Anomalous Behavior of Spin Systems with Dipolar Interactions

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SFB TRR21:
Tailored quantum matter
B6/B8

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Dipole-dipole interactions

Cold atoms

- magnetic dipole moments between electron spins: $d \sim \mu_B = \frac{e\hbar}{2m_e c}$

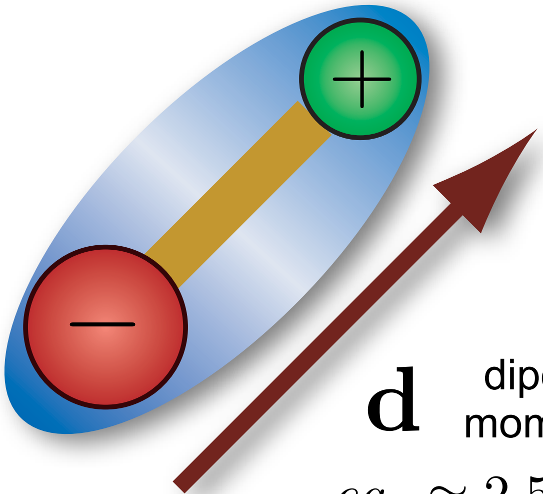
- Cr/Dy one of the strongest magnetic dipole moments

Polar Molecules

- permanent dipole moment: $d \sim ea_0 = \frac{e\hbar}{m_e c\alpha}$

- interactions are increased by $1/\alpha^2 \sim 137^2$

- rotational energy



$\mathbf{d}$ dipole moment

$ea_0 \approx 2.5\mathrm{Debye}$

Rydberg atoms

- electric dipole moment $d \sim n^2 ea_0$

- similar internal structure as polar molecules

- finite life time

n: principal quantum number, $n \sim 10 - 100$

Internal structure of polar molecule

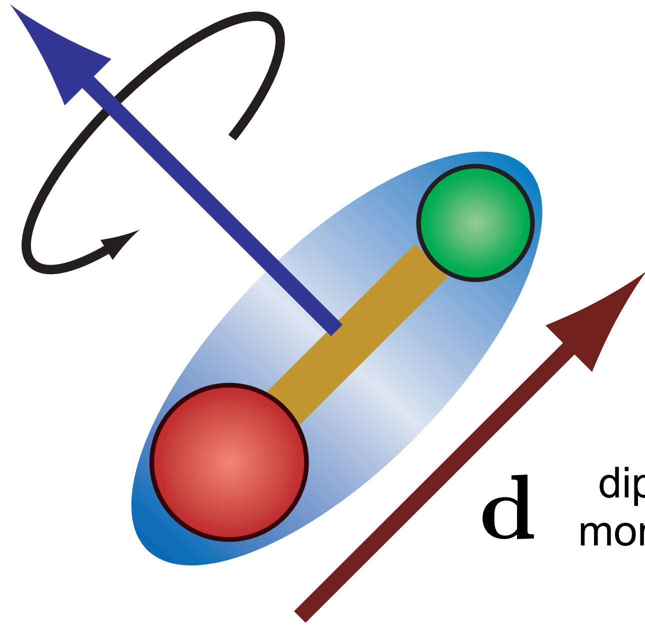
Low energy description

- rigid rotor in an electric field

$$H^{(i)}_{\rm rot} = B\mathbf{N}_i^2 - \mathbf{d}_i\mathbf{E}(t)$$

$\mathbf{N}_i$: angular momentum

$\mathbf{d}_i$: dipole operator



$BN_i(N_i+1)$

$N = 2$ — — — — —

$N = 1$ — — —

$N = 0$ —

$N=1$ to $N=0$: $\sim 20\text{GHz}$

Accessible via microwave

- anharmonic spectrum
- electric dipole transition

$$\Delta N = \pm 1 \qquad \Delta m_z = -1, 0, 1$$

- microwave transition frequencies
- no spontaneous emission

Dipole-dipole interaction

$$V_{ij}(\mathbf{r}_i - \mathbf{r}_j) = \frac{\mathbf{d}_j \mathbf{d}_i - 3(\mathbf{d}_i \cdot \mathbf{n}_{ij})(\mathbf{d}_j \cdot \mathbf{n}_{ij})}{|\mathbf{r}_i - \mathbf{r}_j|^3}$$

$$\mathbf{n}_{ij} = \frac{\mathbf{r}_i - \mathbf{r}_j}{|\mathbf{r}_i - \mathbf{r}_j|}$$

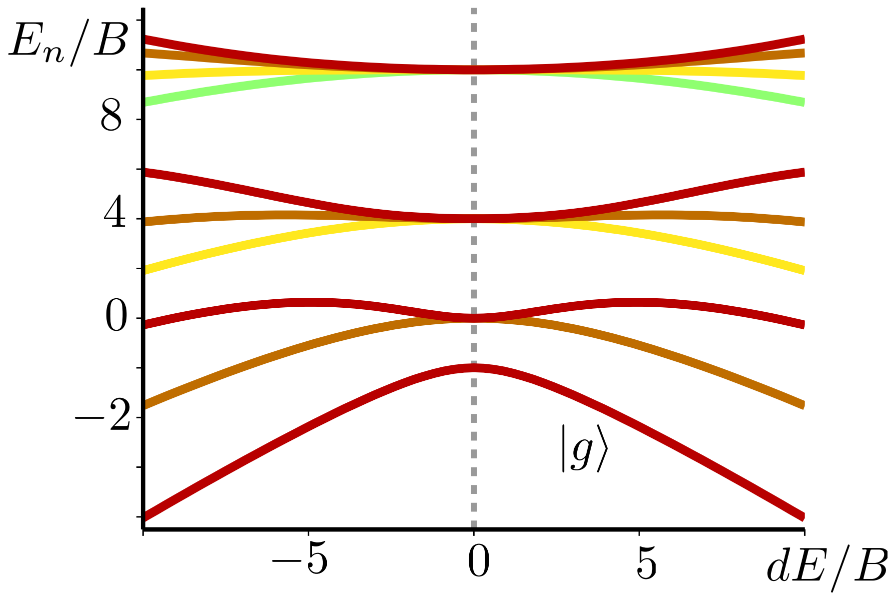
Static electric field

- internal Hamilton

$$H_{\rm rot}^{(i)} = B\mathbf{N}_i^2 - \mathbf{d}_i \mathbf{E}$$

- finite averaged dipole moment

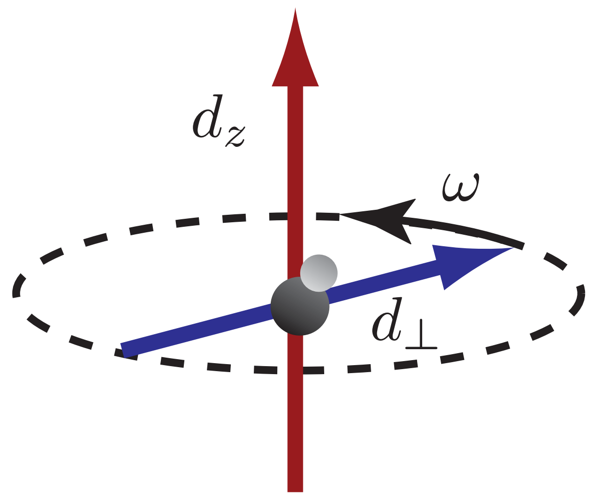
$$D = |\langle g|\mathbf{d}_i|g\rangle|^2 \leq d^2$$



Microwave coupling

- rotating dipole moments via coupling of rotational states with microwave fields

- design interaction potentials



Dipole-dipole interaction

$$\mathbf{n}_{ij} = \frac{\mathbf{r}_i - \mathbf{r}_j}{|\mathbf{r}_i - \mathbf{r}_j|}$$

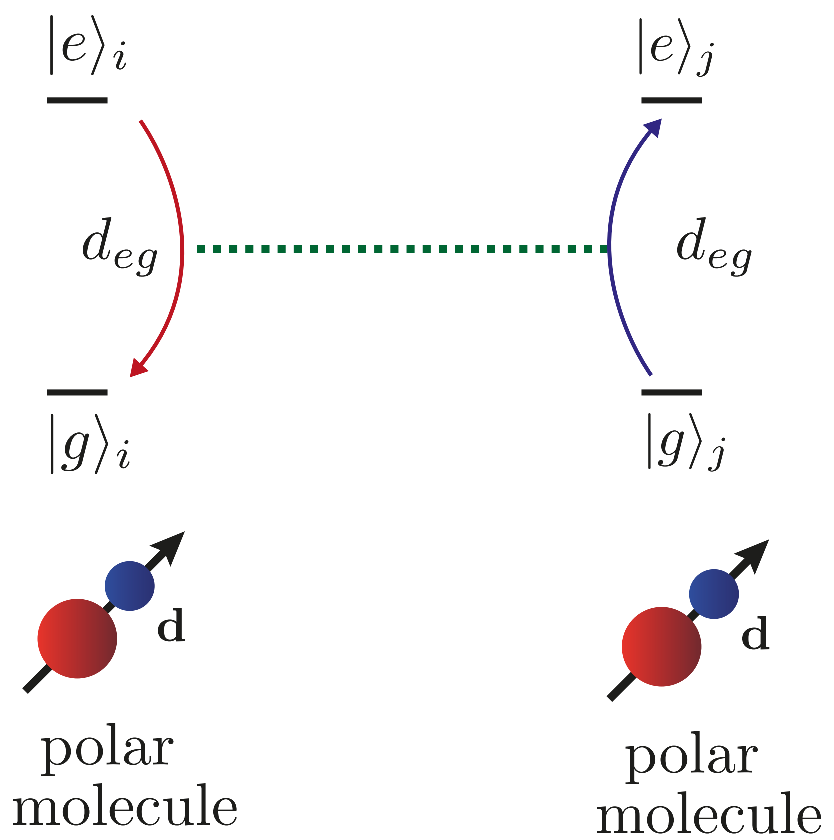
$$V_{\mathrm{ij}}(\mathbf{r}_i - \mathbf{r}_j) = \frac{\mathbf{d}_j \mathbf{d}_i - 3\,(\mathbf{d}_i \cdot \mathbf{n}_{ij})\,(\mathbf{d}_j \cdot \mathbf{n}_{ij})}{|\mathbf{r}_i - \mathbf{r}_j|^3}$$

