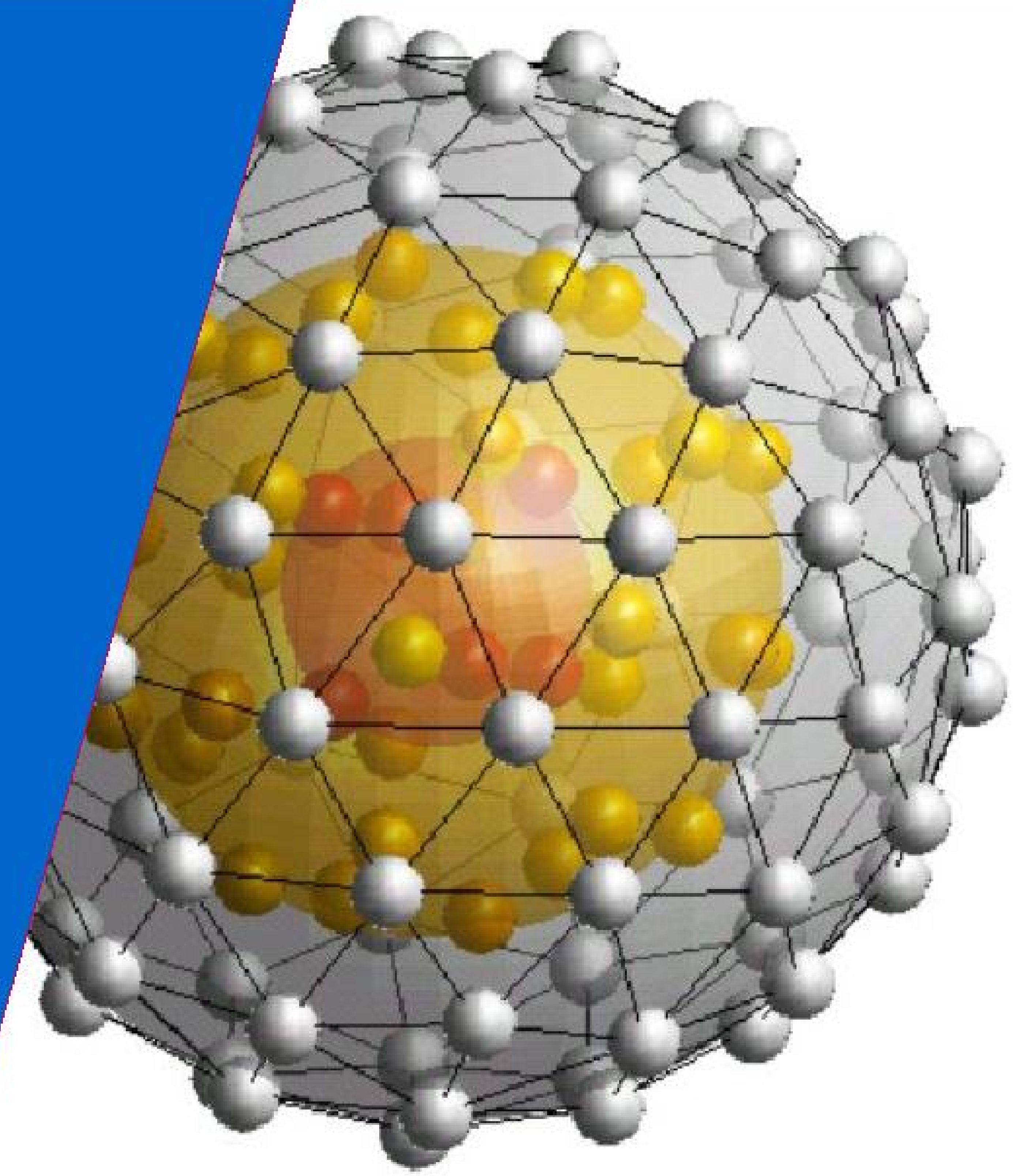
At the boundary of short-range and long-range potentials: molecules and Rydberg atoms with strong interactions

12-2-13, KITP, Santa Barbara

Servaas Kokkelmans



Technische Universiteit
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University of Technology

Where innovation starts

Outline

- **Asymtotic Bound state Model**
  - **Short range: van der Waals length** $r_{vdW} = \frac{1}{2}\left(\frac{2\mu C_6}{\hbar^2}\right)^{1/4}$
  - **Long-range:** $r > r_{vdW}$
  - **Asymptotic region weakly-bound molecules:**

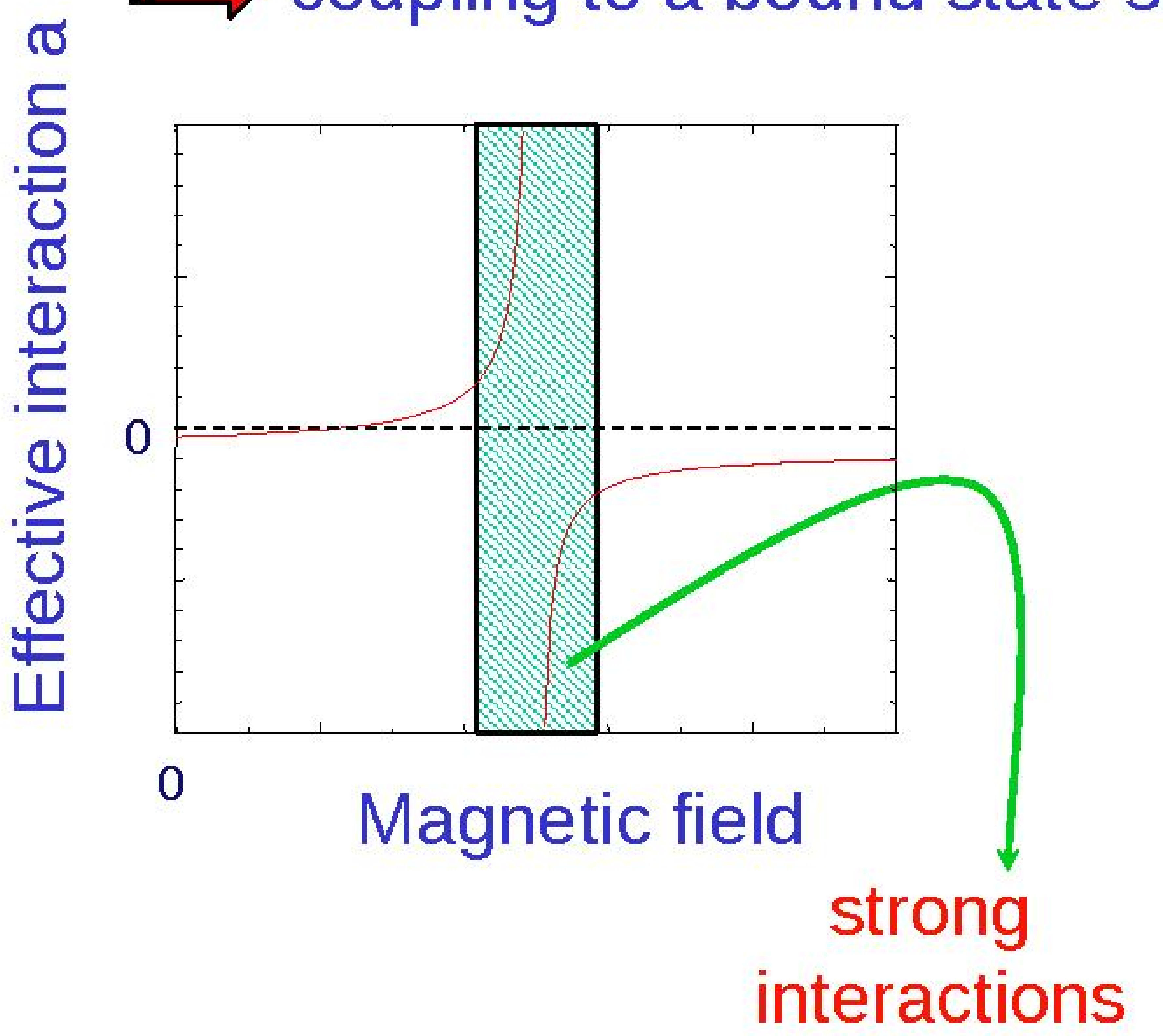
$$\psi(r) = \frac{1}{\sqrt{2\pi a}} \frac{e^{-r/a}}{r}$$ with $a > r_{vdW}$

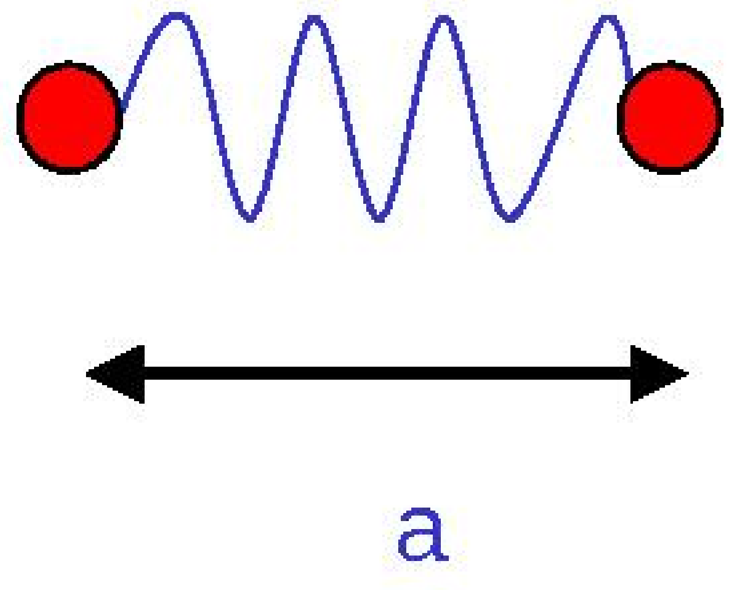
  - **Dipolar interactions**
  - **Feshbach spectroscopy**
- **Rydberg crystals**
  - **Highly-excited electronic s-states:** $r_{vdW} > n^{-1/3}$
  - **Long-range interactions**
  - **Blockade effect**

Feshbach resonances: Strong interactions

External field: Tuning atom-atom interaction: Feshbach resonance

➡ coupling to a bound state state





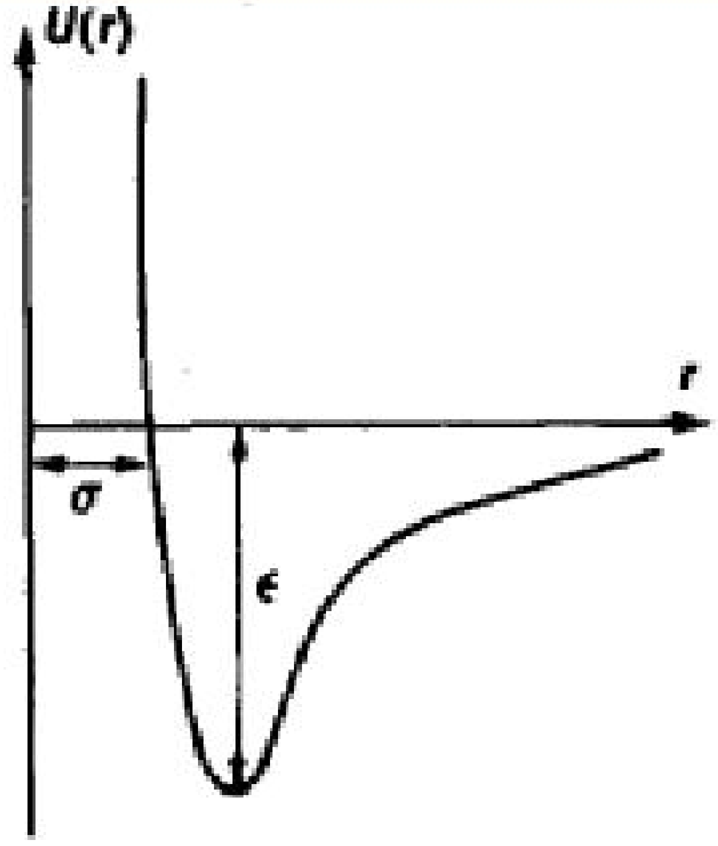
Weakly-bound molecules

Interaction parameter is the scattering length *a*

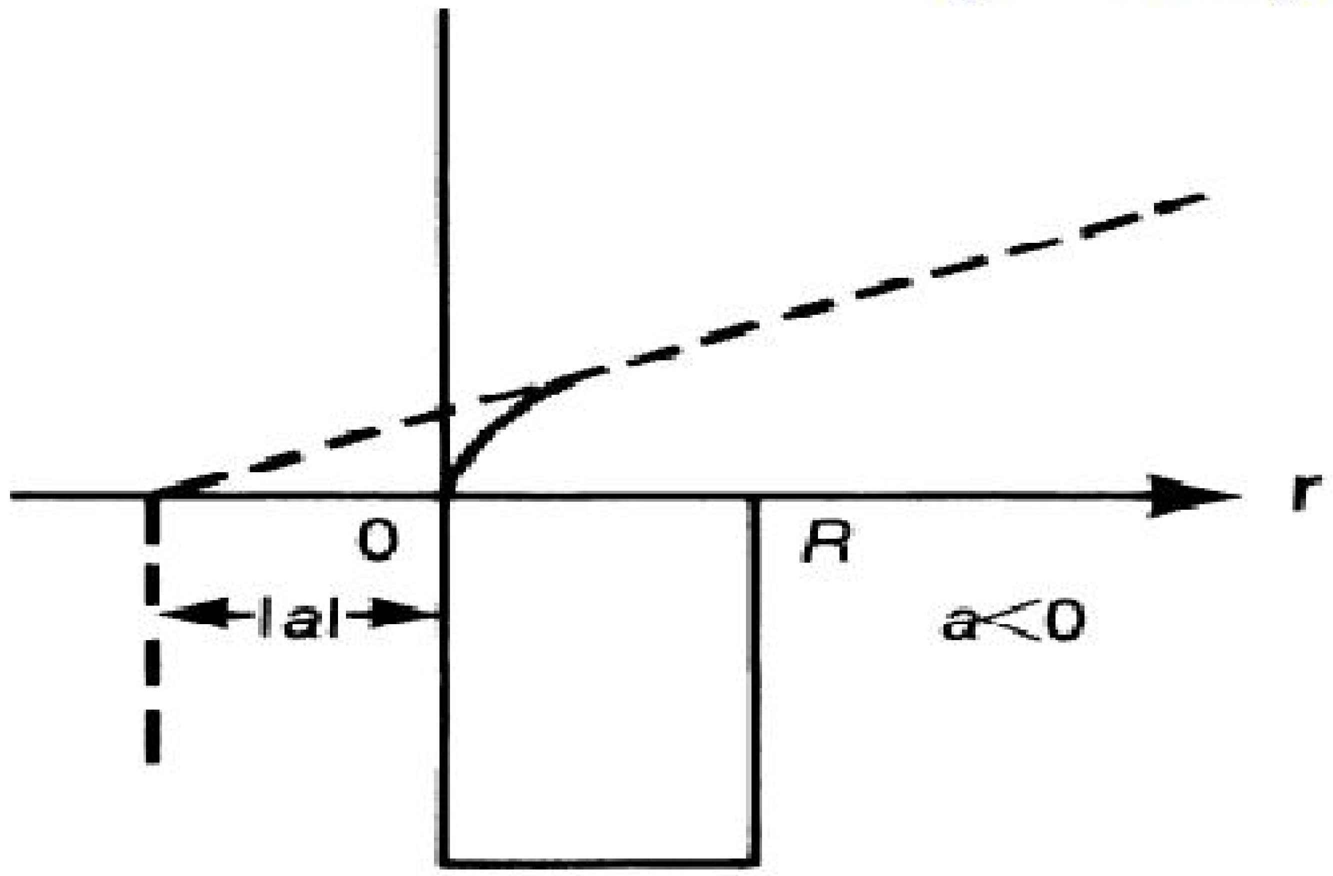
Short-range potentials: no spin

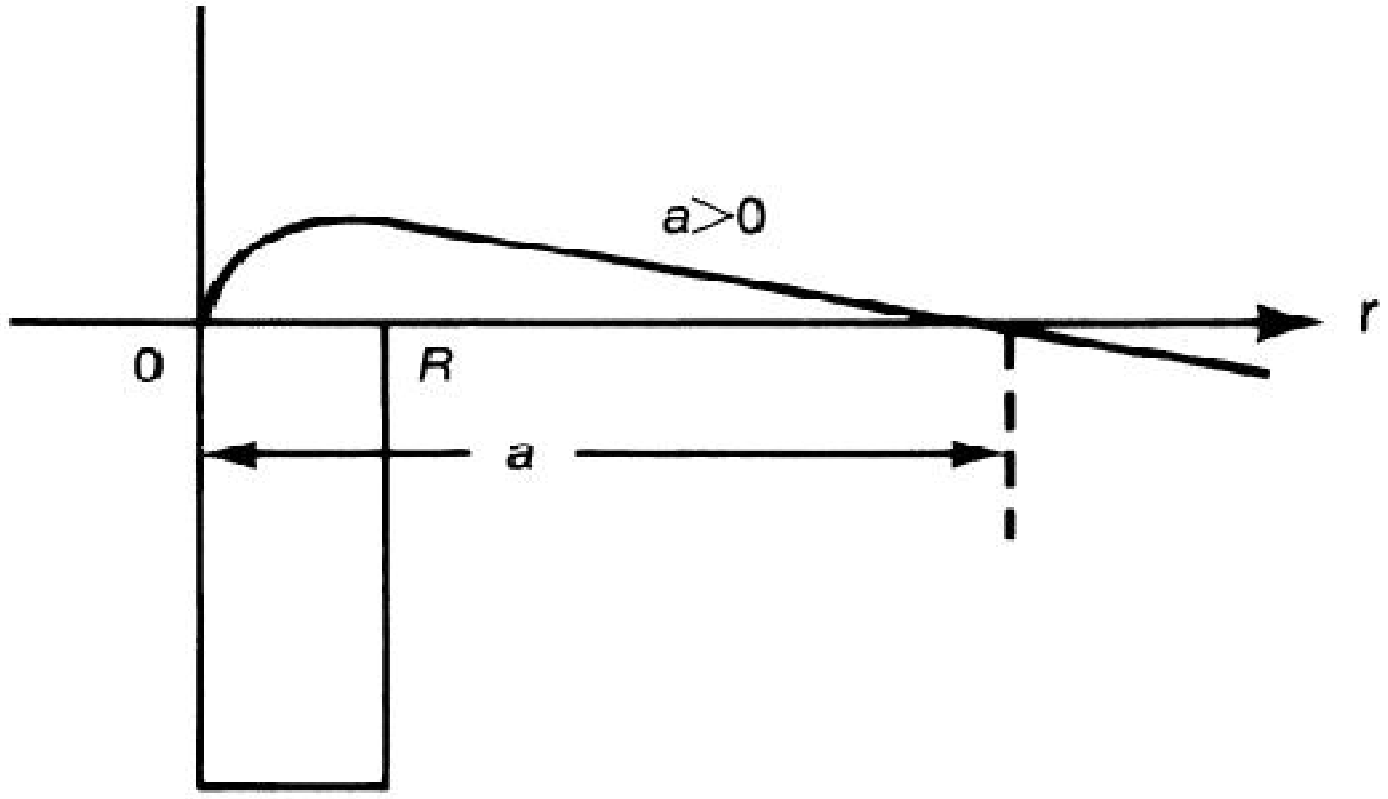
- **Short range two-body interactions**

  ➡ **s-wave scattering length *a***



$$\psi = \sin[k(r-a)]$$





$$a \rightarrow \pm\infty$$

inbetween

Two-body interactions including spin

Hamiltonian of two colliding particles:

