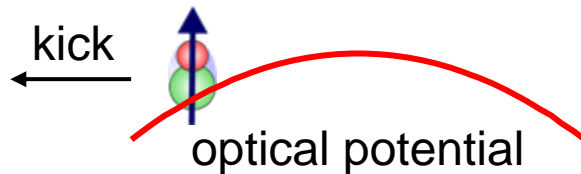• **Other requirements: dipole interaction < Level splitting**

Preparation of a Crystal

- Preparation from an optical lattice:
  - e.g. from a Mott state: switch optical lattice off, and dipoles on
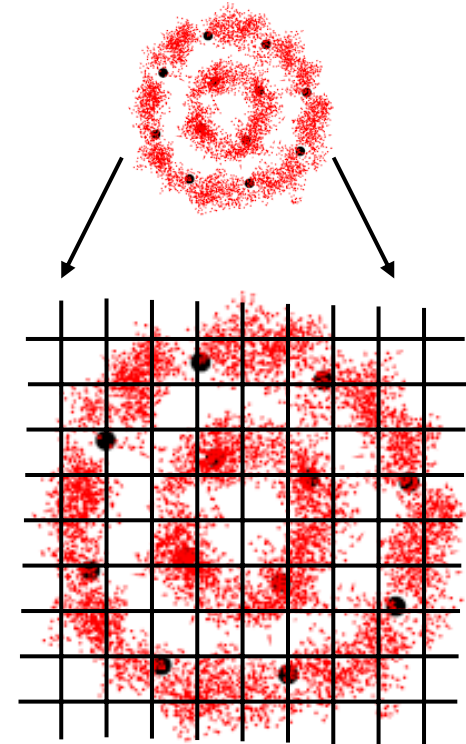
Detection of the shell-structure in real space

- "Magnifying lens" to see the shell structure

  - defocusing „lens"
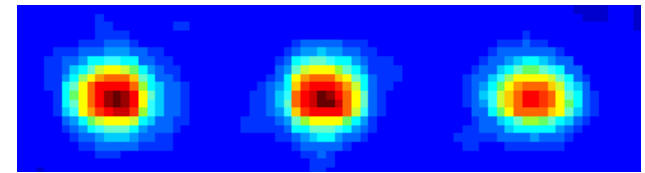
    kick

    optical potential
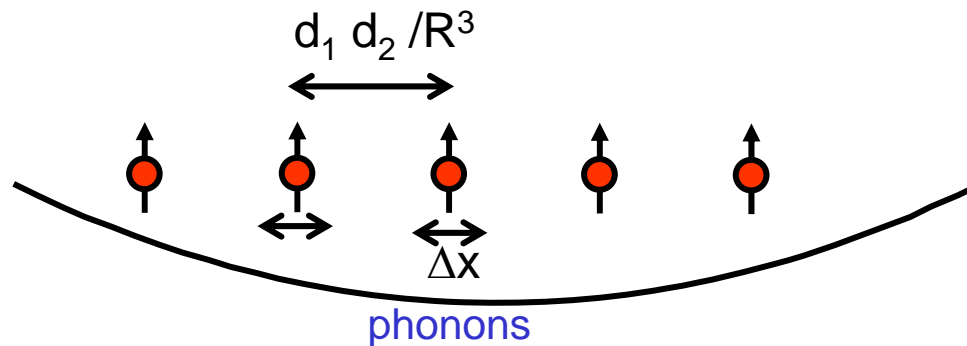
  - expand
  - focus = recapture, e.g. in optical lattice

Crystals for Quantum Computing

compare: ionic Coulomb crystal

- Ion trap like quantum computing with phonons as a bus; moving molecules

$$d_1\ d_2\ /R^3$$

Δx

phonons

(breathing mode indep of # molecules)

ion trap like qc, however:

✓ dipole moment variable

✓ spin dependent dipole

✓ qu melting / quantum statistics

✓ addressing (?)