Resonant exchange interaction

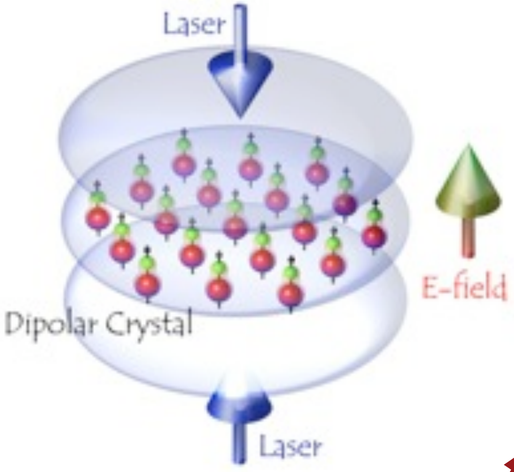
- virtual exchange of a microwave photon

- strong interaction even in absence of electric field

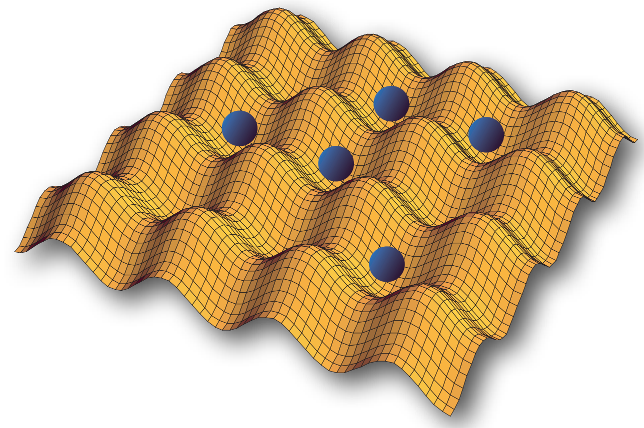
$$V_{\mathrm{eff}} \sim \frac{d^2}{|\mathbf{r}|^3}$$



Many-body physics

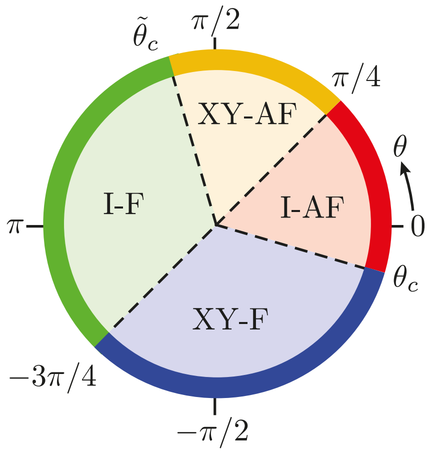


Crystalline phase
(Büchler et al, PRL 2007)

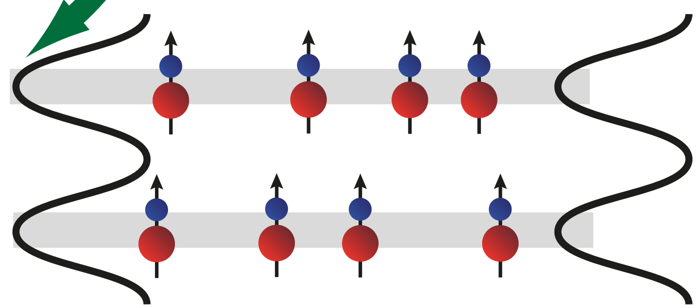


Novel phases in optical lattices
(Pollet, PRL 2010, Bonnes et al, NJP, 2010, Bühler et al, PRA, 2011)

Strongly correlated many-body physics



Magnetism and t-J model
(Goroshkov et al. PRL)



Bilayer systems

How to cool polar molecules?

Cooling of polar molecules

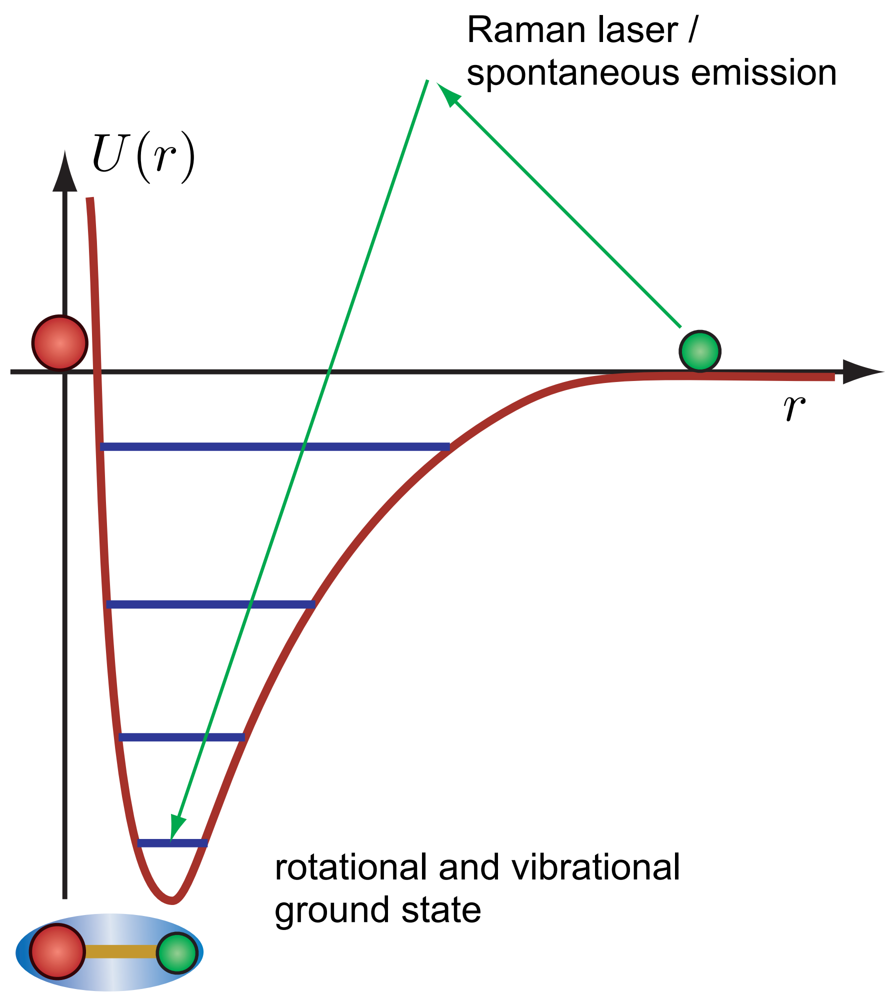
D. De Mille (Yale), J. Doyle (Harvard), G. Rempe(Munich), G. Meijer (Berlin), F. Merkt (Zürich)

- stark deceleration
- buffer gas cooling
- etc ....

Coherent Formation

- heteronuclear mixtures of cold atomic gases
- coherent formation of molecules using Stimulated Raman Adiabatic Passage (STIRAP)

Jun Ye et al, Science (2008) : K Rb
M. Weidemüller et al, PRL (2008) : Li Cs
H. C Nägerl, et al., Science (2008): CsCs