$$H=\frac{p^2}{2\mu}+\sum_{i=1}^{2} H_i^{intern}+V^{centr}+V^{dip}$$

Hyperfine and Zeeman interaction

$$H^{intern}=V^{hf}+V^{z}$$

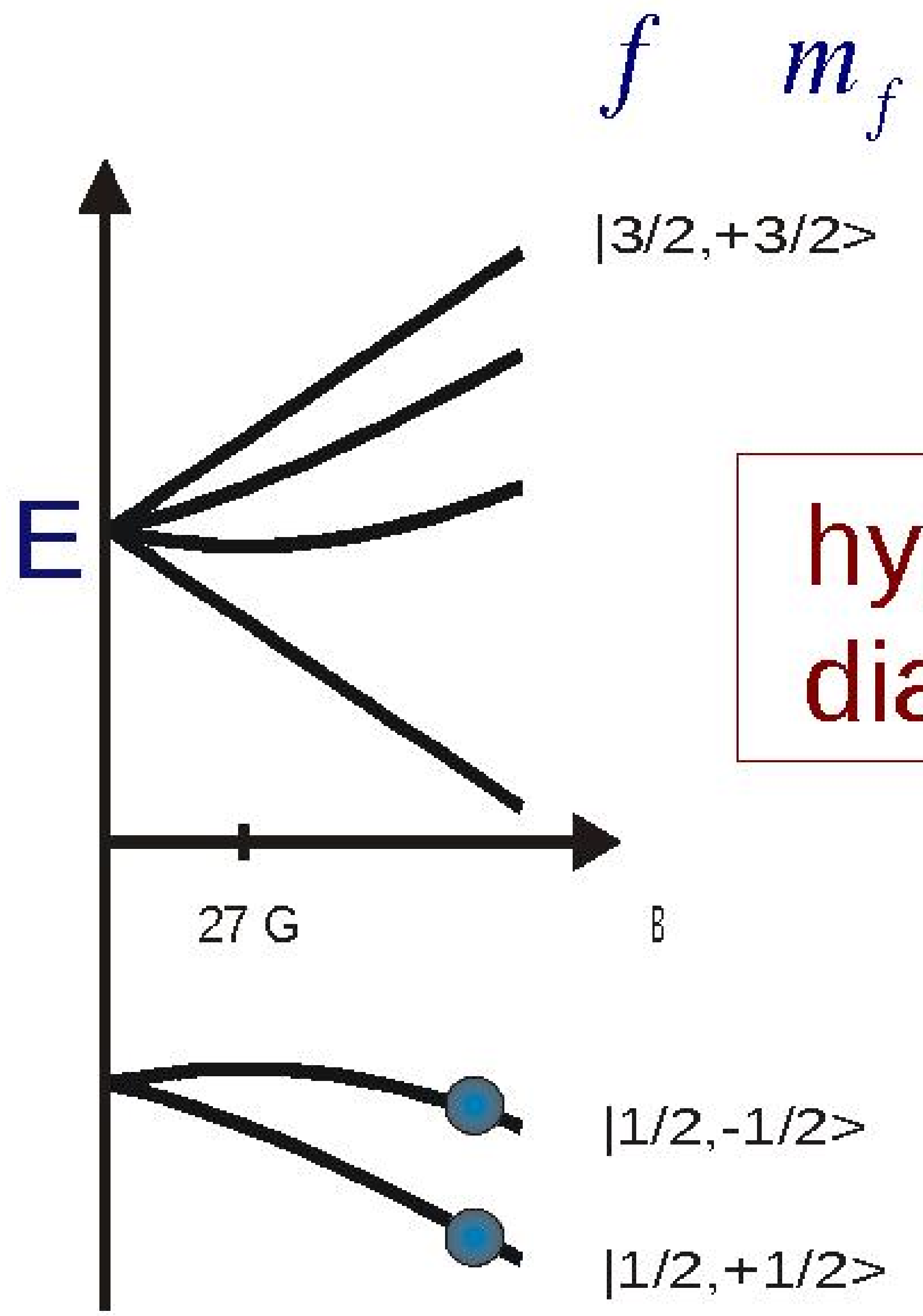
$$=\frac{a_{hf}}{\hbar^2}\vec{s}\,.\,\vec{i}+\frac{1}{\hbar}(\gamma_e s_z-\gamma_N i_z)B_z$$

Total single-particle spin:

Two particle “hyperfine” basis:

$$\vec{f}=\vec{s}+\vec{i}\;;m_f=m_s+m_i$$

$$\left|f_1,m_{f_1};f_2,m_{f_2}\right\rangle$$



hyperfine diagram

Two-body interactions including spin (2)

Hamiltonian of two colliding particles:

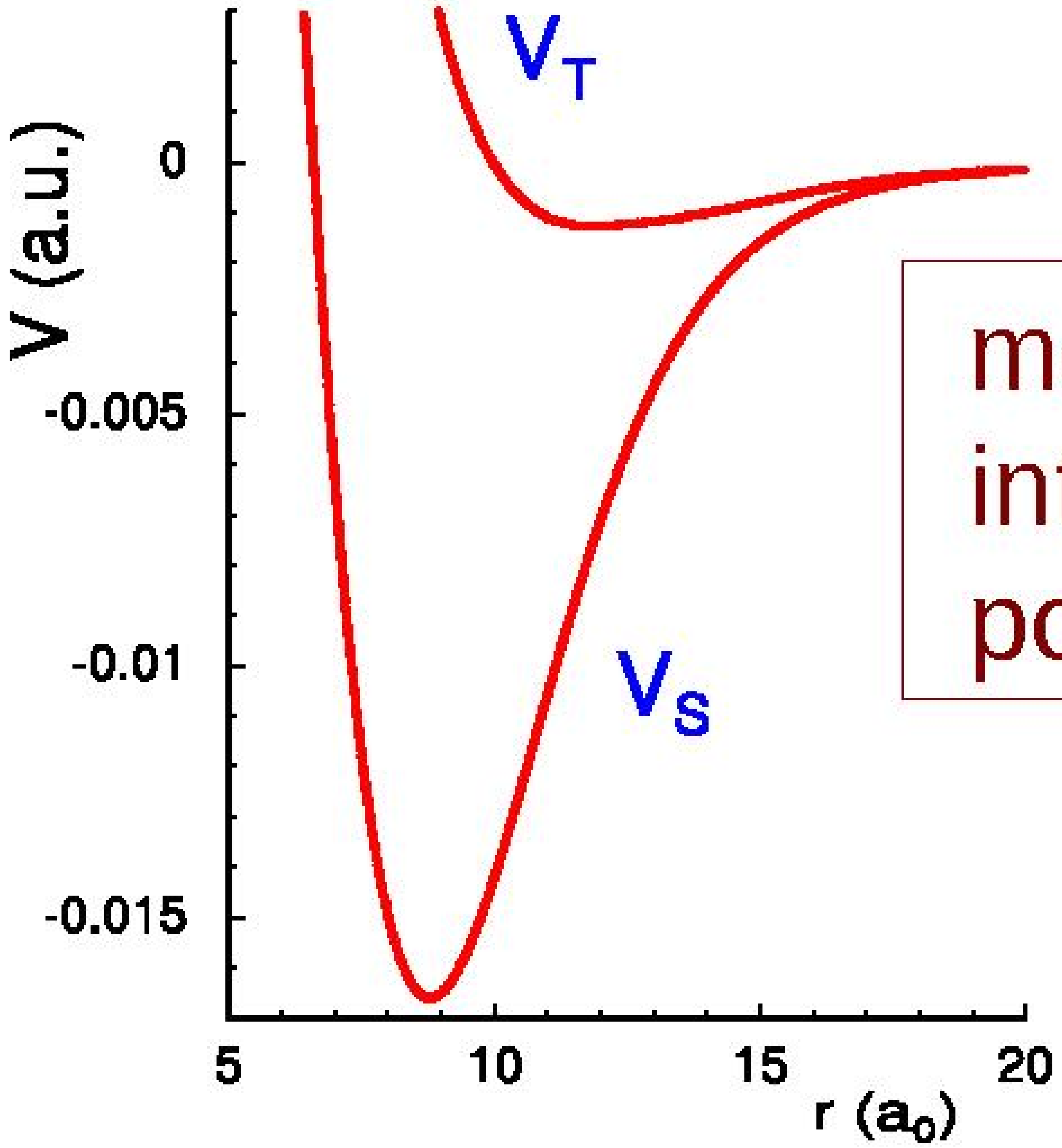
$$H=\frac{p^2}{2\mu}+\sum_{i=1}^{2} H_i^{intern}+V^{centr}+V^{dip}$$

Central interaction

$$V^{centr}=V^S P_S+V^T P_T$$

(All coulomb interactions)

Electronic ground state
Singlet and Triplet
potentials
(dep. on electron spin)



molecular interaction potentials

Total electron/nuclear spin: $\vec{S}=\vec{s}_1+\vec{s}_2 ; \vec{I}=\vec{i}_1+\vec{i}_2$

Two particle "singlet/triplet" basis: $|S, m_S ; I, m_I\rangle$

Coupled channels equation

These two bases are incompatible:

$$\left|f_1, m_{f_1}; f_2, m_{f_2}\right\rangle \quad \Longleftrightarrow \quad \left|S, m_S; I, m_I\right\rangle$$

Good basis for $r \rightarrow \infty$

Good basis for $r \rightarrow 0$

Gives rise to coupling between different channels:

with coupling matrix

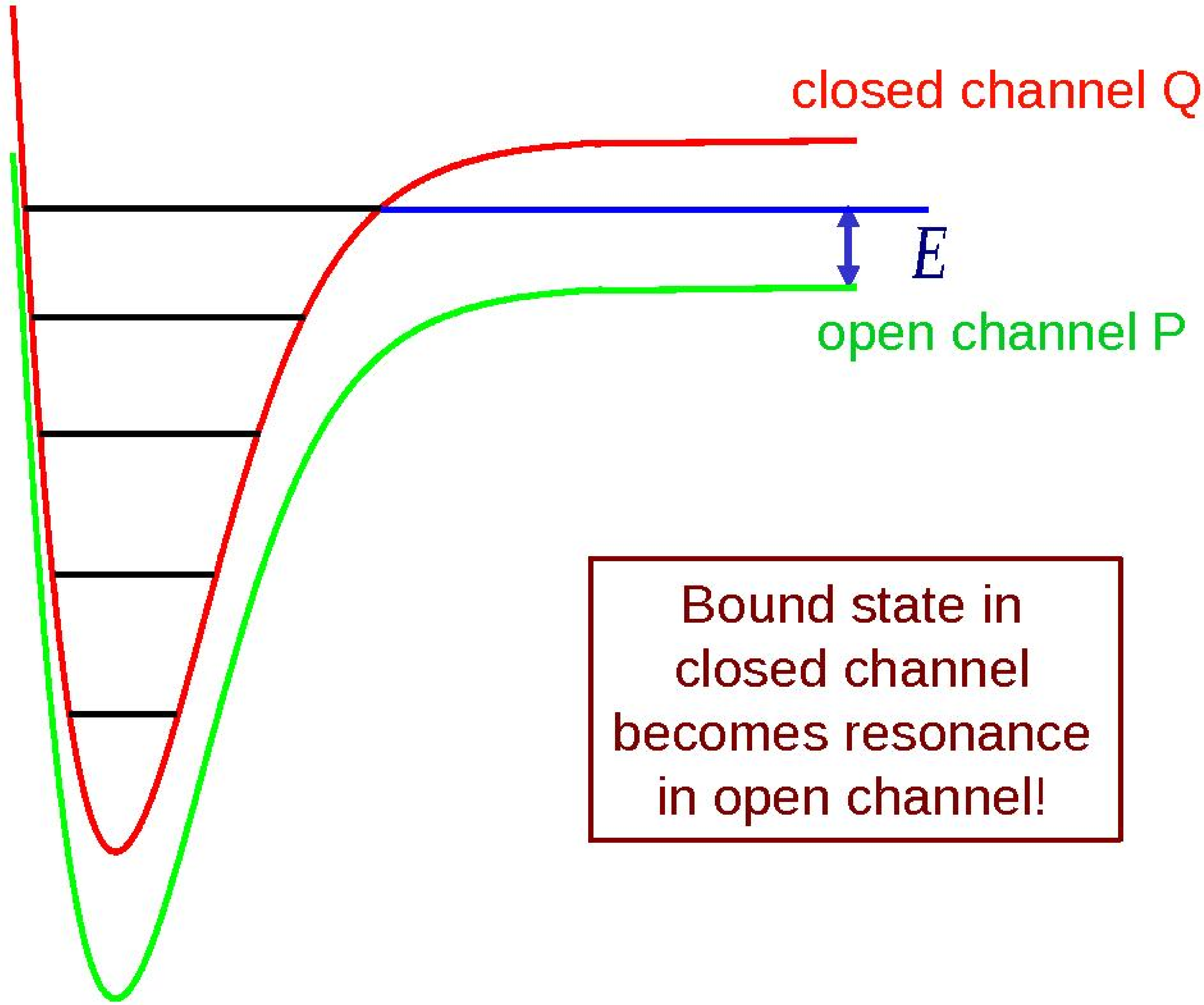
$$V_{lm\{\alpha\beta\}}^{l'm'\{\alpha'\beta'\}} = i^{l'-l} \left\langle lm\{\alpha\beta\}\right| V^{centr} \left| l'm'\{\alpha'\beta'\}\right\rangle$$

Cold collisions: Only few *l* needed (*l*=0,1,2)