- Ensemble memory: dephasing / avoiding collision dephasing in a 1D and 2D WC
    - ensemble qubit in 2D configuration
    - [there is an instability: rotationally excited qubit -> spin waves]

- … vs. coupling *single* molecules to a strip line cavity
    - CQED type quantum computing

Molecular ensembles + Circuit CQED

R. Schoelkopf, M. Devoret,
S. Girvin (Yale)

1. strong CQED with superconducting circuits

- Cavity QED

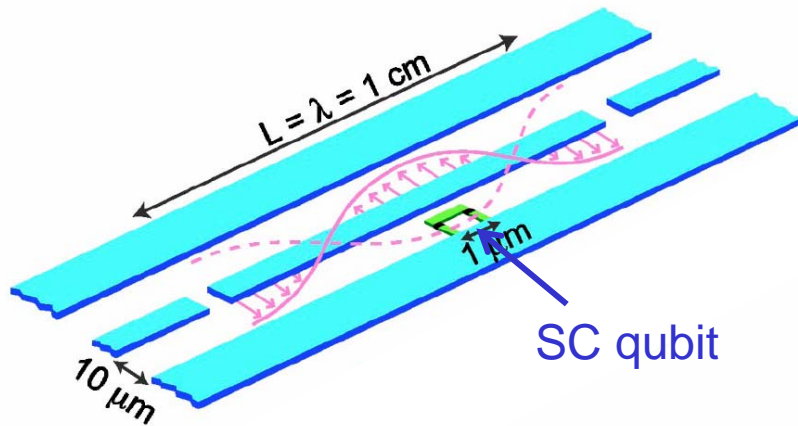$L = \lambda = 1\ cm$

$1\ \mu m$

SC qubit

$10\ \mu m$

$$H = \omega_c a^\dagger a + \frac{1}{2}\omega_q(t)\sigma_z + g(a\sigma_+ + \text{h.c.})$$

Jaynes-Cummings

parameters:

cavity frequency $\omega_c \sim 2\pi \times 10$ GHz

cavity damping $\kappa \sim 2\pi \times 1\ldots 0.01$ MHz — good cavity

SC qubit - cavity coupling $g \sim 2\pi \times 30$ MHz — strong coupling!
(mode volume V/ $\lambda^3 \approx 10^{-5}$)

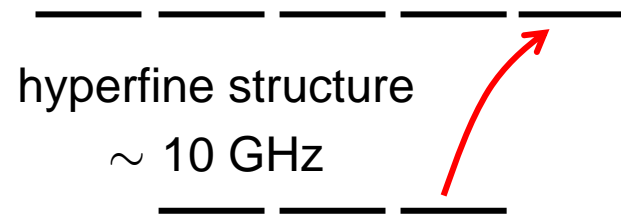SC qubit damping $\Gamma \sim 2\pi \times 1$ MHz — "not so great" qubits

- [... similar results expected for coupling to quantum dots (Delft)]
- [compare with CQED with atoms in optical and microwave regime]

2. ... coupling atoms or molecules

- **superconducting transmission line cavities**

atoms / molecules

$L = \lambda = 1$ cm

$1\,\mu m$

$10\,\mu m$

SC qubit

- Remarks:
  - time scales compatible
  - laser light + SC is a problem: we must *move* atoms / molecules to interact with light (?)
  - traps / surface ~ 10 μm scale
  - low temperature: SC, black body…

- **hyperfine excitation of BEC / atomic ensemble**

hyperfine structure
~ 10 GHz

$g \sim 2\pi \times 80$ Hz $\sqrt{\#\text{atoms}}$

- **rotational excitation of polar molecule(s)**

N=1

rotational excitations
~ 10 GHz

μ large!