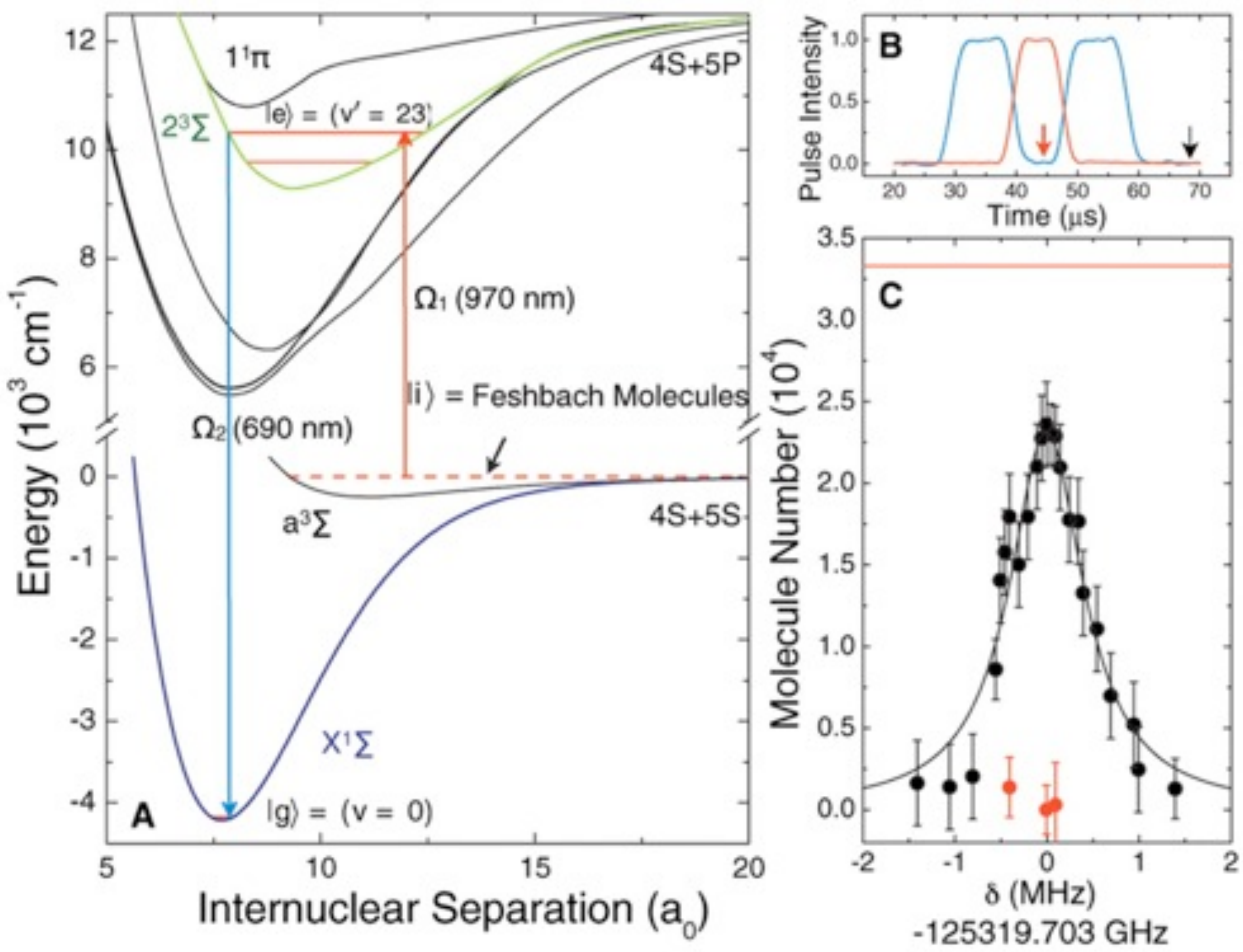


Coherent Formation

- formation of weakly bound molecules via a Feshbach resonance

- Stimulated Raman Sdiabatic Passage (STIRAP) via an electronic excited state

Jun Ye et al, Science (2008) : K Rb



Why is dipolar different?

Low energy scattering in 3D

- dipolar interactions are not described by a single s-wave scatering length
- observation in atomic BEC with large magentic moments

Absence of a first order phase transition (Spivak Kivelson, PRB 2004)

- dipolar interactions prevent first order phase transition:

$$E \sim L - \alpha L \log L$$

domain wall boundary — dipole interaction

- second order transition via the formation of large scale patterns with domains

Dipolar dispersion

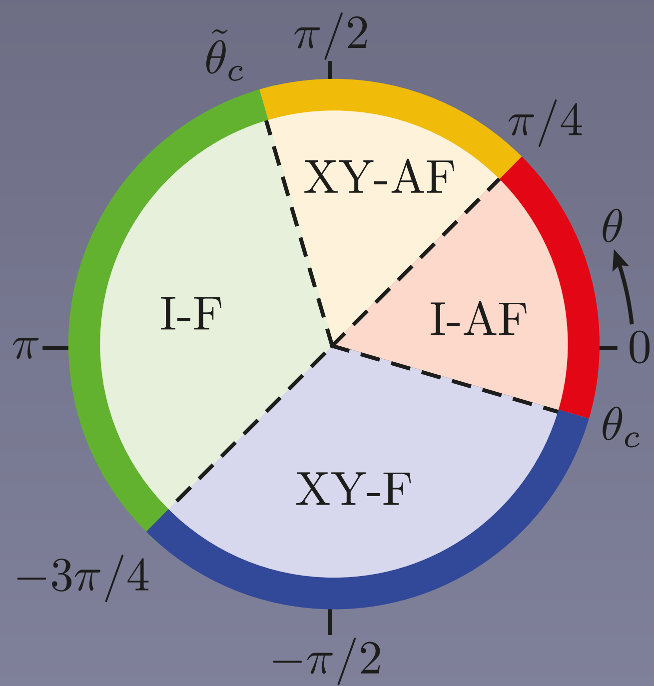
- lattice Fourier transformation in two dimension

$$\epsilon_{\mathbf{q}} = \sum_{i\neq 0} \frac{e^{i\mathbf{R}_i \mathbf{q}}}{|\mathbf{R}_i|^3} = -2\pi a|\mathbf{q}| \mathrm{Erfc}(a|\mathbf{q}|/2\sqrt{\pi}) + 4\pi \left(e^{-\frac{a^2|\mathbf{q}|^2}{4\pi}} - \frac{1}{3} \right)$$

$$+2\pi \sum_{i\neq 0} \int_1^\infty \frac{d\lambda}{\lambda^{3/2}} \left[e^{-\pi\lambda\left(\frac{\mathbf{R}_i}{a} + \frac{a\mathbf{q}}{2\pi}\right)^2} + \lambda^2 e^{-\frac{\pi\lambda|\mathbf{R}_i|^2}{a^2} + i\mathbf{R}_i\mathbf{q}} \right]$$

Ewald summation

Spin Systems with polar molecules



Spin Hamiltonian

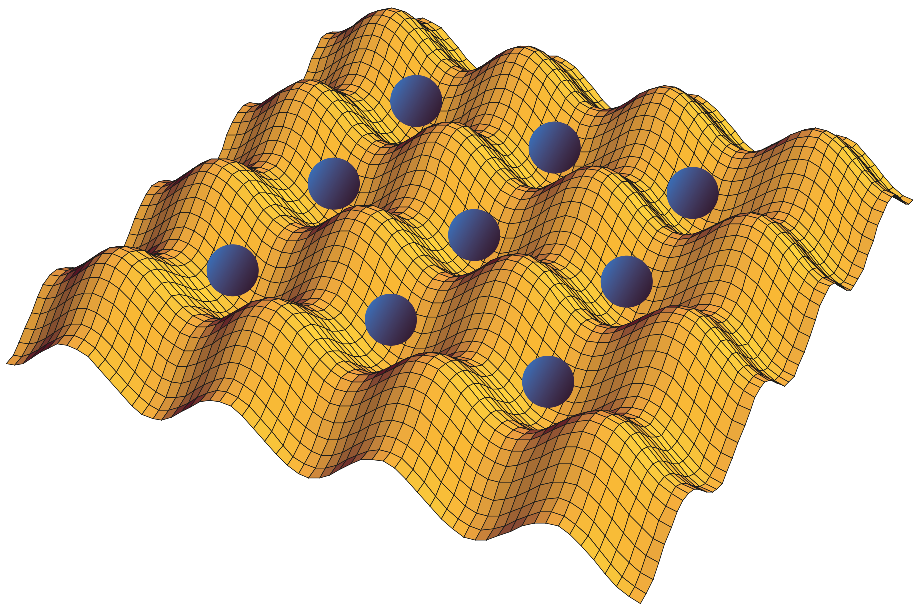
Spin Hamiltonian

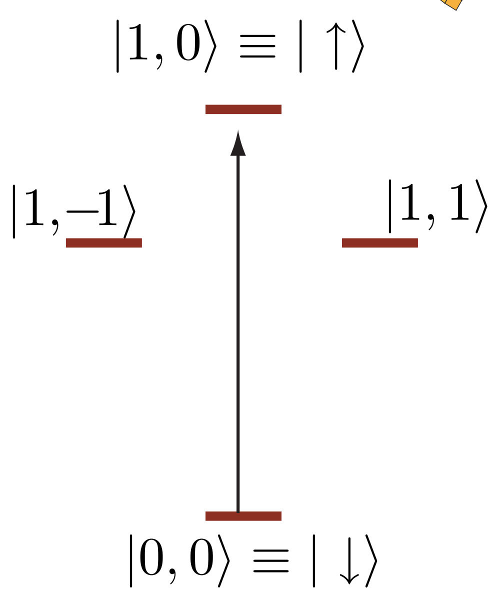
- polar molecules trapped in an optical lattice
- suppressed tunneling
- one particle per lattice site
- electric field perpendicular to the plane splits rotational excitations
- two levels: spin 1/2 system

$$|\downarrow\rangle = |0,0\rangle$$

$$|\uparrow\rangle = |1,0\rangle$$

- dipole-dipole interaction gives rise to spin Hamiltonian





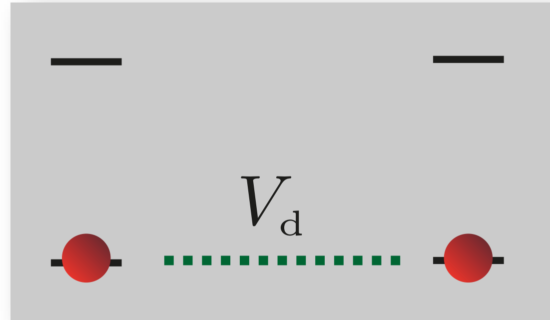
Spin Hamiltonian

Ising interactions

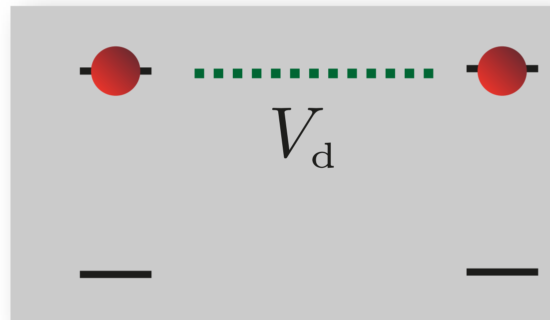
- static dipole moments within each rotational state