Conservation of m_{tot}

Open and closed channels in hyperfine basis

Two-body spin states are coupled: coupled channels

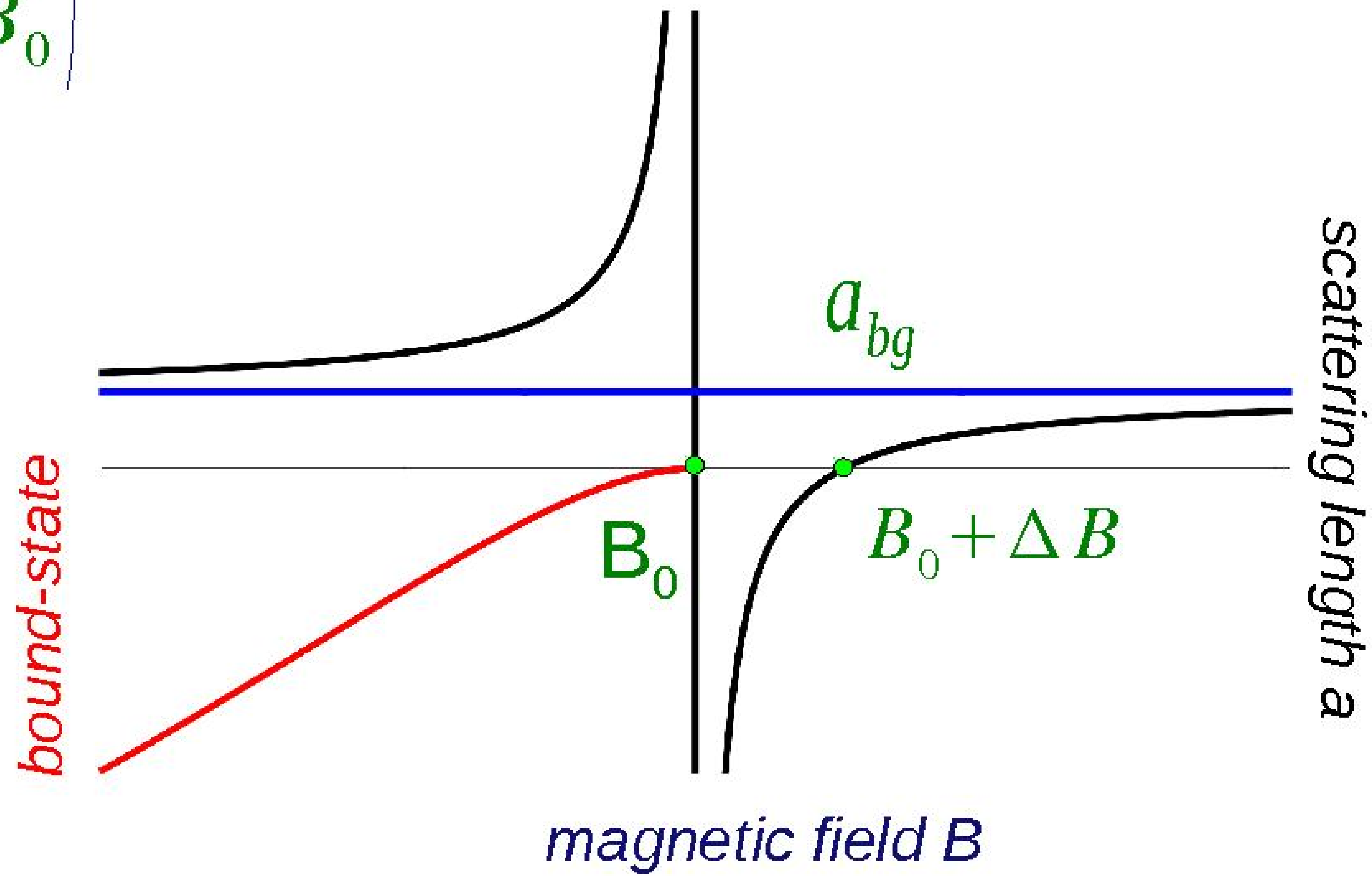


Bound state in
closed channel
becomes resonance
in open channel!

Feshbach resonance

- **Control over scattering length with magnetic field**

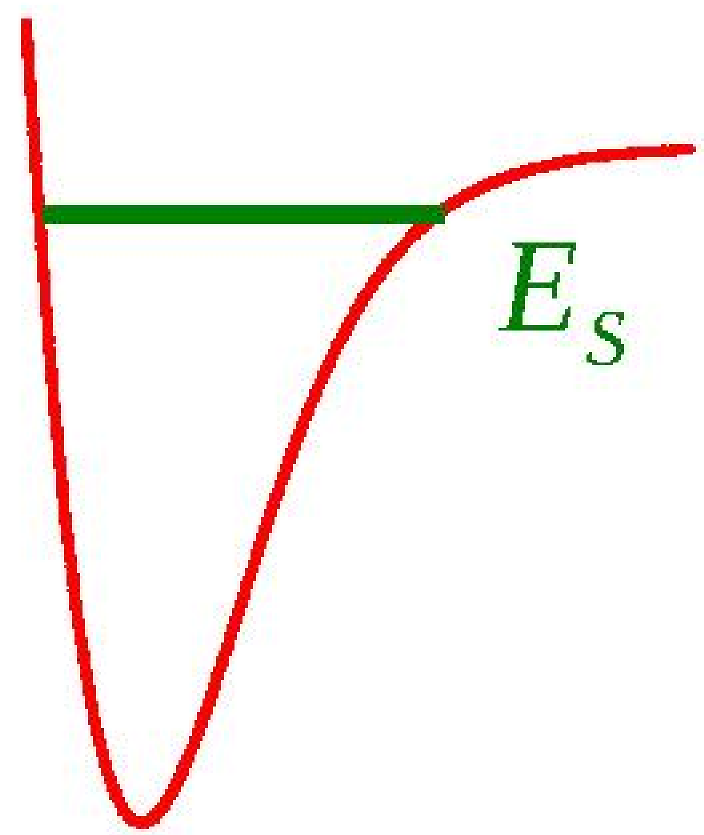
$$a = a_{bg}\left(1 - \frac{\Delta B}{B - B_0}\right)$$



Asymptotic bound-state model

For the prediction and analysis of Feshbach resonances

- **Simple model: based on highest bound state**



Replace whole potential
by only a number

$$E_S = \frac{-\hbar^2 \kappa_S^2}{m}$$

$$\psi(r) = \sqrt{\frac{\kappa_S}{2\pi}} \frac{e^{-\kappa_S r}}{r}$$

- **Find bound states of coupled system**

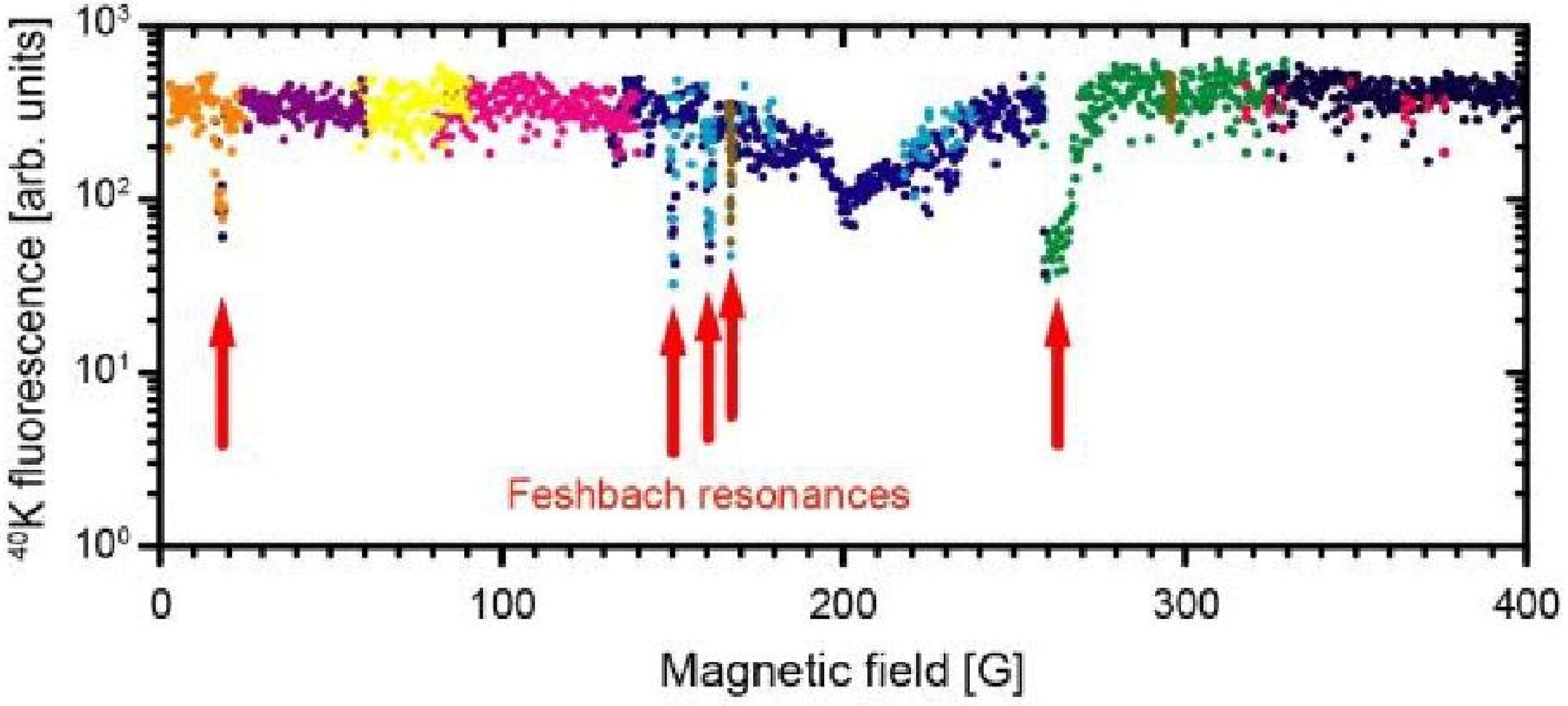
Feshbach spectroscopy

- ~~Complicated coupled radial equations~~

  → simple matrix diagonalization

  gives rise to coupled field-dependent dimer states

- Easy-to-use tool for analyzing Feshbach spectroscopic data: fingerprint of interatomic interactions



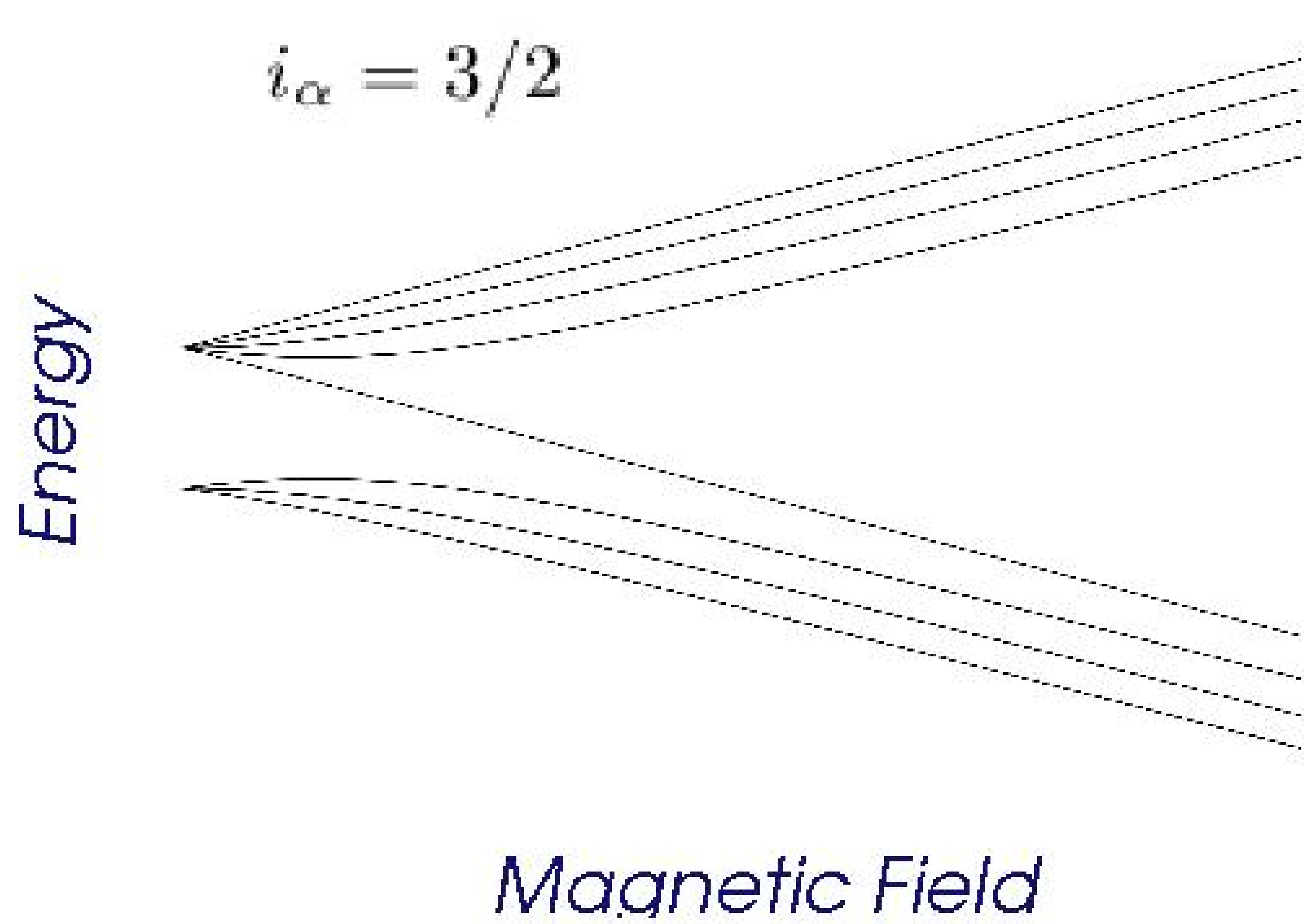
See e.g.: [T. G. Tiecke et al, Phys. Rev. Lett. 104, 053202 (2010)]

Internal Hamiltonian

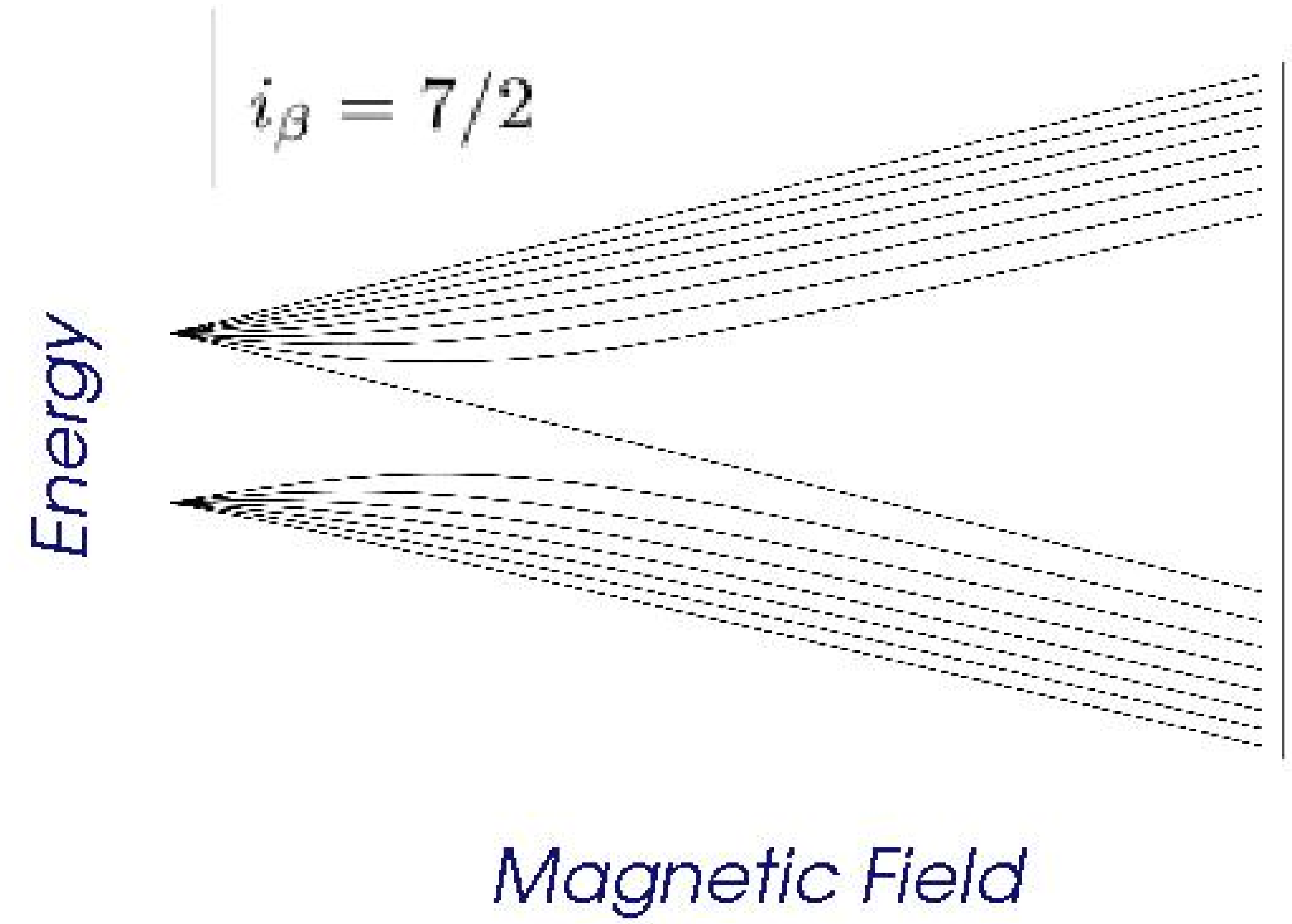
- **Hyperfine and Zeeman interaction**

$$H_{\mathrm{hf}} = \underbrace{\frac{a_{\mathrm{hf}}}{\hbar^2}\underline{s}\cdot\underline{i}}_{\text{couples spins } \underline{f} = \underline{s} + \underline{i}} + \underbrace{(\gamma_e \underline{s} - \gamma_n \underline{i})\cdot\underline{B}}_{\text{lifts degeneracy}}$$