N=0

$g \sim 2\pi \times 10$ KHz $\sqrt{\#\text{molecules}}$

$\sim 2\pi \times 1 \dots 10$ MHz ensemble ☺

3. Atomic / molecular ensembles: collective excitations as Qubits

- ground state

$$|g^N\rangle = |g_1\rangle|g_2\rangle\ldots|g_N\rangle$$
$$\equiv |\text{vac}\rangle \equiv |0\rangle_e$$

- one excitation (Fock state)

$$|g^{N-1}q\rangle = \frac{1}{\sqrt{N}}\sum_i^N |g_1\rangle\ldots|q_i\rangle\ldots|g_N\rangle$$
$$\equiv m^\dagger|\text{vac}\rangle \equiv |1\rangle_e$$

$[m, m^\dagger] \approx 1$ harmonic oscillator

- two excitations ... eliminate?
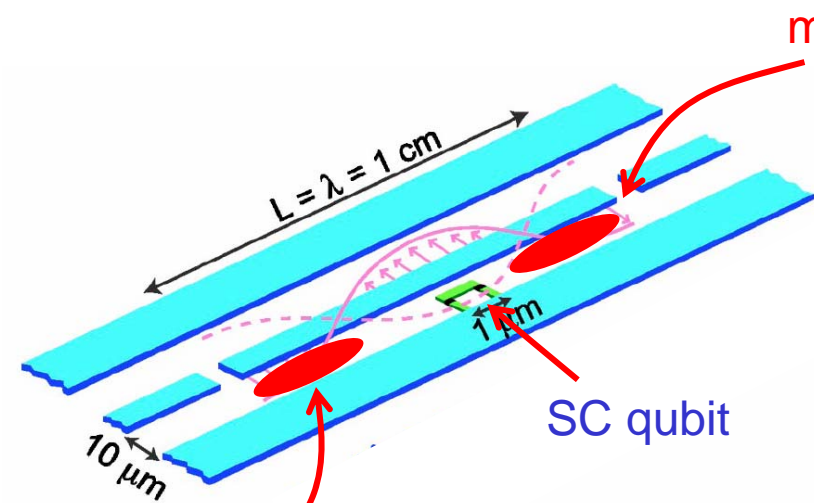 – in AMO: dipole blockade, measurements ...

microwave

microwave

nonlinearity due to Cooper Pair Box.

etc.

- also: ensembles as continuous variable quantum memory (Polzik, ...)
- collisional dephasing (?)

4. Hybrid Device: solid state processor & molec memory

molecules:
qubit 1

L = λ = 1 cm

1 μm

SC qubit

10 μm

molecules:
qubit 2

time independent

$$H = \sum_{i=1}^{2} \omega_e(t) m_i^\dagger m_i + \omega_c a^\dagger a + \frac{1}{2}\omega_q(t)\sigma_z + g_c(a\sigma_+ + \text{h.c.}) + \sum_{i=1}^{2} g_{i;e}(t)(am_i^\dagger + \text{h.c.})$$

ensemble
qubits

solid state system

swap molecule -
cavity

+ dissipation (master equation)