$$P_g^{(i)} = |g\rangle\langle g|_i$$

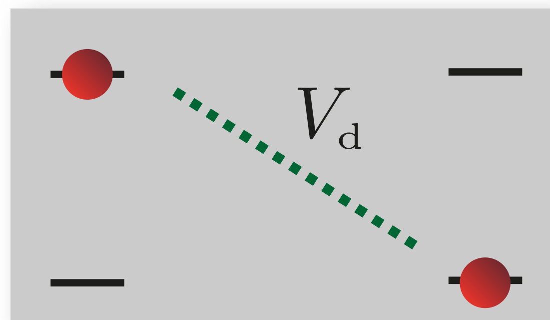
$$H = \frac{1}{2}\sum_{i\neq j}\frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3}\Big[d_g^2 P_g^{(i)} P_g^{(j)}$$



$$+d_e^2 P_e^{(i)} P_e^{(j)}$$



$$+2 d_e d_g P_e^{(i)} P_g^{(j)}\Big]$$



$$= \frac{1}{2}\sum_{i\neq j}\frac{J_z}{|\mathbf{R}_i - \mathbf{R}_j|^3}\sigma_z^{(i)}\sigma_z^{(j)} + \sum_i h_\mathrm{eff}\sigma_z^{(i)} + E_0$$

Ising — magnetic field — energy shift

$$J_z = (d_g - d_e)^2$$

Spin Hamiltonian

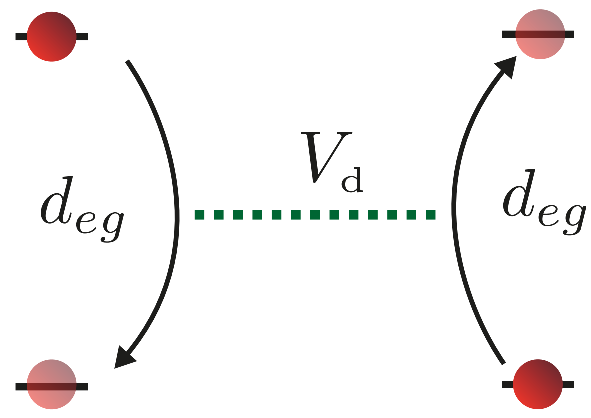
XY interactions

- resonant exchange interactions

$$\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+$$

$$H = \pm \frac{1}{2} \sum_{i \neq j} \frac{d_{eg}^2}{|\mathbf{R}_i - \mathbf{R}_j|^3} \left[\sigma_x^{(i)} \sigma_x^{(j)} + \sigma_y^{(i)} \sigma_y^{(j)} \right]$$

+ for m=0
- for m=1



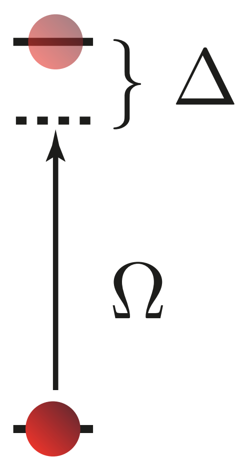
- ferro/ antiferro - magnetic interaction depending on excited rotational state

Magnetic field

- microwave field coupling ground and excited state

- rotating frame

$$H = \frac{\hbar}{2} \sum_i \left[\Delta \sigma_z^{(i)} + \Omega \sigma_x^{(i)} \right] = \sum_i \mathbf{h} \cdot \mathbf{S}^{(i)}$$



Spin Hamiltonian

XXZ model

- dipolar decay of the coupling parameters

- highly tunable from ferro- to anti-ferromagnetic coupling

$$H = J \sum_{i \neq j} \frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3} \left[\cos\theta \sigma_z^i \sigma_z^j + \sin\theta \left(\sigma_x^i \sigma_x^j + \sigma_y^i \sigma_y^j \right) \right]$$

$\theta = 0, \pi$

$\theta = \pm\pi/2$

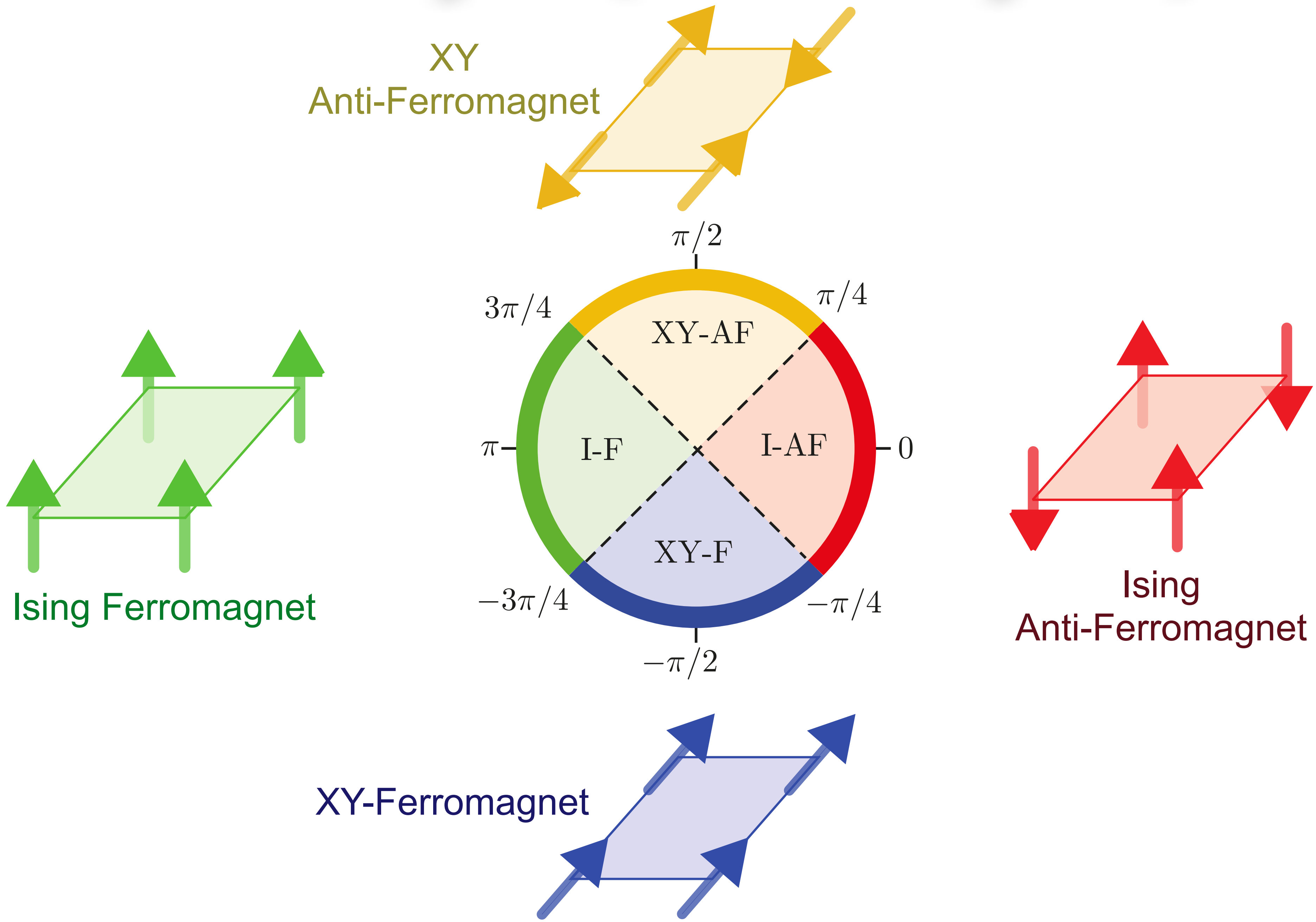
Ising (anti-) ferromagnetic coupling

$$H = \mp J \sum_{i \neq j} \frac{\sigma_z^i \sigma_z^j}{|\mathbf{R}_i - \mathbf{R}_j|^3}$$