couples spins $\underline{f} = \underline{s} + \underline{i}$ — lifts degeneracy



$|f_\alpha m_{f,\alpha}\rangle$



$|f_\beta m_{f,\beta}\rangle$

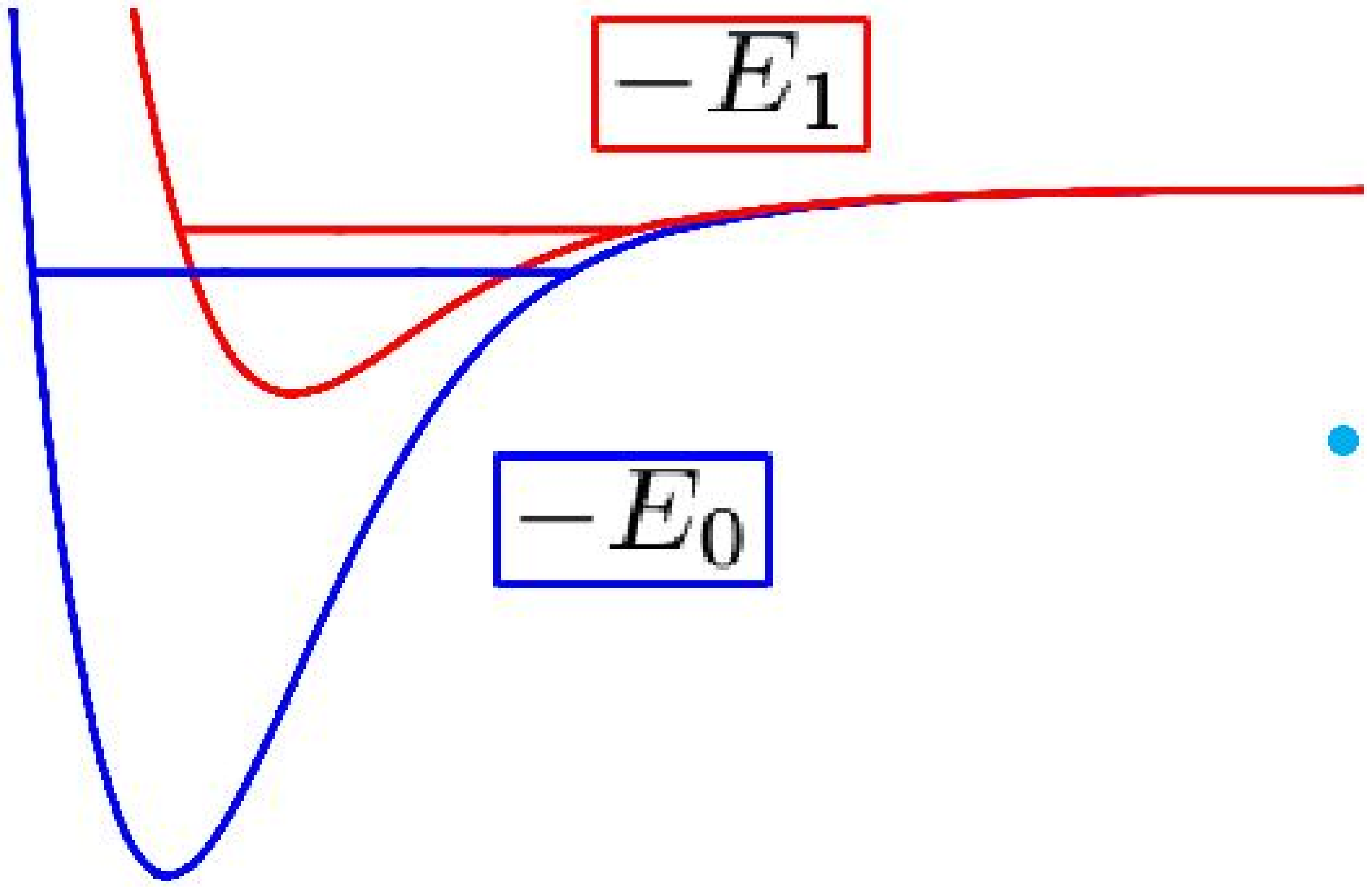
Relative Hamiltonian

- **Define the relative Hamiltonian**

$$H_{\mathrm{rel}} = \frac{p^2}{2\mu} + V$$

- **Define two (least bound) eigenstates**

$$H_{\mathrm{rel}}|\psi_S\rangle = -E_S|\psi_S\rangle$$



- **Eigenvalues are energies least bound states (model parameters)**

ABM: coupled bound states

- **Find coupled bound-states** $|\langle \Psi_{S'}|H - E|\Psi_S\rangle| = 0$
  - **ABM basis**

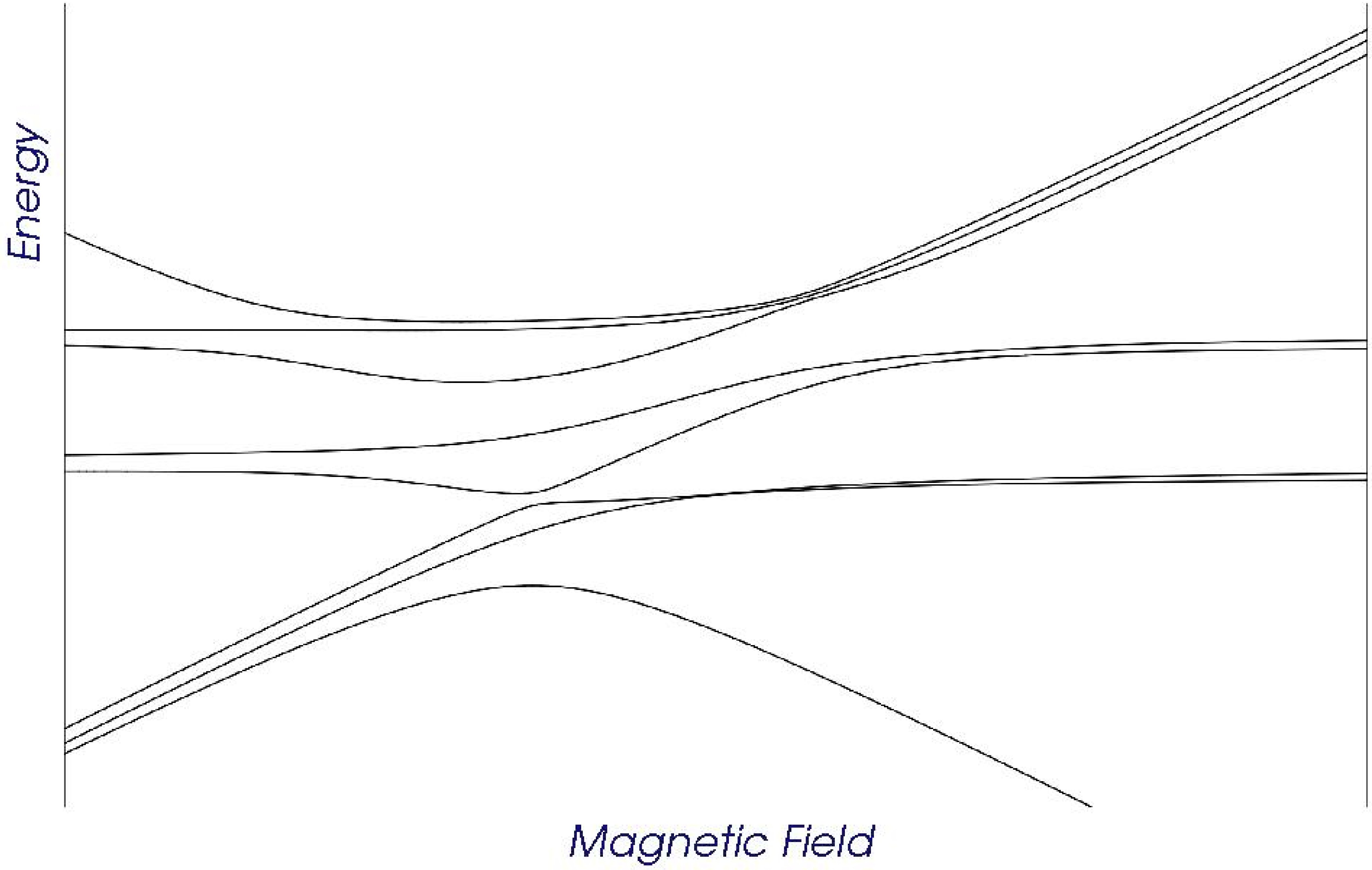
$$|\Psi_S\rangle \equiv |\psi_S\rangle \otimes |S m_S m_{i_\alpha} m_{i_\beta}\rangle$$

$$\Downarrow$$

$$H_{\mathrm{rel}} + \sum H_{\mathrm{hf}}$$

ABM: coupled bound states

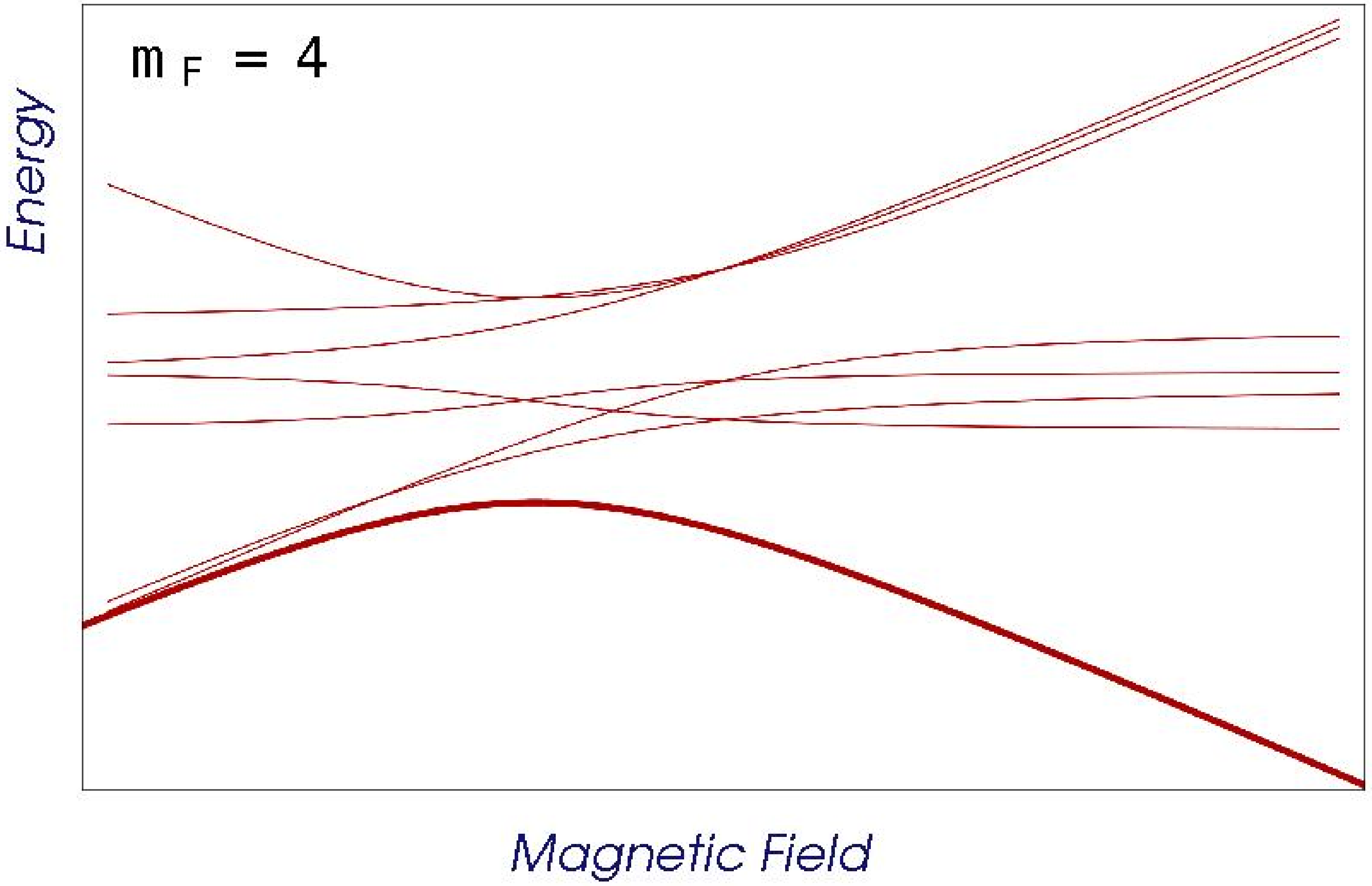
- **Find coupled bound-states** $|\langle\Psi_{S'}|H - E|\Psi_S\rangle| = 0$



ABM: open & closed channels

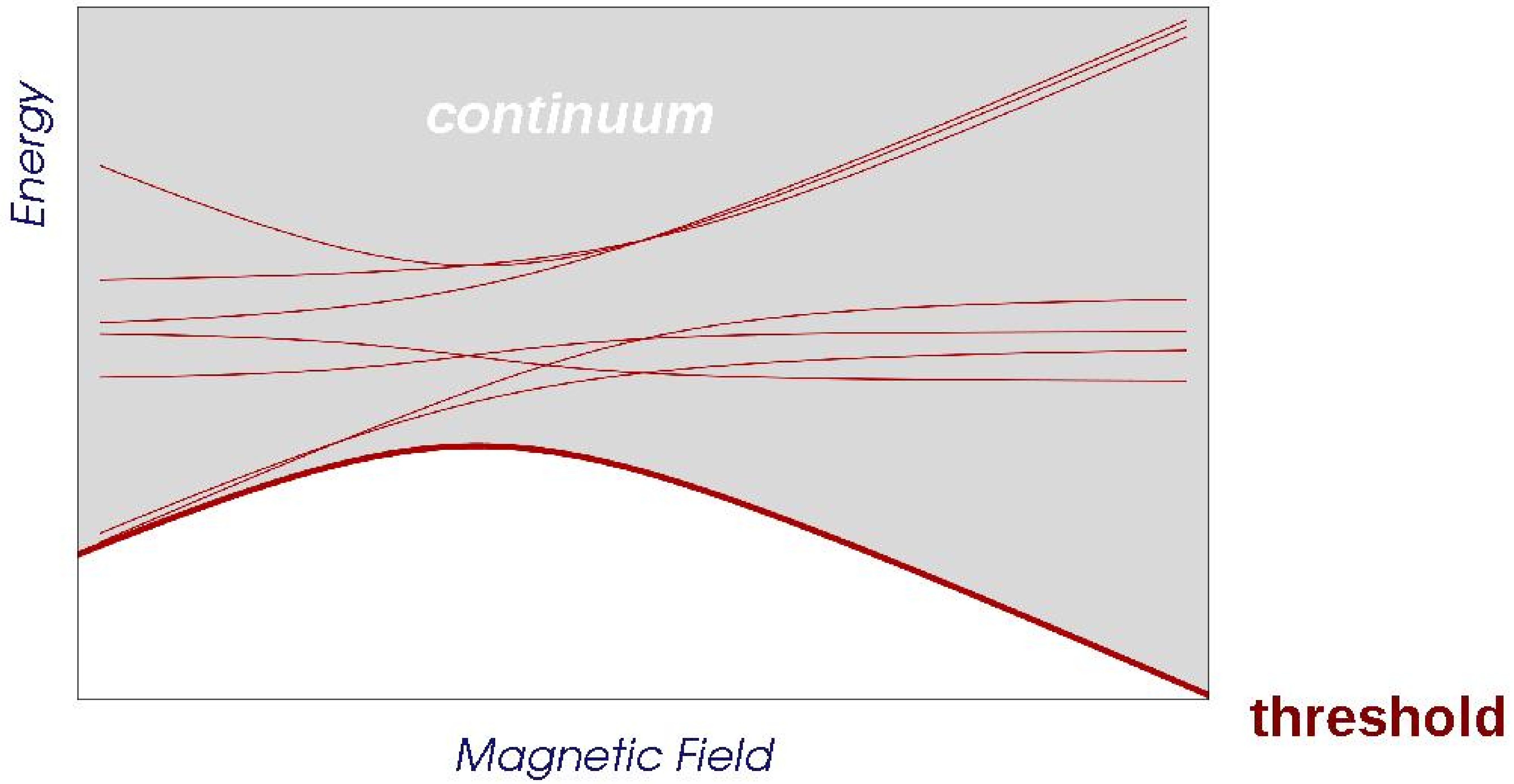
- **Prepare atoms in spin state: open channel**