5. Examples of Quantum Info Protocols

- SWAP

$$(\alpha|0\rangle_{cq} + \beta|1\rangle_{cq})\ |0\rangle_c|0\rangle_e$$

Cooper Pair

$$\leftrightarrow |0\rangle_{cq}(\alpha|0\rangle_c + \beta|1\rangle_c)\ |0\rangle_e$$
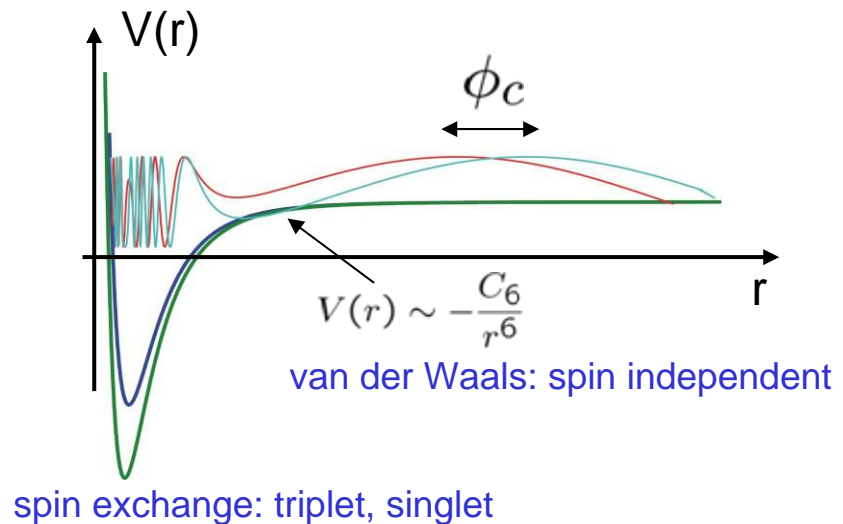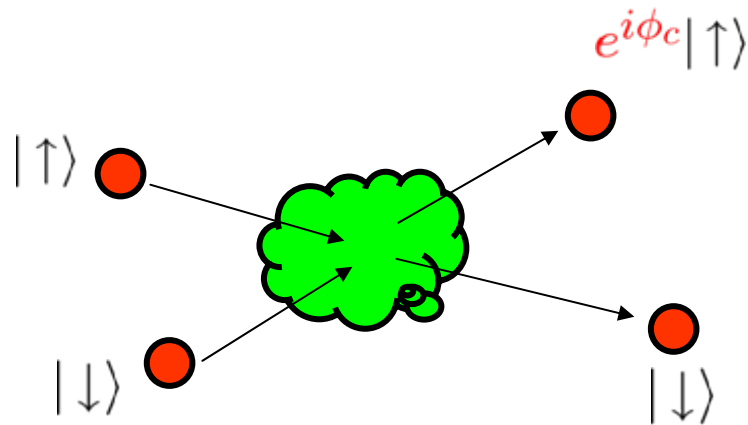
cavity (bus)

$$\leftrightarrow |0\rangle_{cq}|0\rangle_c\ (\alpha|0\rangle_e + \beta|1\rangle_e)$$

molec ensemble

- Single qubit rotations via SC qubit

- Universal 2-Qubit Gates via SC qubit

- measurement via ensemble / optical readout or SC qubit / SET

Atomic ensembles complemented by deterministic entanglement operations

[Study of Decoherence of Molecular Ensemble Qubit : quantum memory & gate operations]

$e^{i\phi_c}|\uparrow\rangle$

$|\uparrow\rangle$

$|\downarrow\rangle$

$|\downarrow\rangle$

V(r)

ϕ_c

$V(r) \sim -\frac{C_6}{r^6}$

r

van der Waals: spin independent

spin exchange: triplet, singlet

Collision rates (estimate): CaF, n=10^{12} cm^{-3}

T= 1 μK: (s-wave scattering)

$$\bar{a} = \sqrt[4]{mC_6/\hbar^2} \approx 450 a_B$$

$\gamma_{col} \approx 50$ Hz

T= 1 mK: (unitarity limit)

$$\sigma \le \frac{4\pi}{k^2} \sum_{l=0}^{l_{max}} (2l+1)$$

$\gamma_{col} < 400$ Hz

Quantum kinetic theory: Bradley, Gardiner, J. Phys. B 35, 4299 (2002)
detailed collision physics: J. Bohn, R. Krems, …

Summary: QIPC & Cond Mat with Polar Molecules

- single molecules / molecular ensembles

- coupling to optical & microwave fields
    - trapping / cooling
    - CQED (strong coupling)
    - spontaneous emission / engineered dissipation
- interfacing solid state / A<u>M</u>O & microwave / optical
    - strong coupling / dissipation
- collisional interactions
    - quantum deg gases / Wigner crystals (ion trap like qc)
    - WC / dephasing