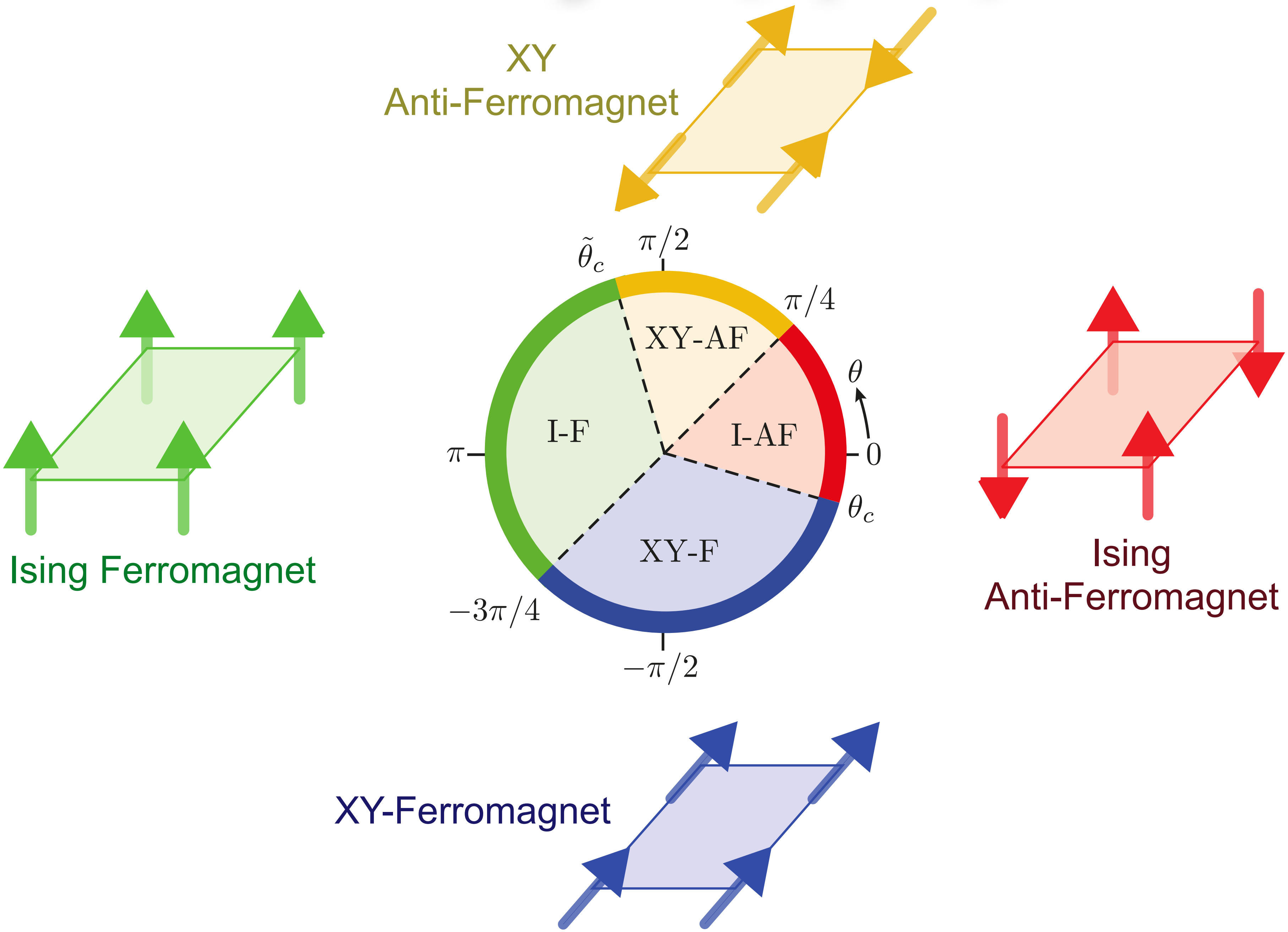
XY (anti-) ferromagnetic coupling

$$H = \mp J \sum_{i \neq j} \frac{\sigma_x^i \sigma_x^j + \sigma_y^i \sigma_y^j}{|\mathbf{R}_i - \mathbf{R}_j|^3}$$

Phase Diagram (nearest neighbor)



Phase Diagram (dipolar)



Spin wave analysis

Holstein-Primakoff transformation

- express spin operators in terms of bosonic operators

$$S_z^i = \frac{\hbar}{2}\left(a_i^\dagger + a_i\right)\varphi(n_i)$$

$$S_y^i = \frac{\hbar}{2i}\left(a_i^\dagger - a_i\right)e^{i\mathbf{R}_i\mathbf{K}}\varphi(n_i)$$

choice of the operators for the XY anti-ferromagnet

factor to guarantee bosonic commutation relations

- quadratic Hamiltonian for the bosonic fields in leading order

- bogoliubov transformation diagonalizes the quadratic Hamiltonian

  - spin wave excitation spectrum

$$H = E_0 + \sum_{\mathbf{k}} E_{\mathbf{k}}\, a_{\mathbf{k}}^\dagger a_{bfk}$$

Fourier transformation

$$\epsilon_{\mathbf{q}} = \sum_{i\neq 0}\frac{e^{i\mathbf{R}_i\mathbf{q}}}{|\mathbf{R}_i|^3} \sim \epsilon_0 - 2\pi a|\mathbf{q}|$$

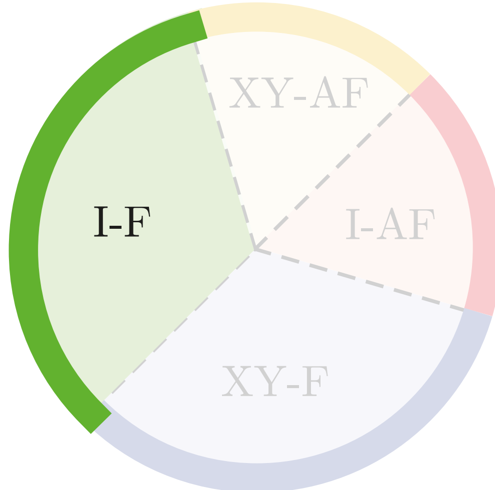
Instead of for $m\mathbf{q}^2$
short range interactions

Ising Ferromagnet

Ground state properties

- broken Z_2 symmetry
- true long range order at low temperatures

$$\langle \sigma_z^i \sigma_z^j \rangle \rightarrow m^2$$

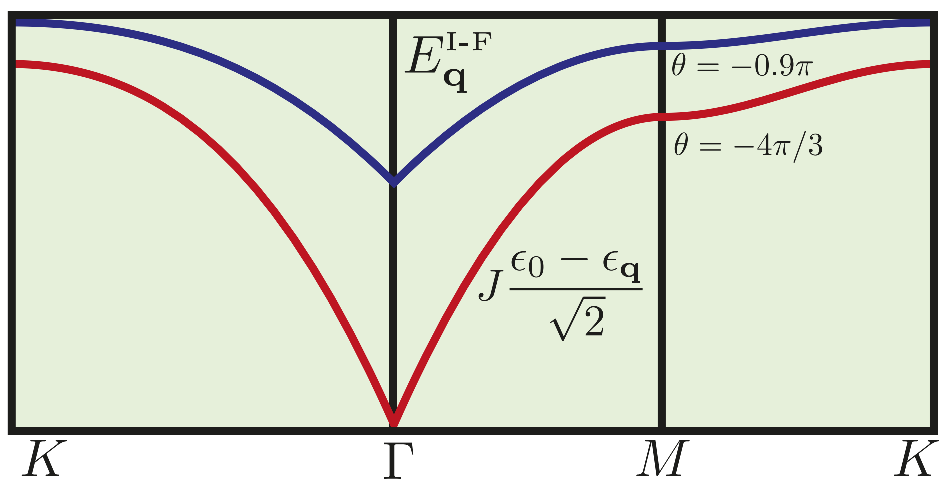


Excitations

- gapped excitations spectrum
- anomalous behavior for $|\mathbf{q}| \rightarrow 0$

$$E_{\mathbf{q}} = \Delta + \hbar c |\mathbf{q}|$$

$$c = 2\pi a J \sin\theta$$

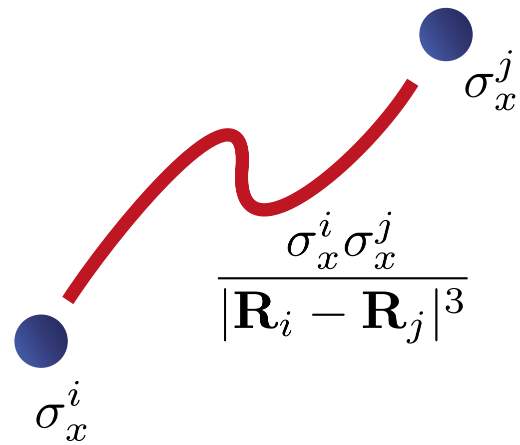


Ising Ferromagnet

Correlation functions:

- gaped system with algebraic correlation functions:

$$\langle \sigma_x^i \sigma_x^j \rangle \to \frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3}$$



lowest order perturbation theory
or high temperature expansion

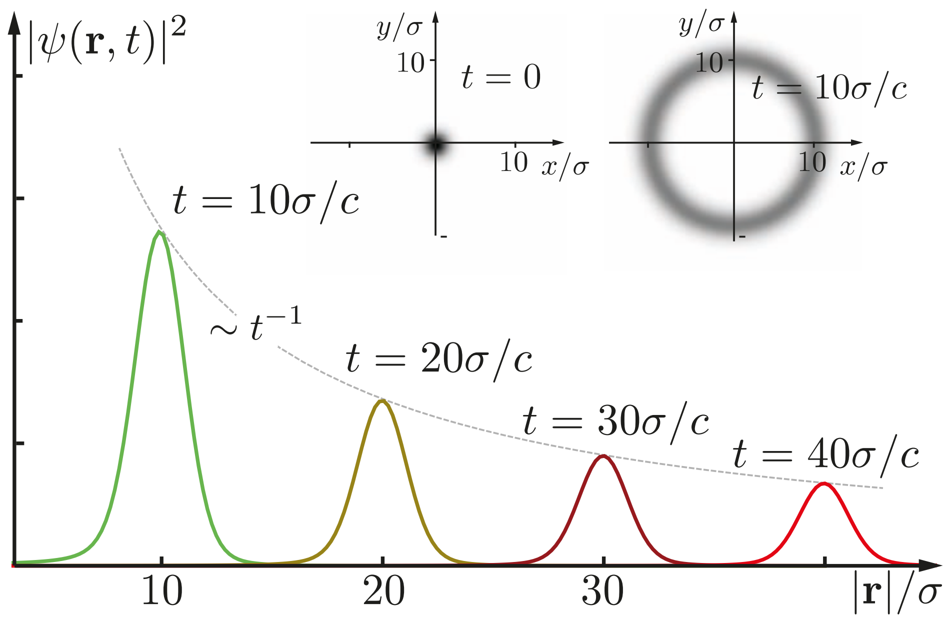
- no correlations can decay faster than the dipolar power law (remains valid for all phases)