$$\sum H_{\rm hf}$$

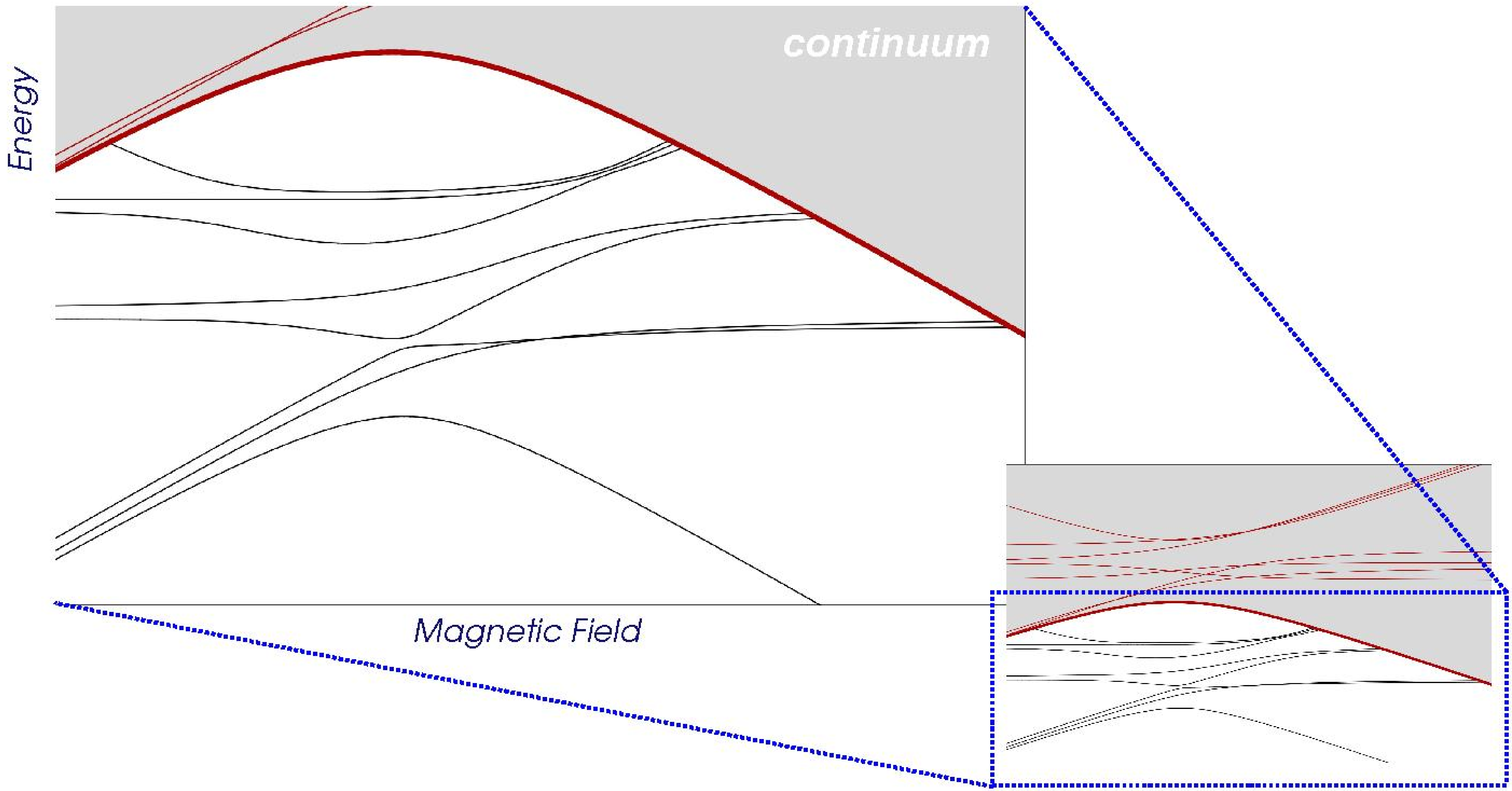


ABM: open & closed channels

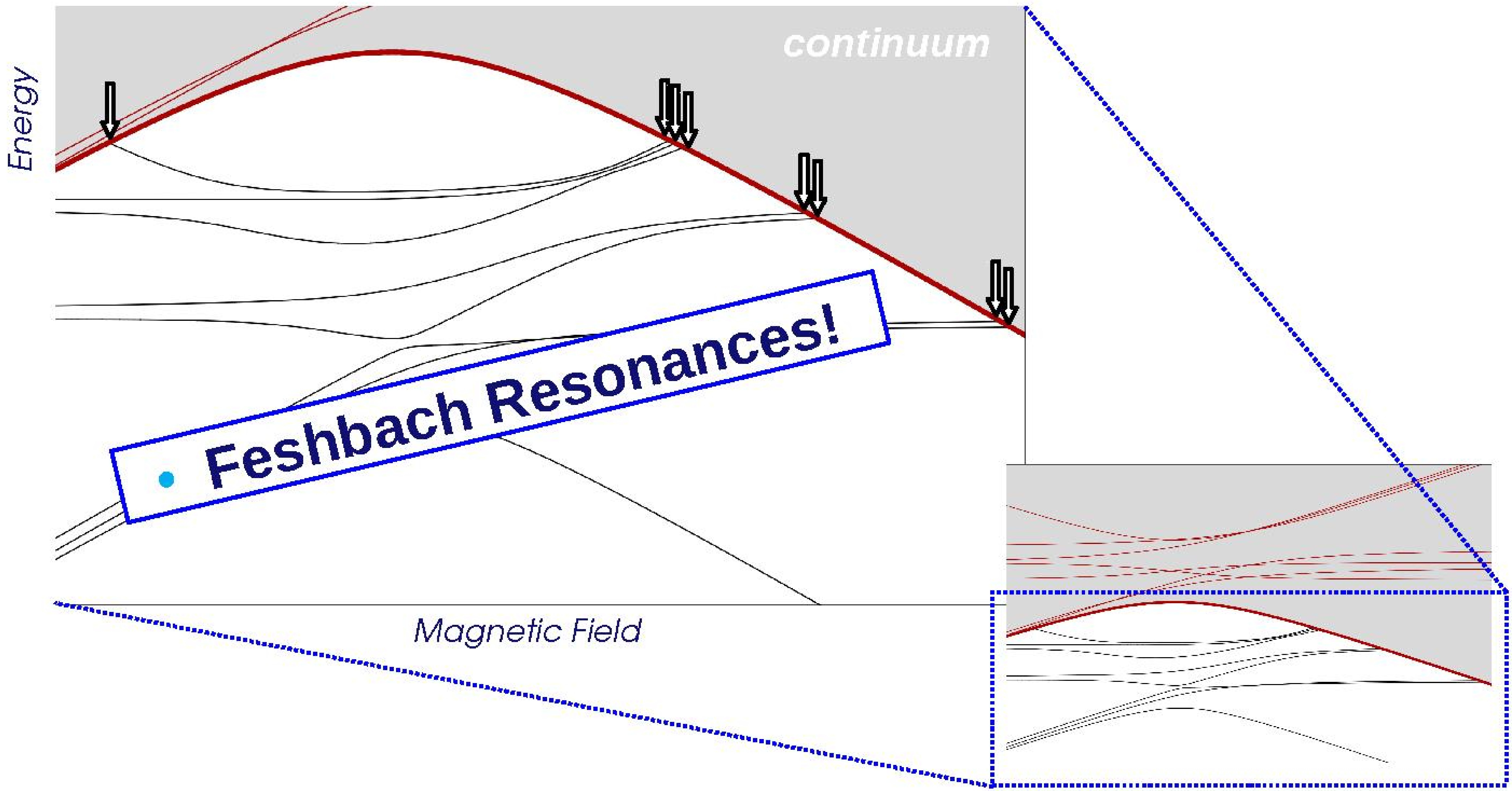
- **Prepare atoms in spin state: open channel**



ABM: predicting Feshbach resonances

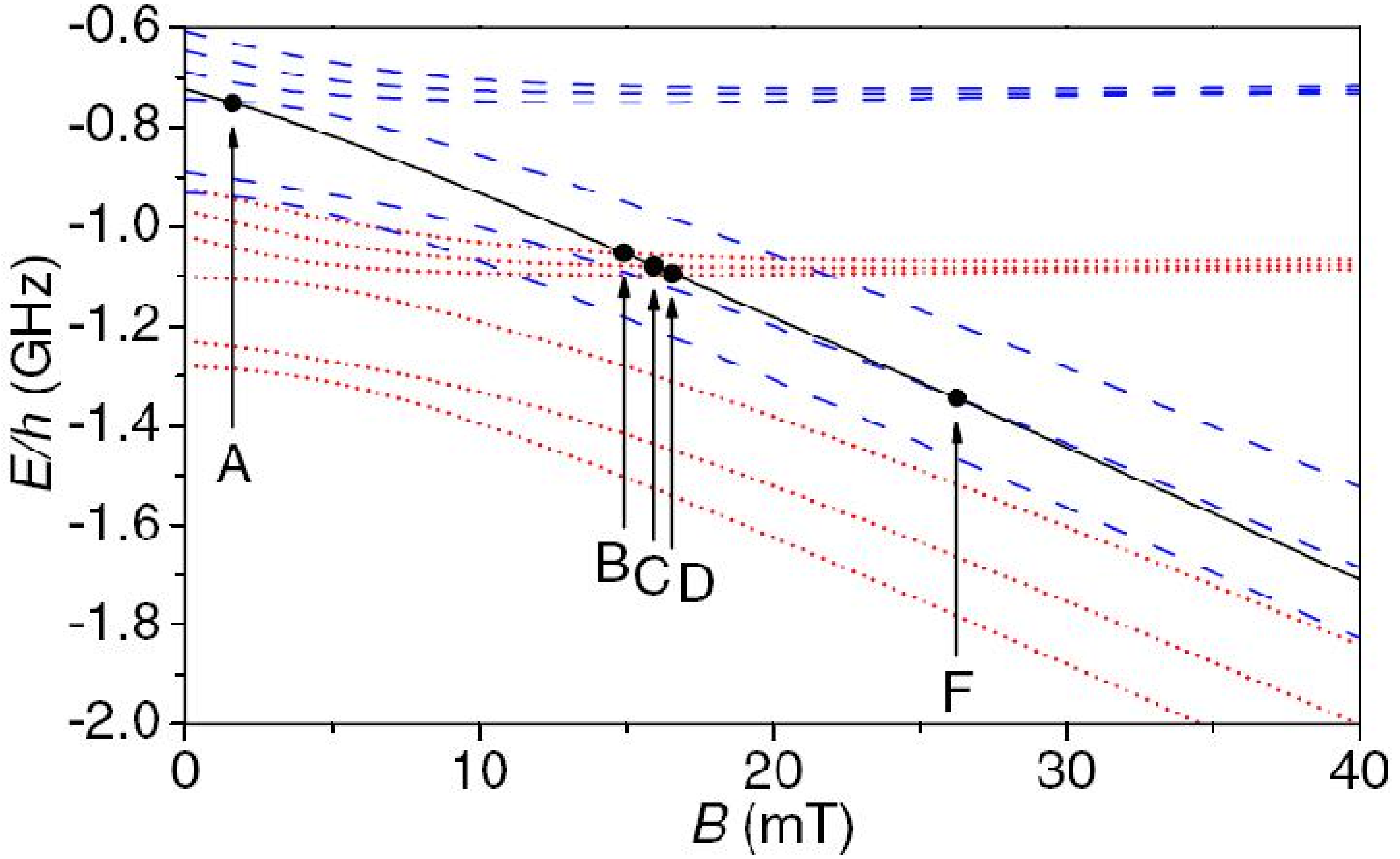


ABM: predicting Feshbach resonances



ABM: applied to Li-K

- **ABM identified s- and p-wave resonances**

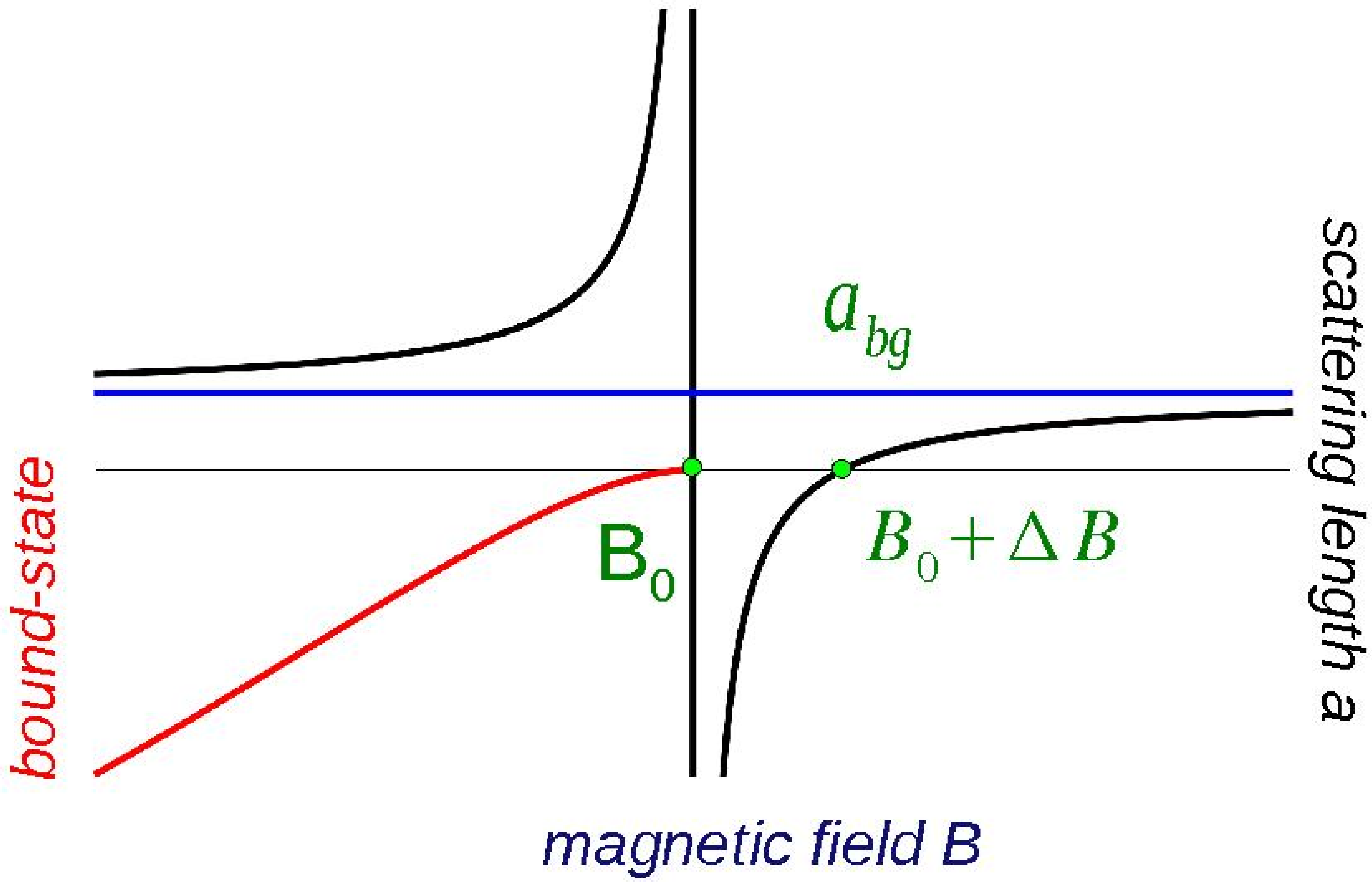


ABM: $E_0/\hbar = 716, E_1/\hbar = 425\,MHz$

Coupled Channels: $E_0/\hbar = 721, E_1/\hbar = 426\,MHz$

[E. Wille, *et al*. PRL 100, 053201 (2008)]

Threshold effects



Bound-state

$$E_{\rm bs} + \Delta\mu(B - B_0) + C\sqrt{E_{\rm bs}} = 0 \qquad C = a_{\rm bg}\Delta B\Delta\mu$$

- **Coupling open & closed channels**
  - **bending bound state, width of the resonance**

$$C = a_{\rm bg}\Delta B\Delta\mu$$

Width of resonances

Transform ABM to hyperfine basis

$$\begin{pmatrix} -E_0 & V_{\rm hf} \\ V_{\rm hf} & -E_1 + H_Z \end{pmatrix} \Rightarrow \begin{pmatrix} H_{QQ} & H_{QP} \\ H_{PQ} & H_{PP} \end{pmatrix}$$

bound-state energy

$$H_{QQ} = \epsilon_Q$$

$$H_{PP} = \epsilon_P$$

threshold energy

$$H_{PQ} = \langle \Psi_P | H | \Psi_Q \rangle = \sqrt{A} = \sqrt{\frac{C}{2\kappa_P |\epsilon_P|}}$$

Coupling P&Q

Solve for bound state energy*

$$(k - i\kappa_P)(E - \epsilon_Q - \Delta + iCk) = 0$$

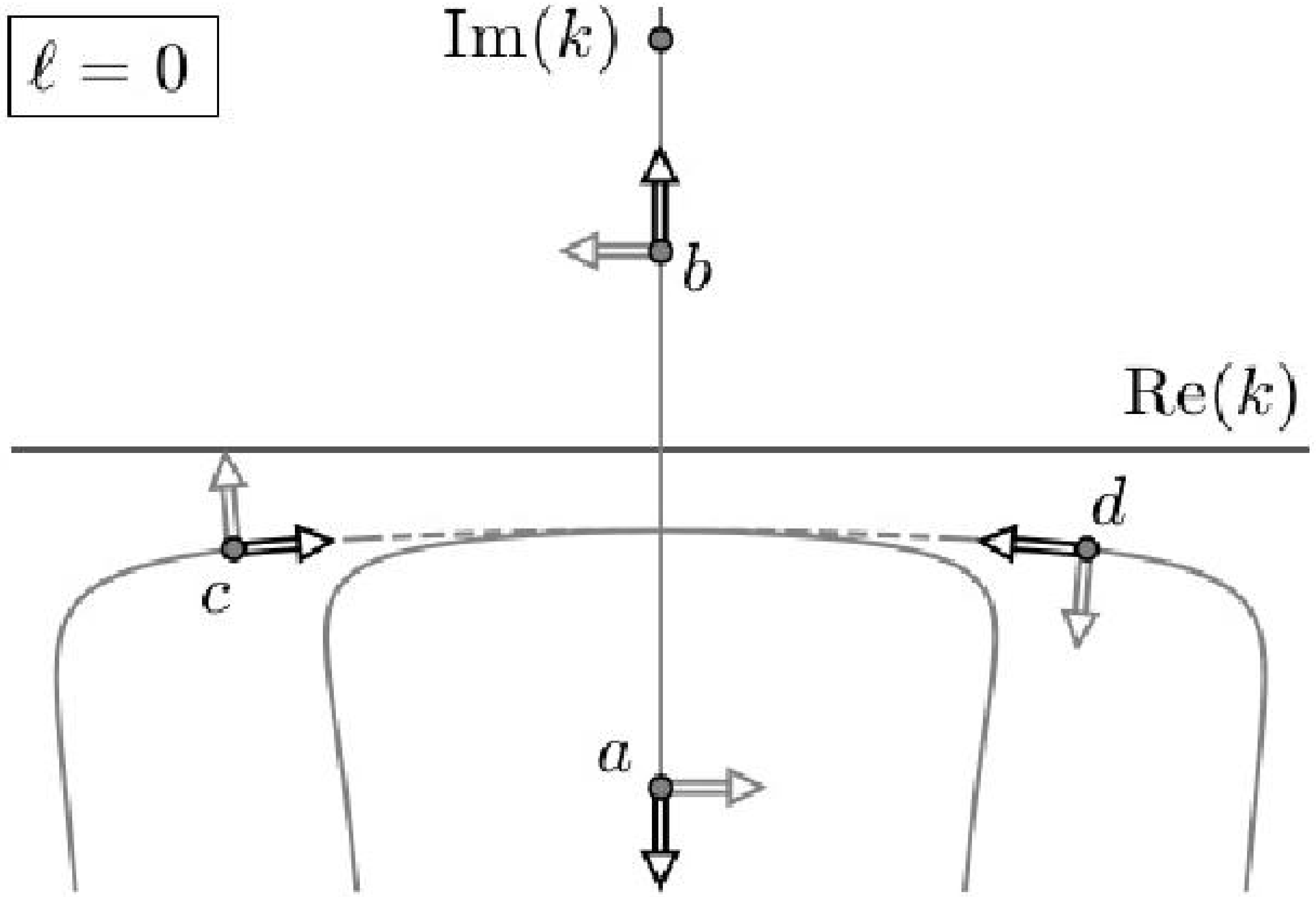
$$\Delta = A / (2|\epsilon_P|)$$

*Double resonance Feshbach theory:
B. Marcelis *et al*. PRA 70, 012701 (2004)

Scattering physics in terms of bound states

- **Bound states: poles of the scattering matrix**
  - **Poles in the complex plane**

$$S(k) = e^{-2i a_{bg}} \prod_n \left(\frac{k_n + k}{k_n - k} \right)$$



- **Background processes:**

$$a_{bg} = \langle f_1 m_{f_1} f_2 m_{f_2} | P_S a_S + P_T a_T | f_1 m_{f_1} f_2 m_{f_2} \rangle$$

[M.R. Goosen, Ph.D thesis]

Dipolar interactions in ABM

- **Long-range dipole interactions**
  - **Use known bound states potentials**

$$\langle \Psi_{S,l} | V^{dip} | \Psi_{S',l'} \rangle = -3\alpha^2 \langle \psi_{S,l} | \frac{1}{r^3} | \psi_{S',l'} \rangle \times$$

$$\sum_{q=-2}^{q=2} (-1)^q \sqrt{\frac{8\pi}{15}} \langle l\, m_l | Y_{2-q} | l'\, m_l' \rangle \langle S\, M_S\, m_{i_\alpha}\, m_{i_\beta} | \{\mathbf{s_1} \otimes \mathbf{s_2}\}_{\mathbf{2q}} | S'\, M_{S'}\, m_{i_\alpha}'\, m_{i_\beta}' \rangle$$

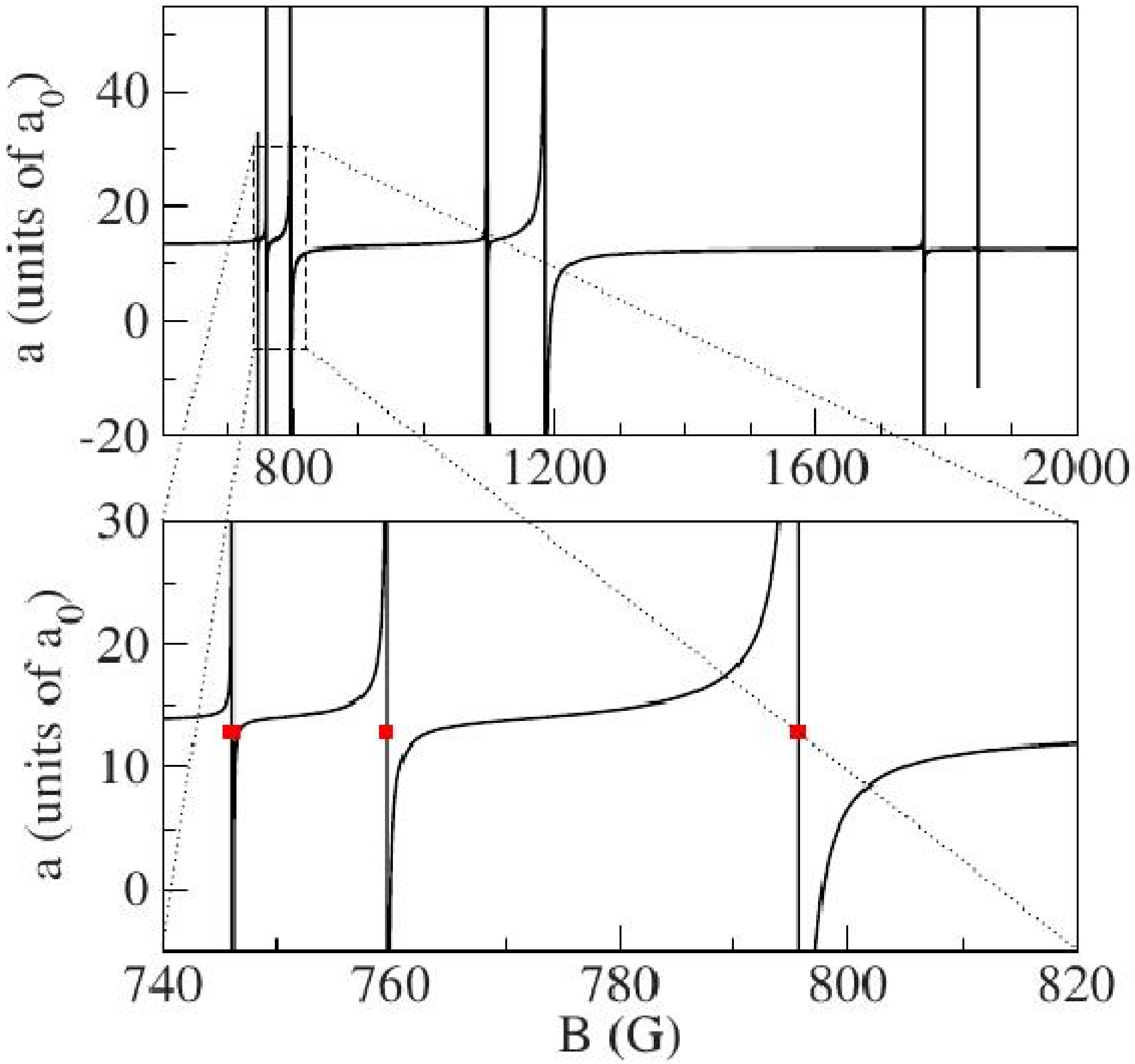
- **Leads to more complex molecular states**
- **Easy to calculate shifted energy levels in presence of the details of the contact interactions**
  - **Gives rise to dipolar-induced Feshbach resonances**

[M.R. Goosen, T.G. Tiecke, W. Vassen, and S.J.J.M.F. Kokkelmans, Feshbach resonances in 3He*-4He* mixtures, Phys. Rev. A 82, 042713 (2010).]

Example: control complexity in ^{6}Li-Na

- **Seemed to be very well known:**
  - **Experiment Ketterle group**

    [C. A. Stan, M. W. Zwierlein, C. H. Schunck, S. M. F. Raupach, and W. Ketterle, Phys. Rev. Lett. 93, 143001 2004]



  - **Analyzed in**

    [M. Gacesa, P. Pellegrini, and R. Côté, Phys. Rev. A78, 010701(R) (2008).]

	^{7}Li+^{23}Na	^{6}Li+^{23}Na
a_S	39.7 ± 0.5 (1.6)	15.9 ± 0.3 (1.5)
a_T	36.1 ± 0.3 (4.6)	12.9 ± 0.6 (4.5)
v^S_{last}	47	45
v^T_{last}	12	11
E^S_{last} (MHz)	-1505 ± 3	-1.6 ± 0.2
E^T_{last} (MHz)	-7112 ± 12	-5720 ± 16

Further experiments: no agreement

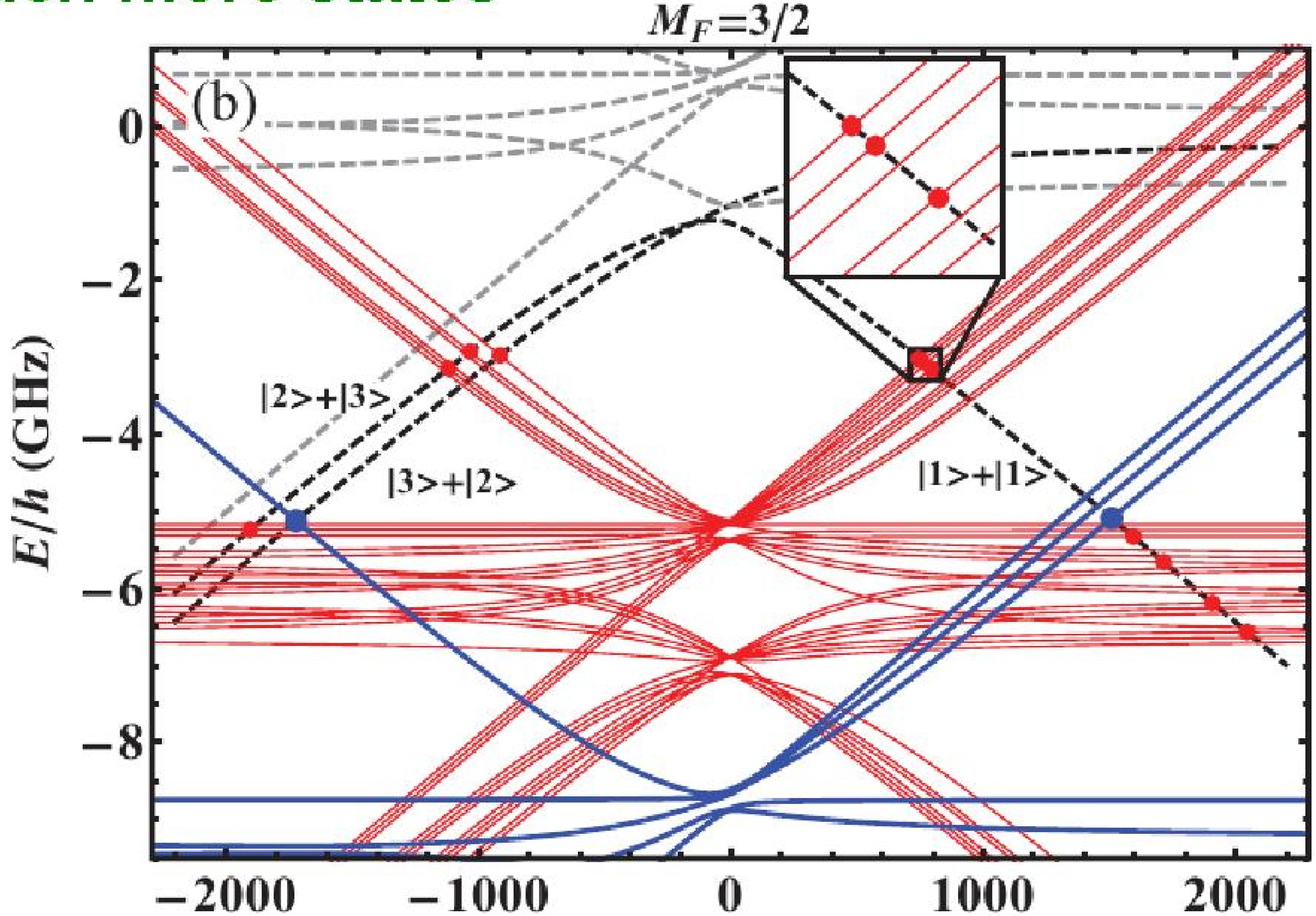
- Experiment in Heidelberg
- 26 interspecies Feshbach resonances at fields up to 2050 G

  - Much more resonances than predicted
  - New resonances didn't match at all with the predictions

^{6}Li + Na	M_F	Expt. B_0^{expt} (G)
$\|2\rangle + \|1\rangle$	1/2	771.8 (5)
		822.9 (5)
		1596.8 (4)
		1716.7 (3)
$\|1\rangle + \|3\rangle$	−1/2	1002.3 (5)
		1088.5 (5)
$\|3\rangle + \|1\rangle$	−1/2	800.9 (2)
		852.0 (7)
		1566.3 (8)
		1597.5 (7)
		1717.3 (2)
$\|1\rangle + \|1\rangle$	3/2	745.2 (3)
		759.0 (3)
		795.2 (2)
		1510.4 (3)
		1596.5 (5)
		1715.6 (8)
		1908.9 (7)
		2046.9 (9)
$\|2\rangle + \|3\rangle$	−3/2	1031.7 (3)
		1117.3 (6)
		1902.4 (6)
$\|3\rangle + \|2\rangle$	−3/2	913.2 (6)
		1720.5 (3)
$\|6\rangle + \|1\rangle$	5/2	1575.8 (9)
		1700.4 (7)

Analyse with ABM

- **Now also include the spin-spin interaction (magnetic dipoles)**
  - **Much more states**