Spin wave dynamics:

- the dynamics of localized spin waves behaves like photons

- finite velocity

- no broadening of the wave packet



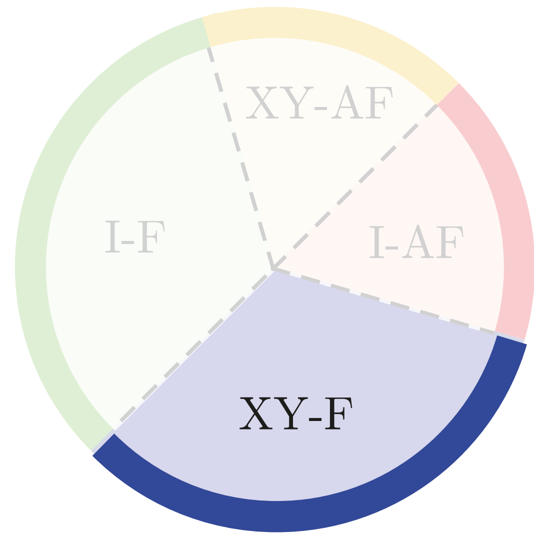
Mermin-Wagner theorem

Ground state properties

- broken U(1) symmetry

- true long range order at low temperatures

$$\langle \sigma_x^i \sigma_x^j + \sigma_y^i \sigma_y^j \rangle \to m^2$$

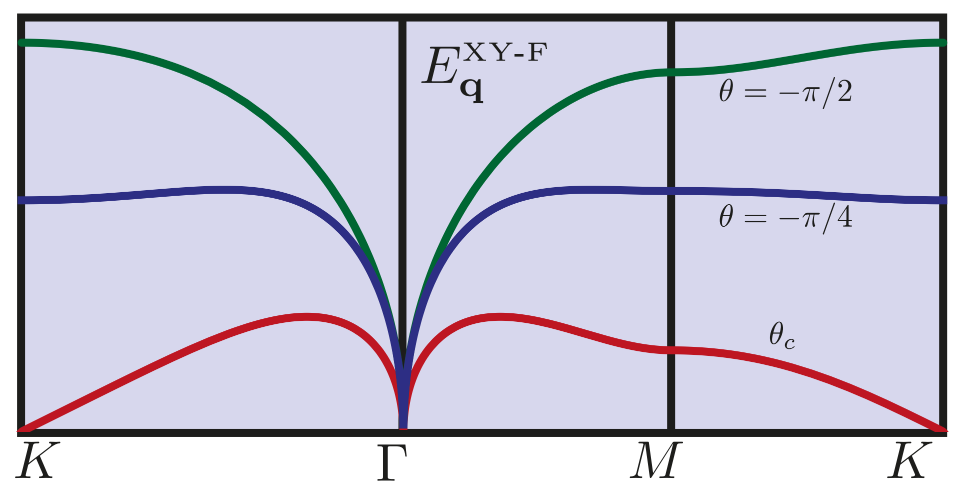


Excitations

- gapless excitations

- absence of linear Goldstone mode:

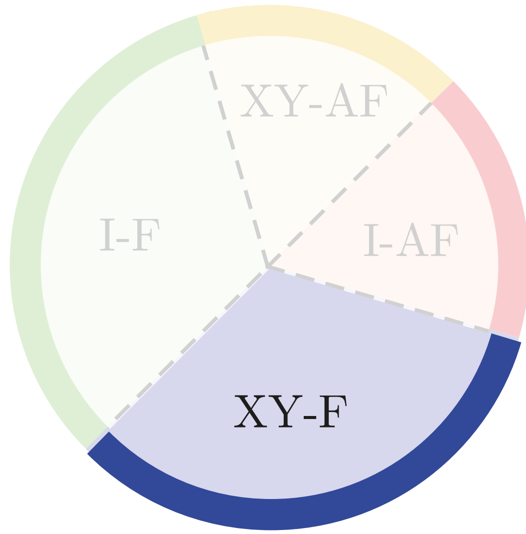
$$E_{\mathbf{q}} \sim \sqrt{|\mathbf{q}|}$$



Mermin-Wagner theorem

A continuous symmetry cannot be spontaneously broken in $d \leq 2$ dimensions at finite temperature.
(sufficiently short range interactions)

$$\sum_i V(\mathbf{R}_i)\, \mathbf{R}_i^2 < \infty$$



- Mermin Wagner theorem does not apply for dipolar interaction

$$\sum_i^{\Lambda} \frac{\mathbf{R}_i^2}{|\mathbf{R}_i|^3} \sim \begin{cases} \Lambda & d=2 \\ \log \Lambda & d=1 \end{cases}$$

Kosterlitz-Thouless transition?

- vortices are linear confining in 2D
  - no free vortices
  - only fluctuations on short scales
- thermal phase transition becomes conventional second order phase transition

Ising Anti-Ferromagnet

Ground state properties

- broken Z_2 symmetry

- true long range order at low temperatures

$$\langle (-1)^{i-j} \; \sigma_z^i \sigma_z^j \rangle \to m^2$$

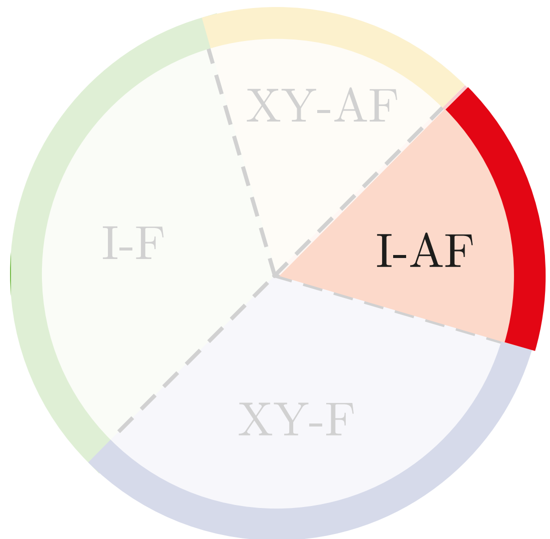
- excitation gap with linear cusp

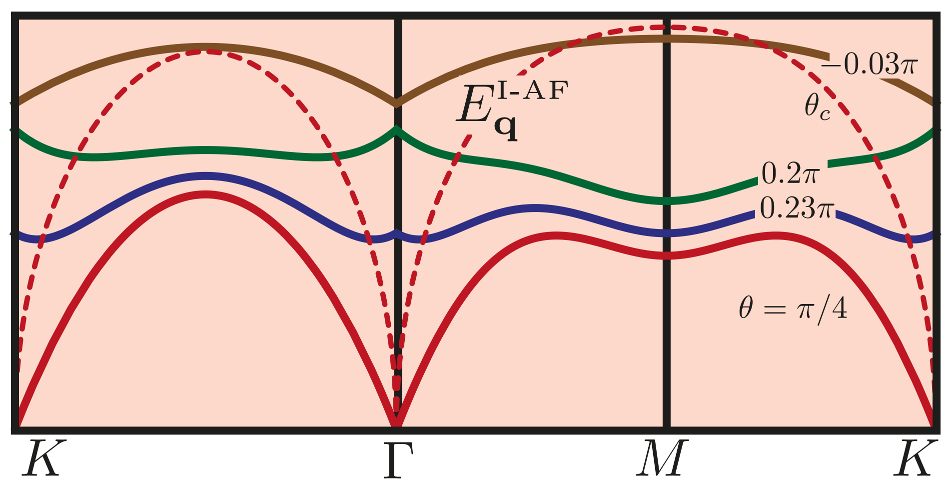
$$E_{\mathbf{q}} = \Delta + \hbar c |\mathbf{q}|$$

- algebraic correlations decaying with dipolar character:

$$\sim \frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3}$$

anti-ferromagnetic interactions provide a frustration and reduce the long-range character





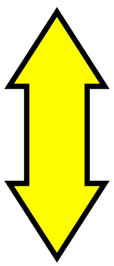
XY Anti-Ferromagnet

Ground state properties

- broken U(1) symmetry
- linear Goldstone mode
- cusp at K point
- Kosterlitz-Thouless transition:

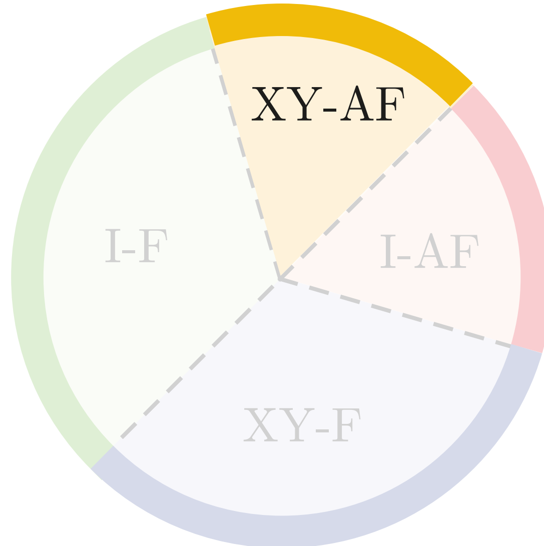
low temperatures

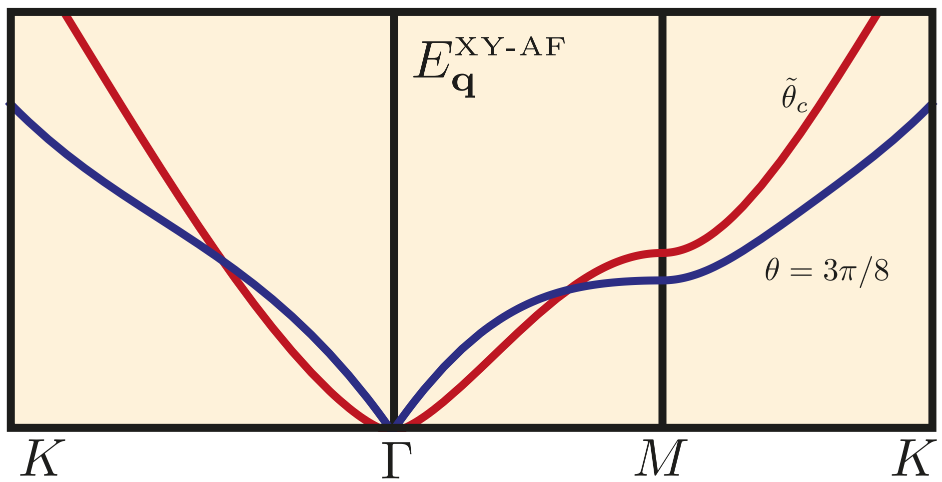
$$\sim |\mathbf{R}_i - \mathbf{R}_j|^{-\alpha}$$



high temperatures

$$\sim |\mathbf{R}_i - \mathbf{R}_j|^{-3}$$





How good is spin wave analysis?

Reduction of mean-field order

- fluctuations suppress the order parameter

- slow decay of dipolar interaction provide only weak reductions

→ good validity of spin wave analysis

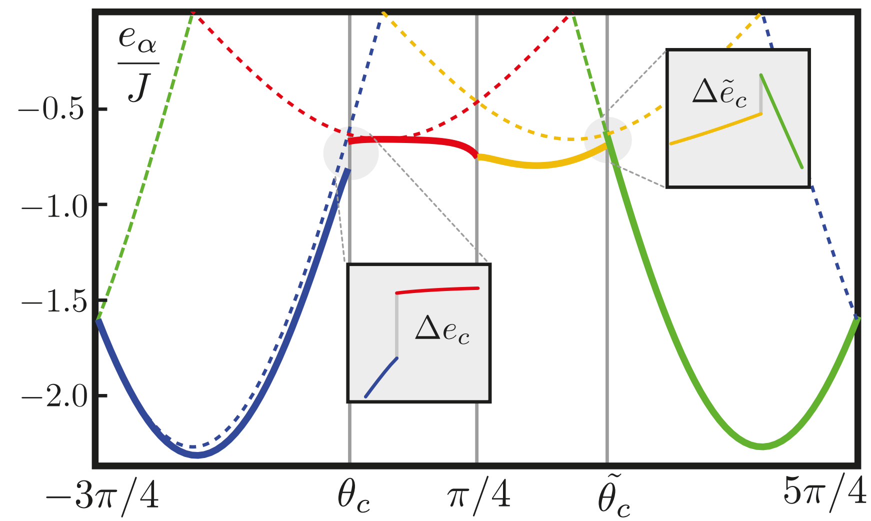
Ferromagnetic order parameter at $\theta = -\pi/2$

$$\langle \sigma_x^i \rangle - 1 \equiv \Delta m \approx 0.08$$

Corrections to mean-field ground state energy:

- the zero point fluctuations of the spin wave provide a substantial contribution at the anti-ferromagnetic Heisenberg point

- predict first order phase transition between ferro and anti-ferromagnetic phases



Conclusion

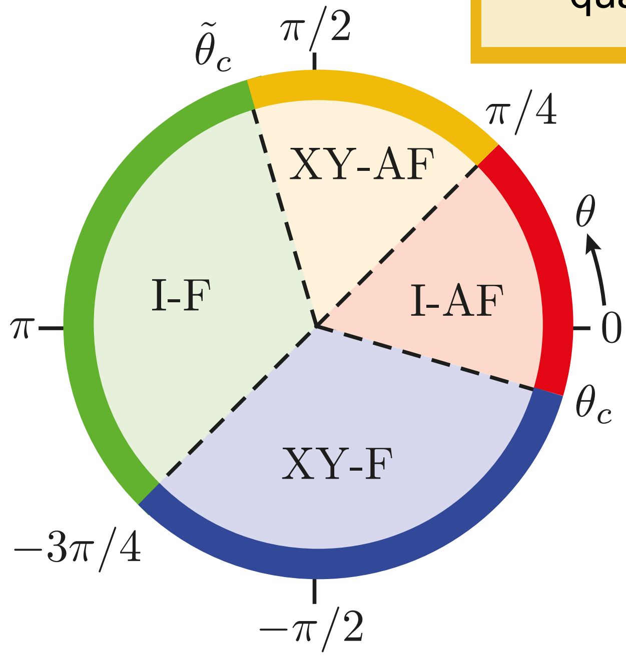
linear Goldstone mode

Kosterlitz-Thouless transition

quasi-long range order

gaped linear spectrum

algebraic correlations



gaped linear spectrum

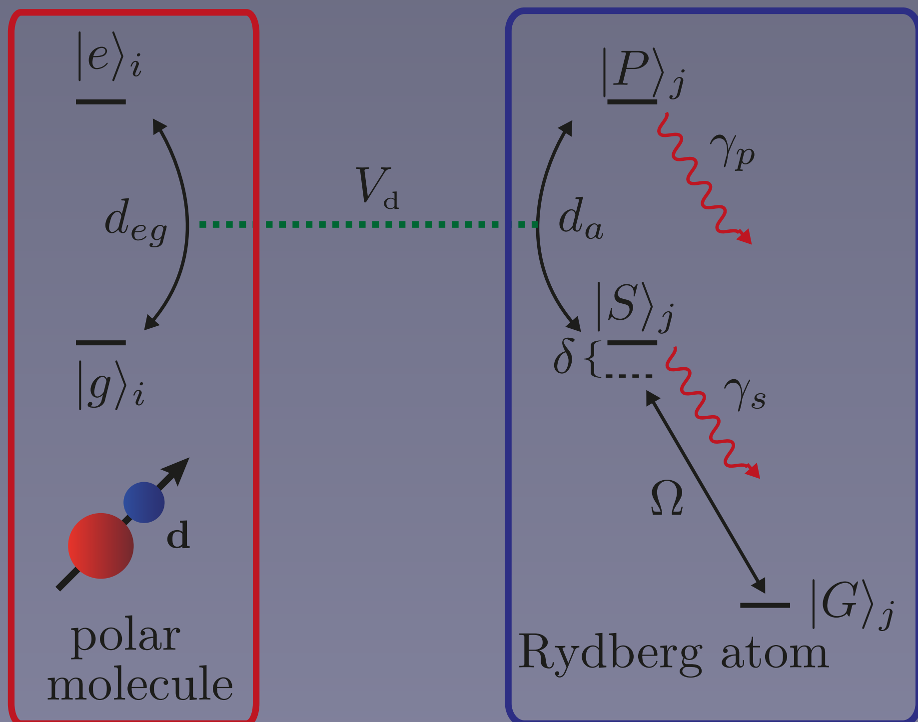
algebraic correlations

anomalous spectrum $\sim \sqrt{|\mathbf{q}|}$

broken U(1) symmetry in 2D

ferromagnetic order at low temperatures

Cooling polar molecules
to
ultra-cold temperatures?



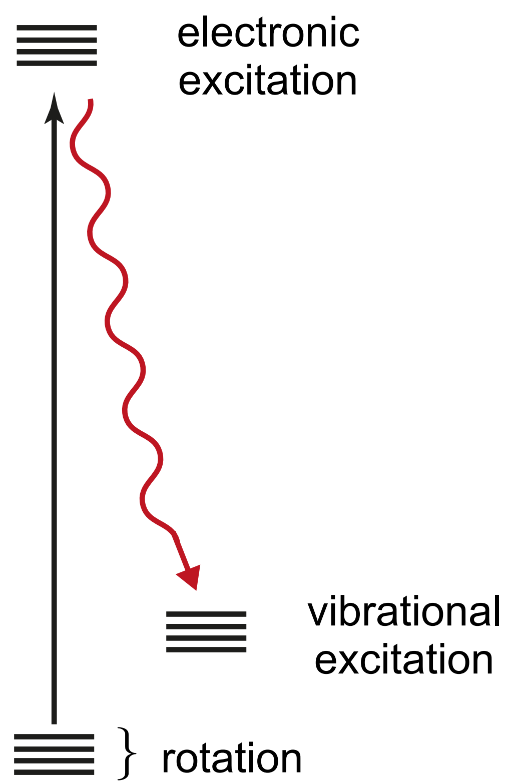
Spontaneous emission

Spontaneous emission of electronic states in polar molecules:

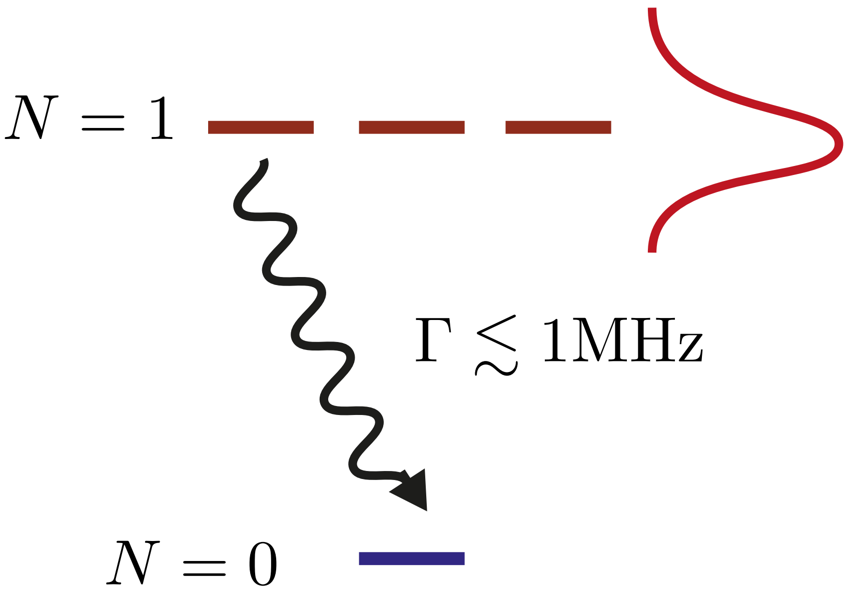
- transition into different rotational and vibrational states

- internal heating of the polar molecule

- absence of cycling transition



Decay rate for rotational state



Design a fast decay rate for an excited rotational state

- decay rate in the range of MHz

- turning the strength externally

- small momentum recoil

Optical pumping

- internal cooling of rotational states

- preparation of a single hyperfine state

→ fundamental requirement for quantum information applications

Laser cooling

- conventional laser cooling techniques
  - absence of momentum recoil
  - tunable decay rate

→ laser cooling into strongly correlated many-body state?

Setup

Hybrid system:
Rydberg atoms and polar molecules

- polar molecules interacting with neutral atoms excited into a Rydberg state

- very strong dipole-dipole interactions

- non-overlapping samples: atoms and molecules are well separated in space

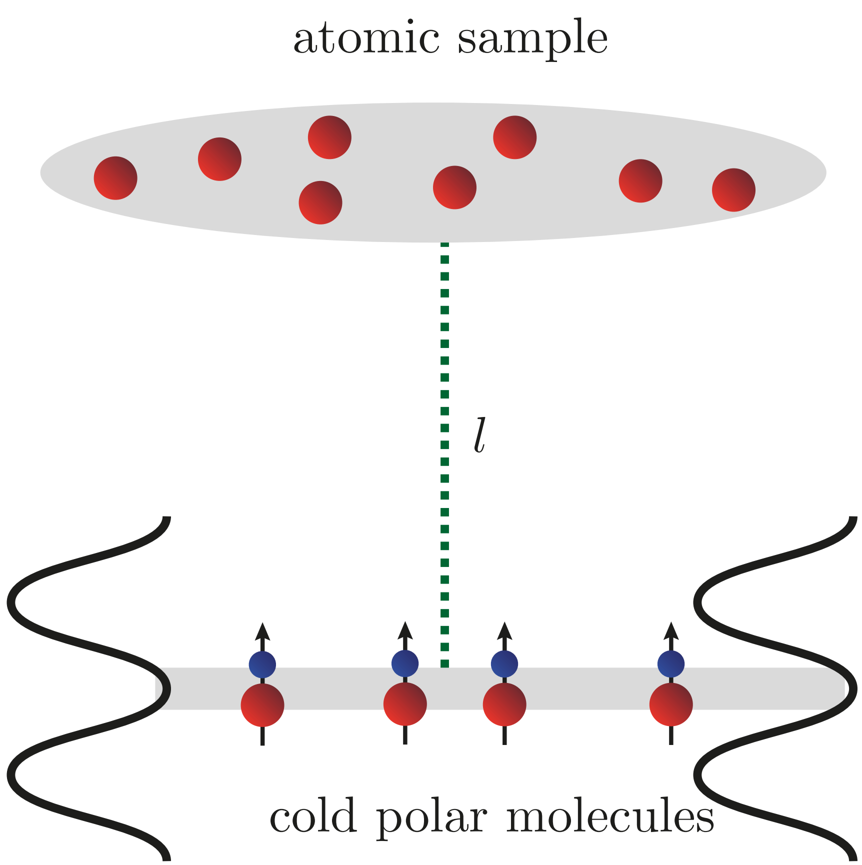
➡

- absence of chemical reactions
- suppressed inelastic collisions
- absence of equilibration between atoms and molecules

- laser cooling of atomic system

- resonant dipole-dipole interactions

- fast decay rate for Rydberg atoms



Setup

Dipole-Dipole interaction

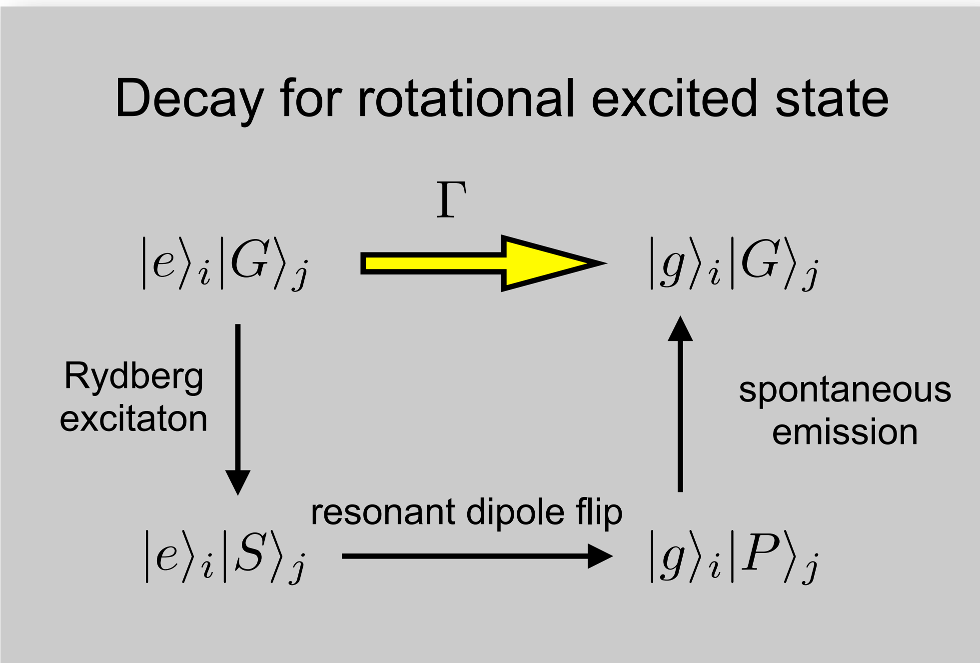
- resonant exchange interaction

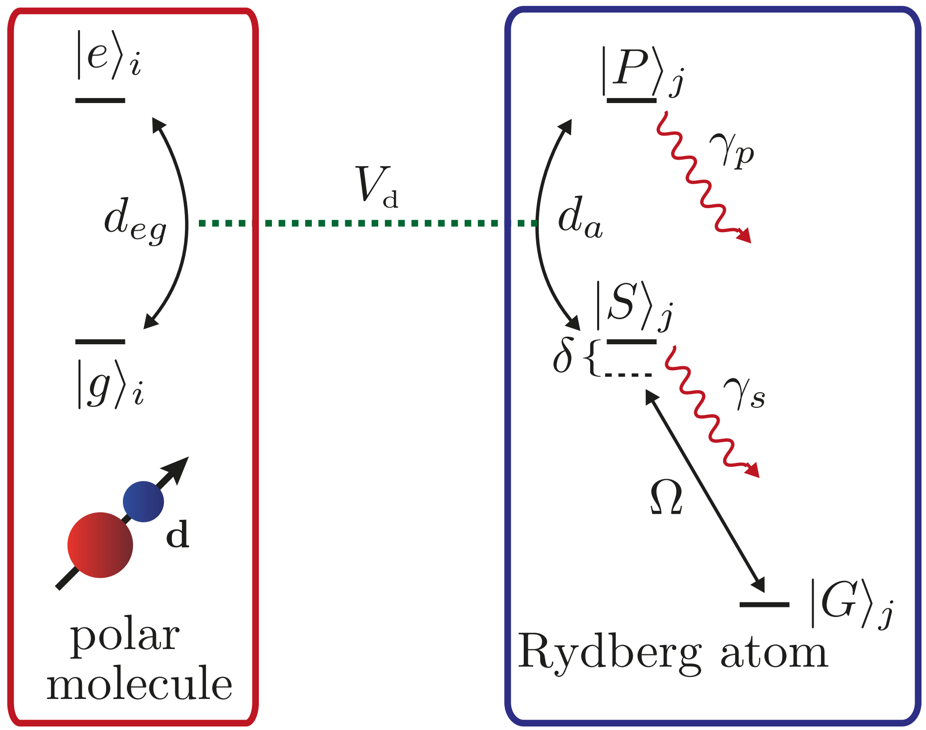
rising/lowering operators for the polar molecule $S_i^- = |g\rangle\langle e|_i$

$$H_c = \sum_{i,j} V_d(\mathbf{r}_i - \mathbf{R}_j)\left[S_i^- T_j^+ + S_i^+ T_j^-\right]$$

rising/lowering operators for Rydberg state $T_j^+ = |P\rangle\langle S|_j$

dipole-dipole interaction





Conclusion

Rotational states

- coupling of different rotational states
- decay of a single rotational state towards the ground state

Laser cooling

- Raman transition into rotational state
- Doppler limit in the quantum degenerate regime

Hyperfine states

- quadrupole coupling between nuclear and rotational states for N=1
- coupling from N=0 to N=1 via microwave field
- select a dark state due to selection rules
  - polarization
  - energy