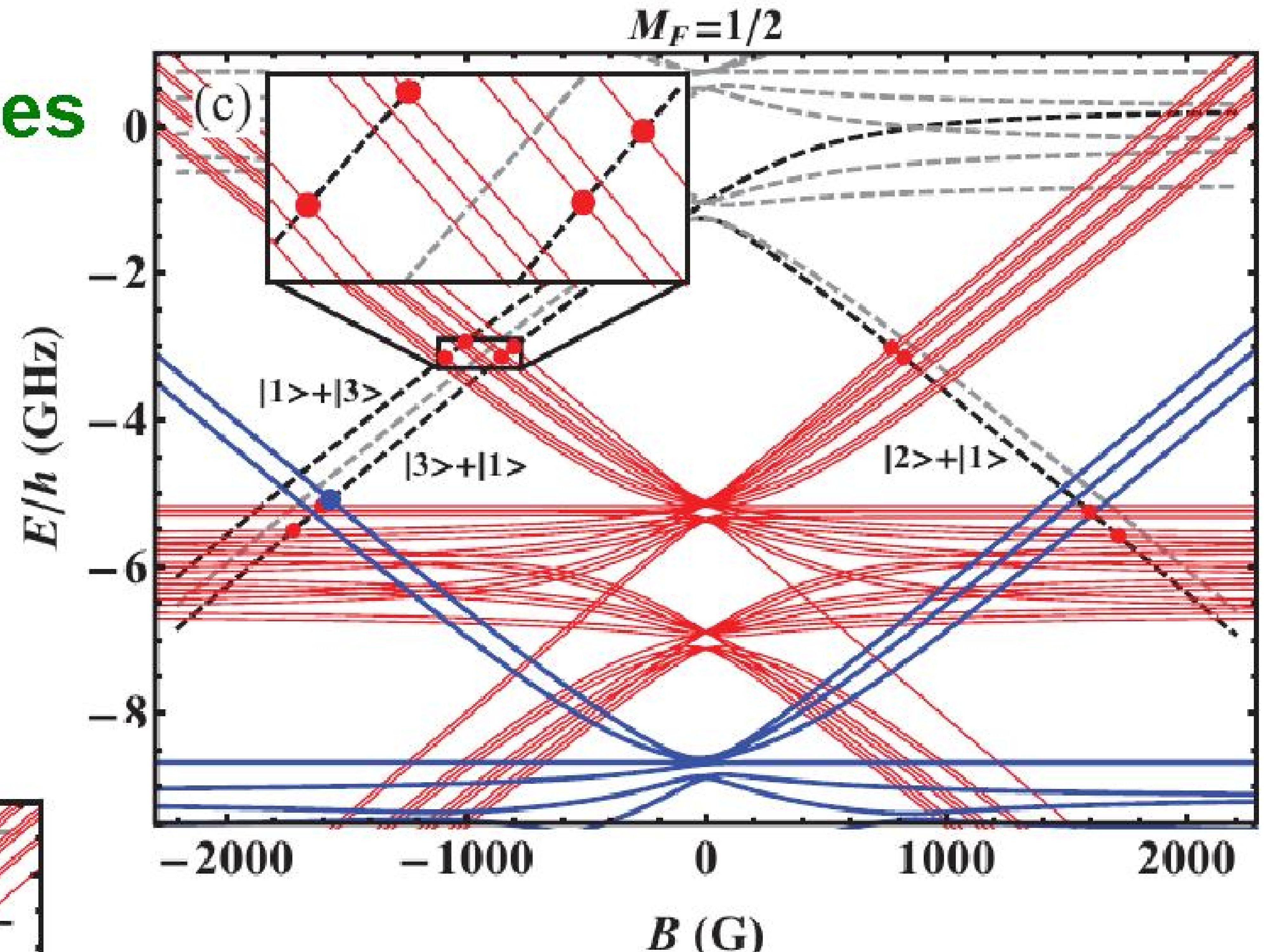


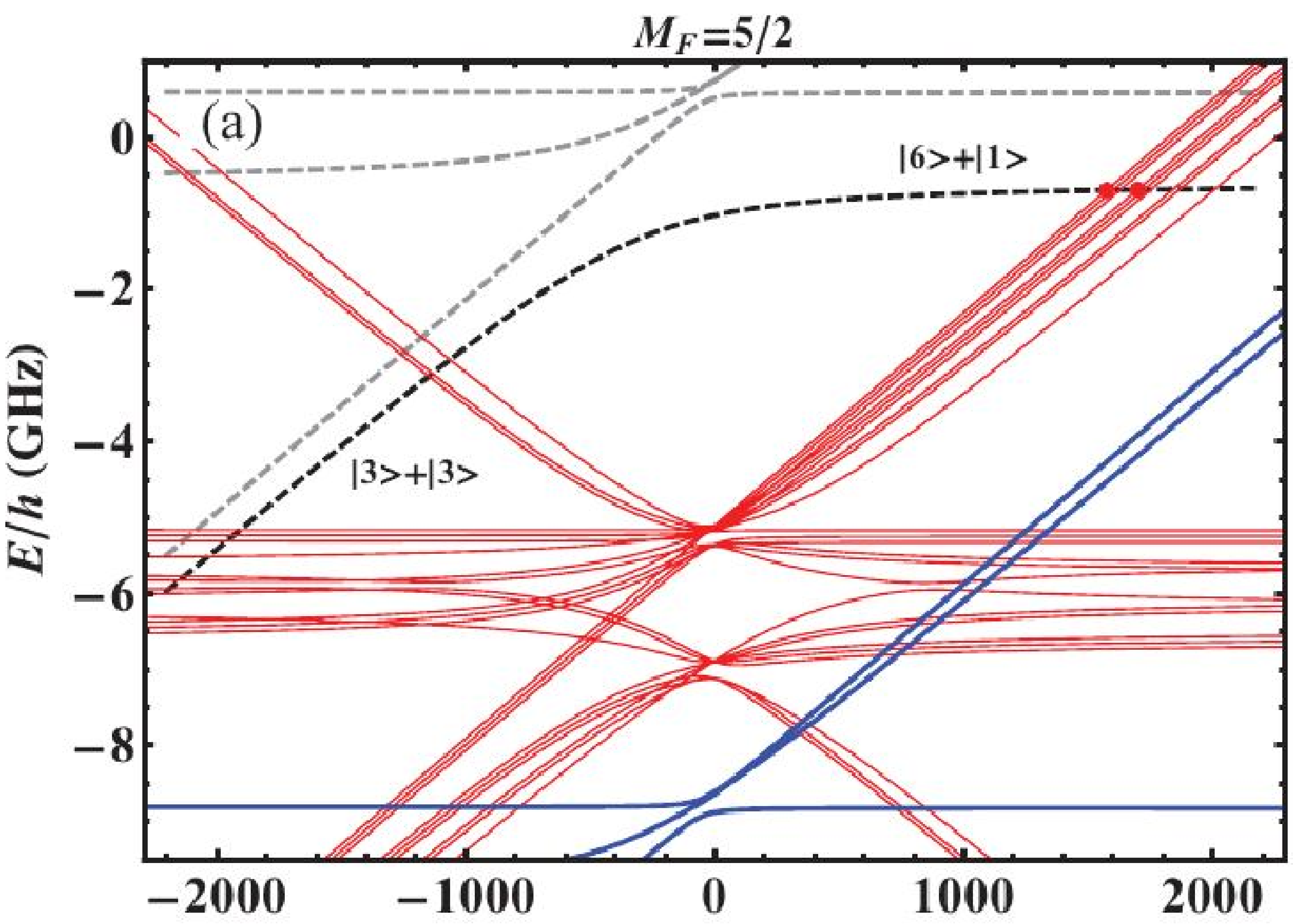
s-wave
d-wave

T. Schuster, R. Scelle, A. Trautmann, S. Knoop, M. K. Oberthaler, M. M. Haverhals, M. R. Goosen, S. J. J. M. F. Kokkelmans, and E. Tiemann, Phys. Rev. A 85, 042721 (2012).

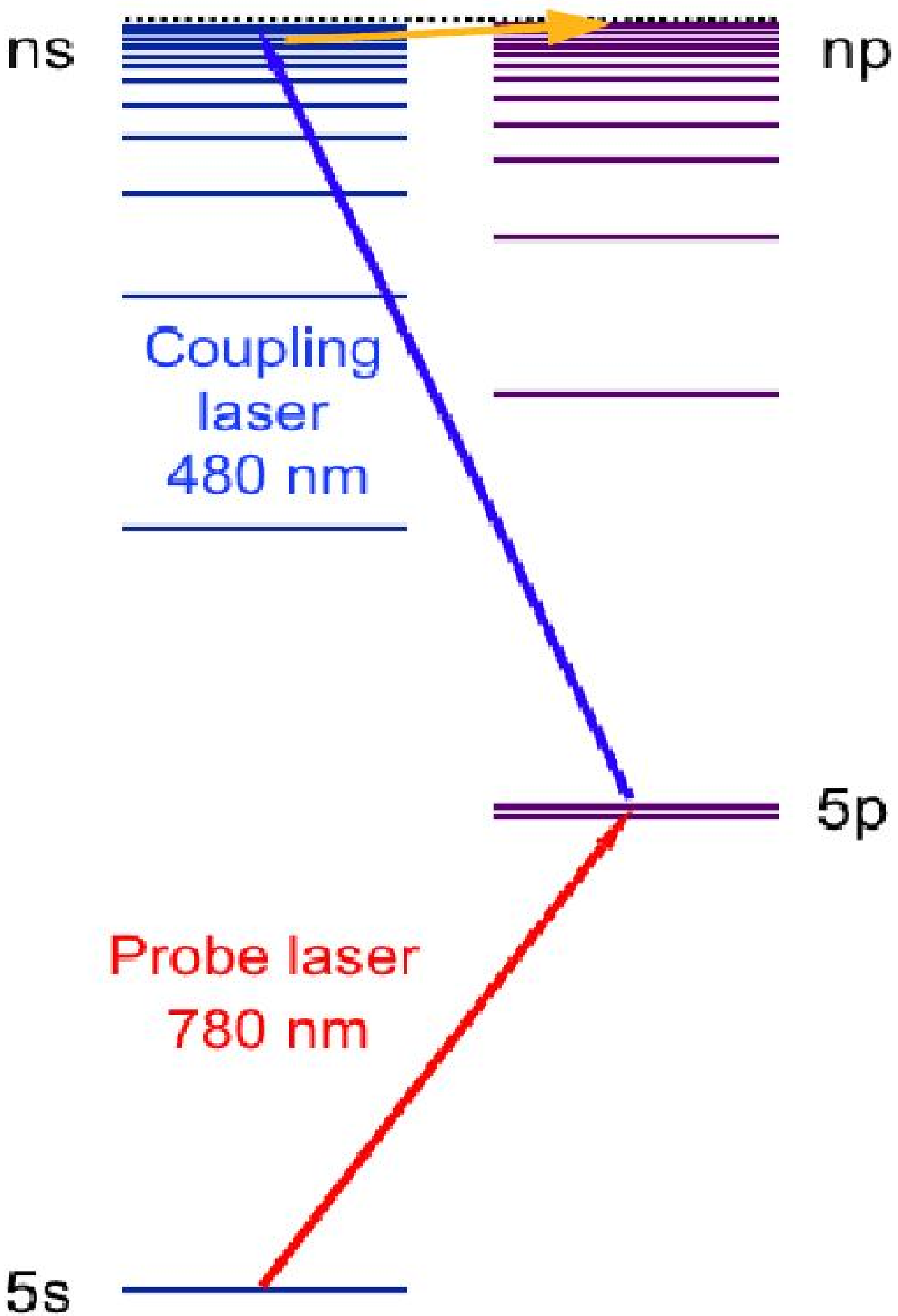
Other spin states

- Mainly dipolar resonances
- d-wave scenario
- Very narrow





Nice properties of Rydberg atoms



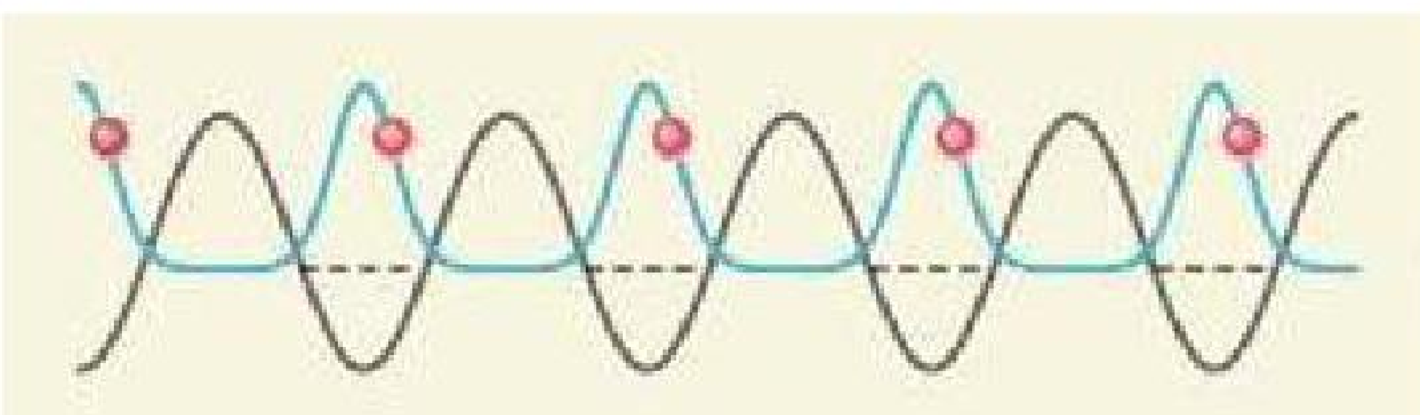
Rubidium atoms

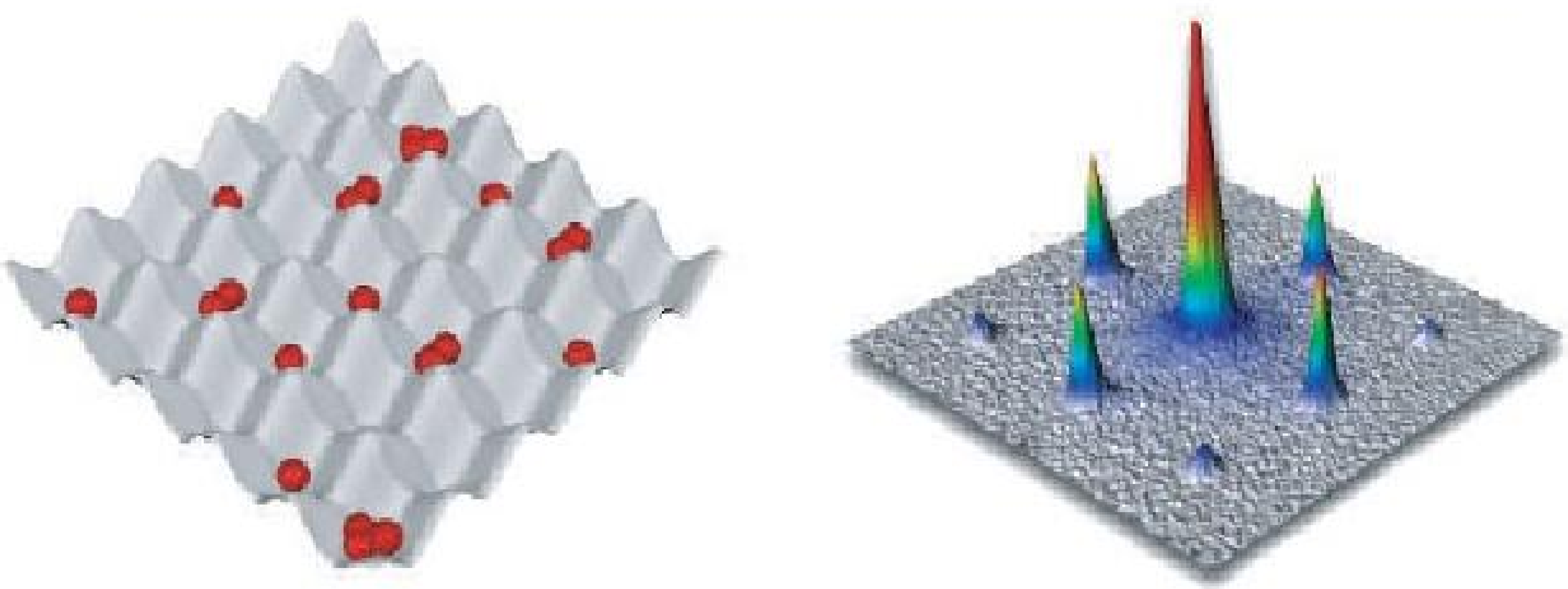
- **Rydberg states: large *n*:**
  - **Size** n^2
  - **Dipole moment** n^2
  - **Lifetime** n^3
  - **Polarizability** n^7 **(E-fields)**
  - **van der Waals coefficient** n^{11}

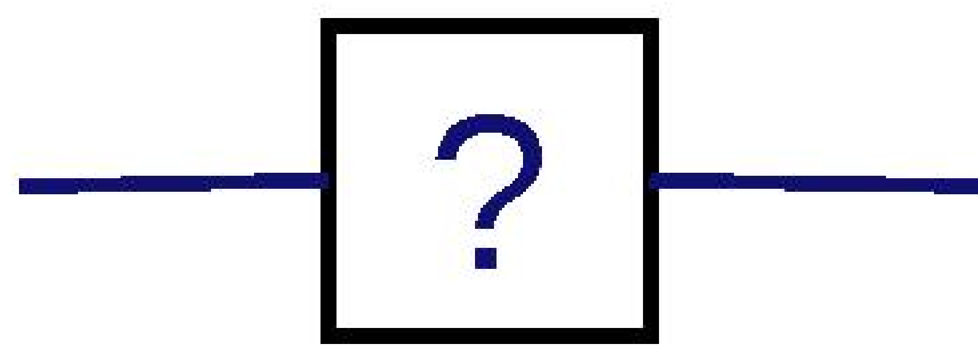
Our interest in Rydberg atoms

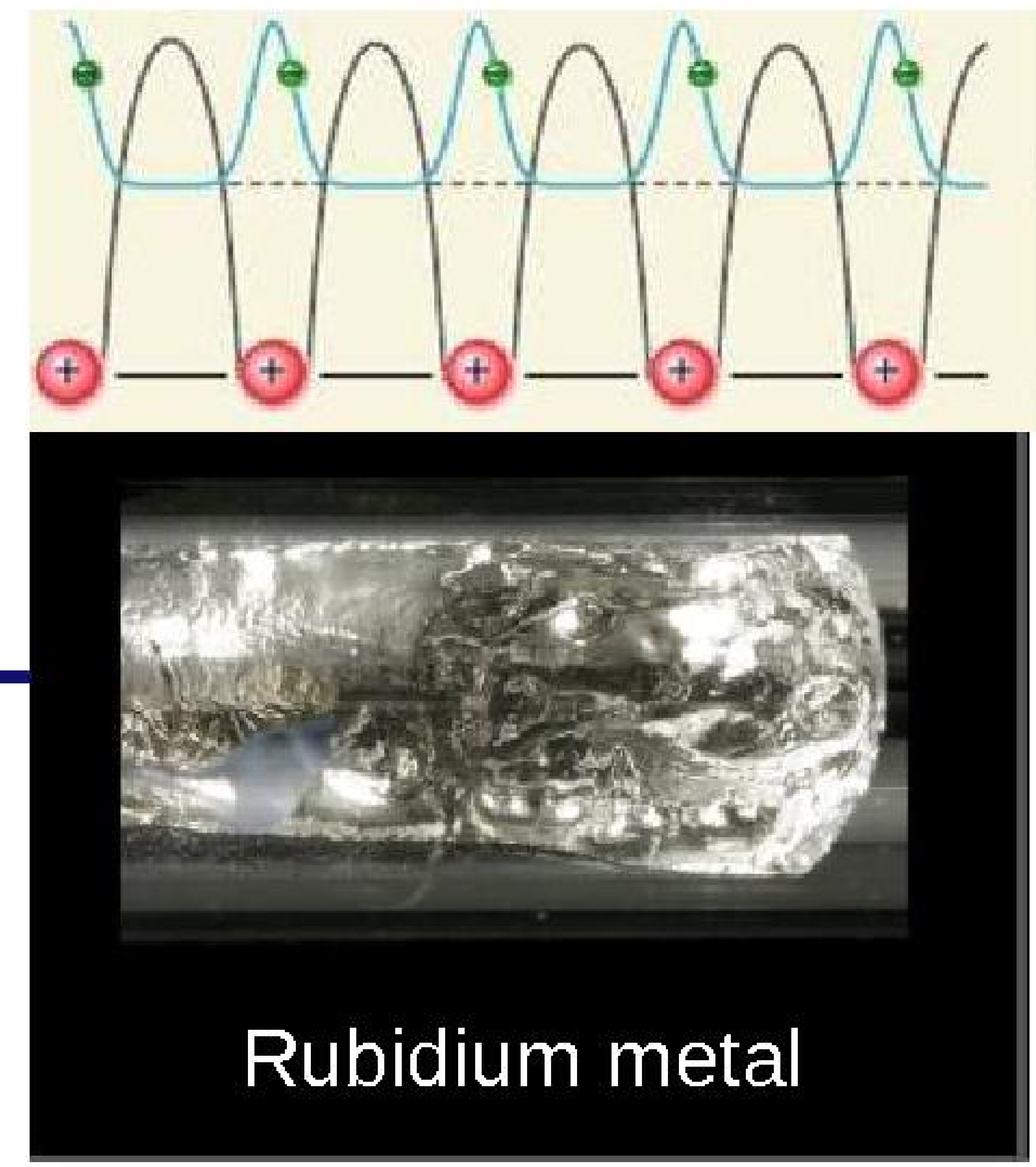
Artificial lattices in atomic physics:

- **Model for real condensed-matter systems?**
- **Crucial problem: the weakness of interactions**







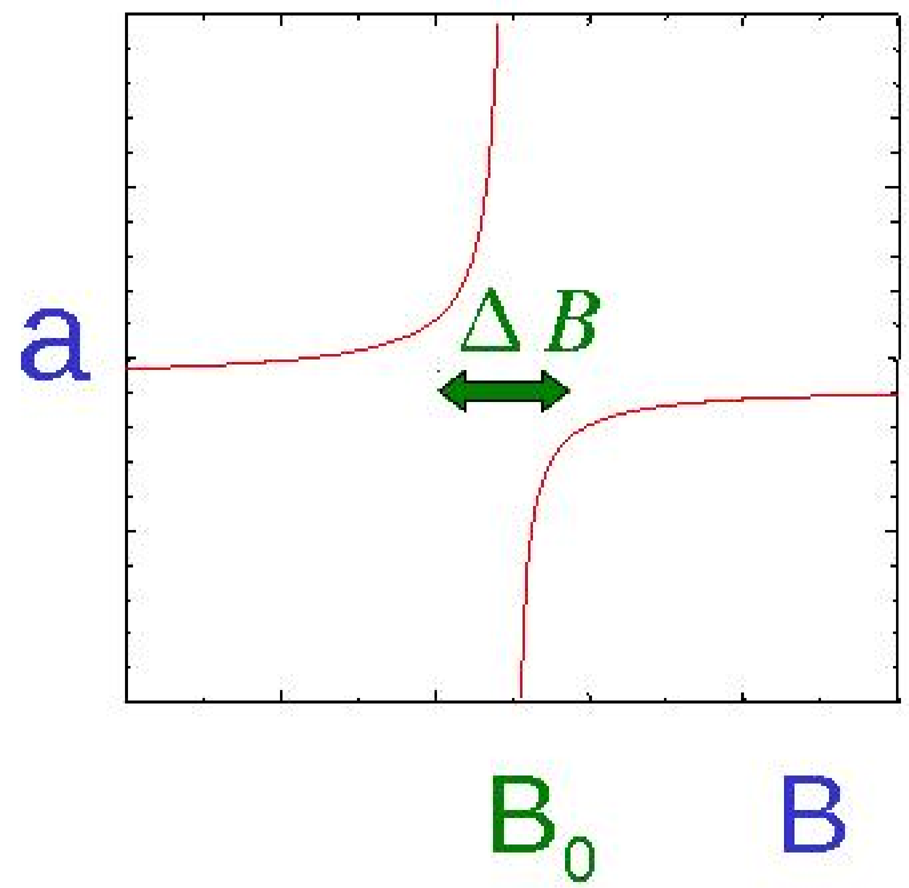


Rubidium atoms in optical lattice

Weak interactions ⟷ Strong interactions

Range of interactions

- **Van der Waals range:** $R_{vdW} \simeq (m C_6 / 16 \hbar^2)^{1/4}$
- **Ultracold ground-state atoms**
  - **Manipulate interactions via Feshbach resonances** $a \gg n^{-1/3}$



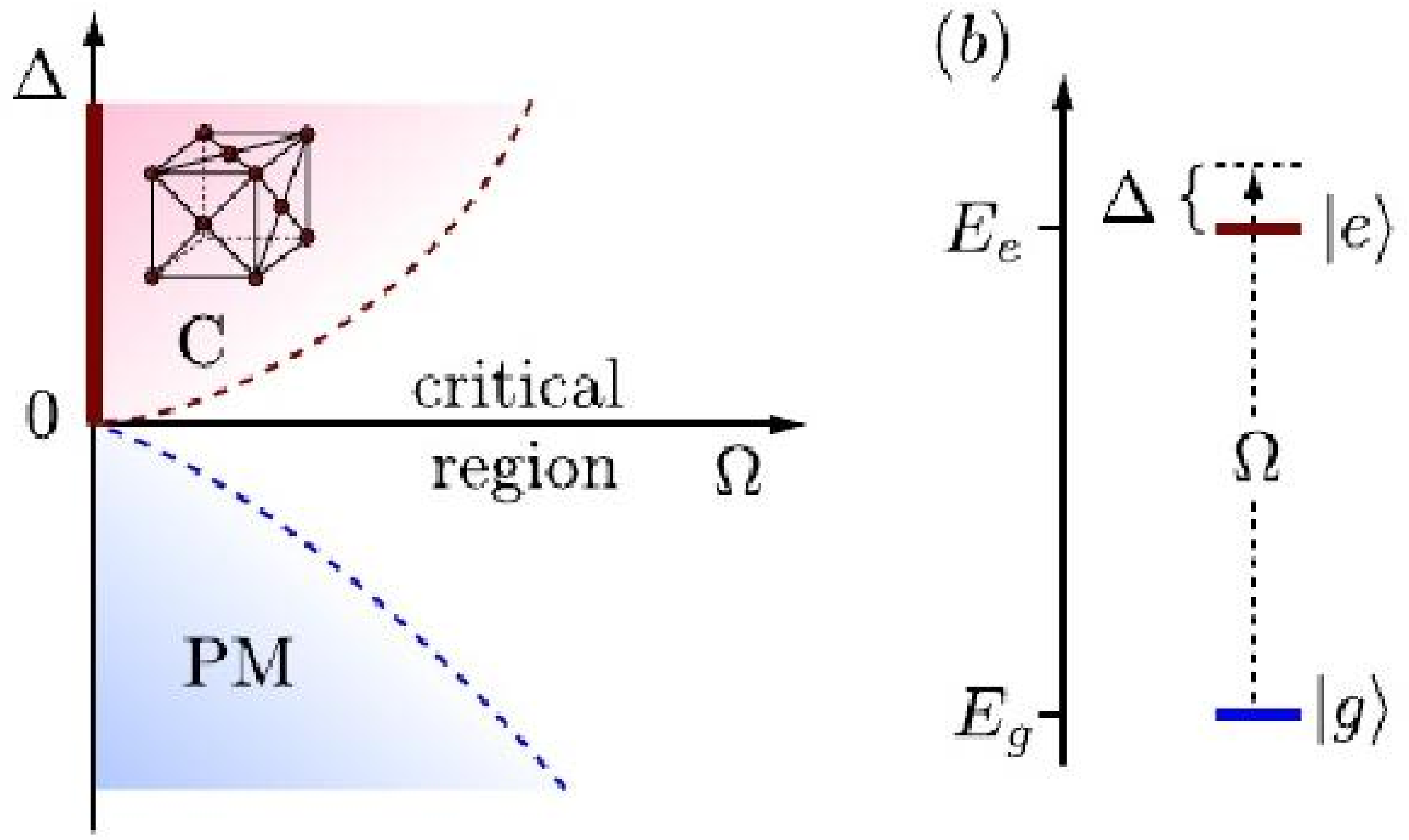
$$a = a_{bg}\left(1 - \frac{\Delta B}{B - B_0}\right)$$

  - **But always** $R_{vdW} \ll n^{-1/3}$ **Fermi gas at unitarity:** $\Gamma = \left|\frac{E_{\text{int}}}{E_F}\right| \leqslant 0.6$

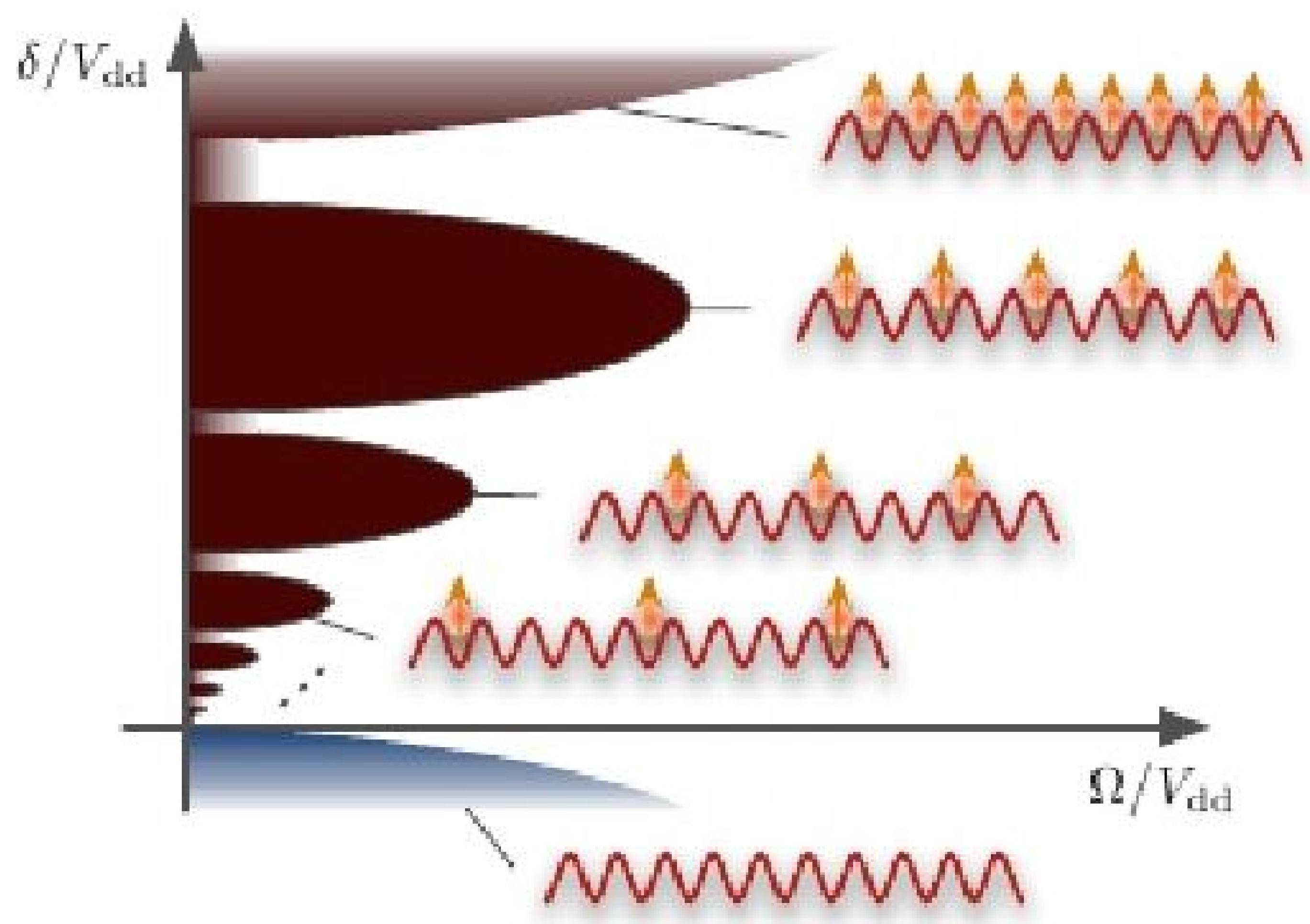
- **Rydberg atoms**

$$C_6 \sim n^{11} \quad \Rightarrow \quad R_{vdW} > n^{-1/3}$$

Predictions many-body Rydberg systems

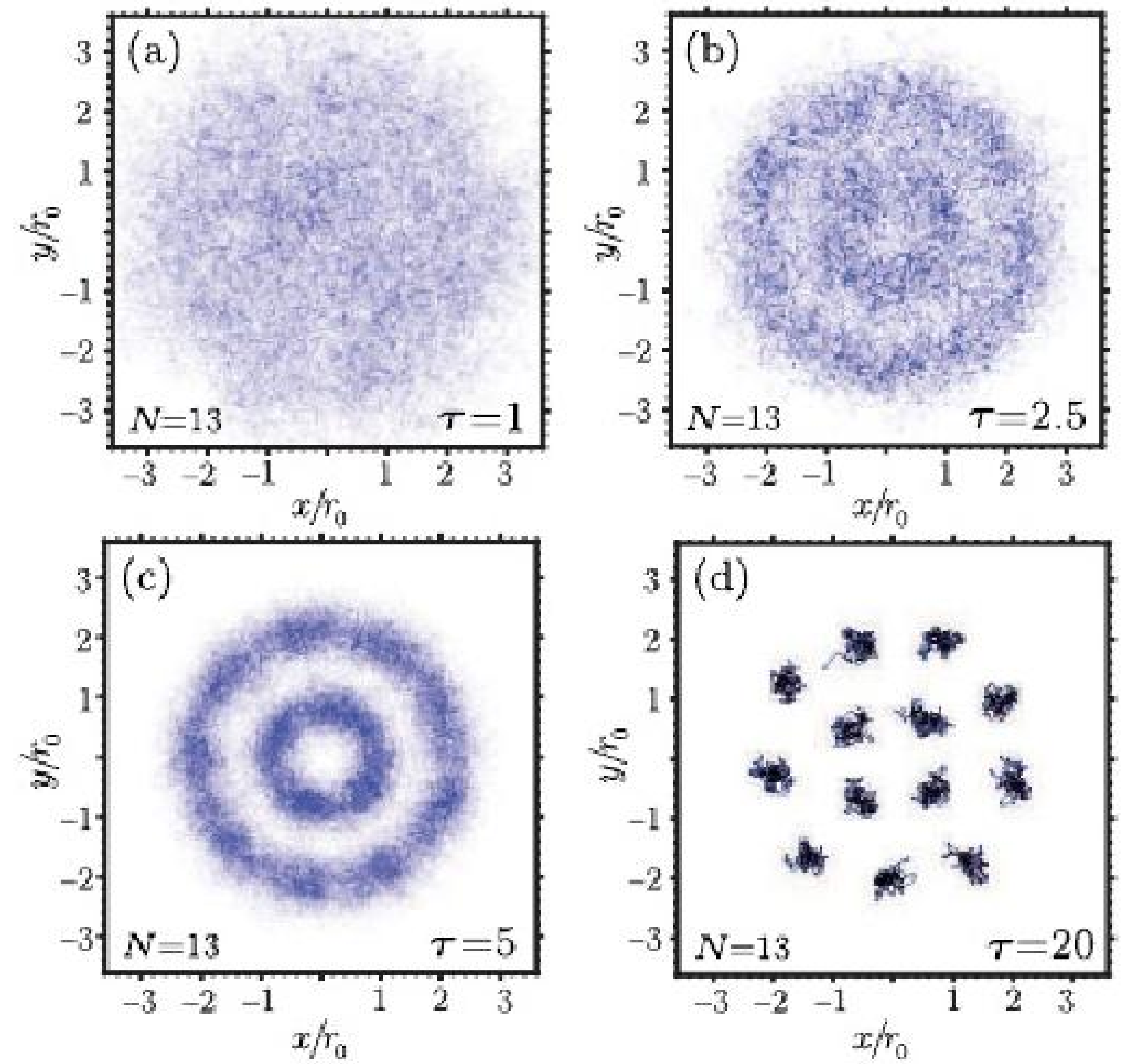


H. Weimer, R. Löw, T. Pfau, and H. P. Büchler, Phys. Rev. Lett. **101**, 250601 (2008)



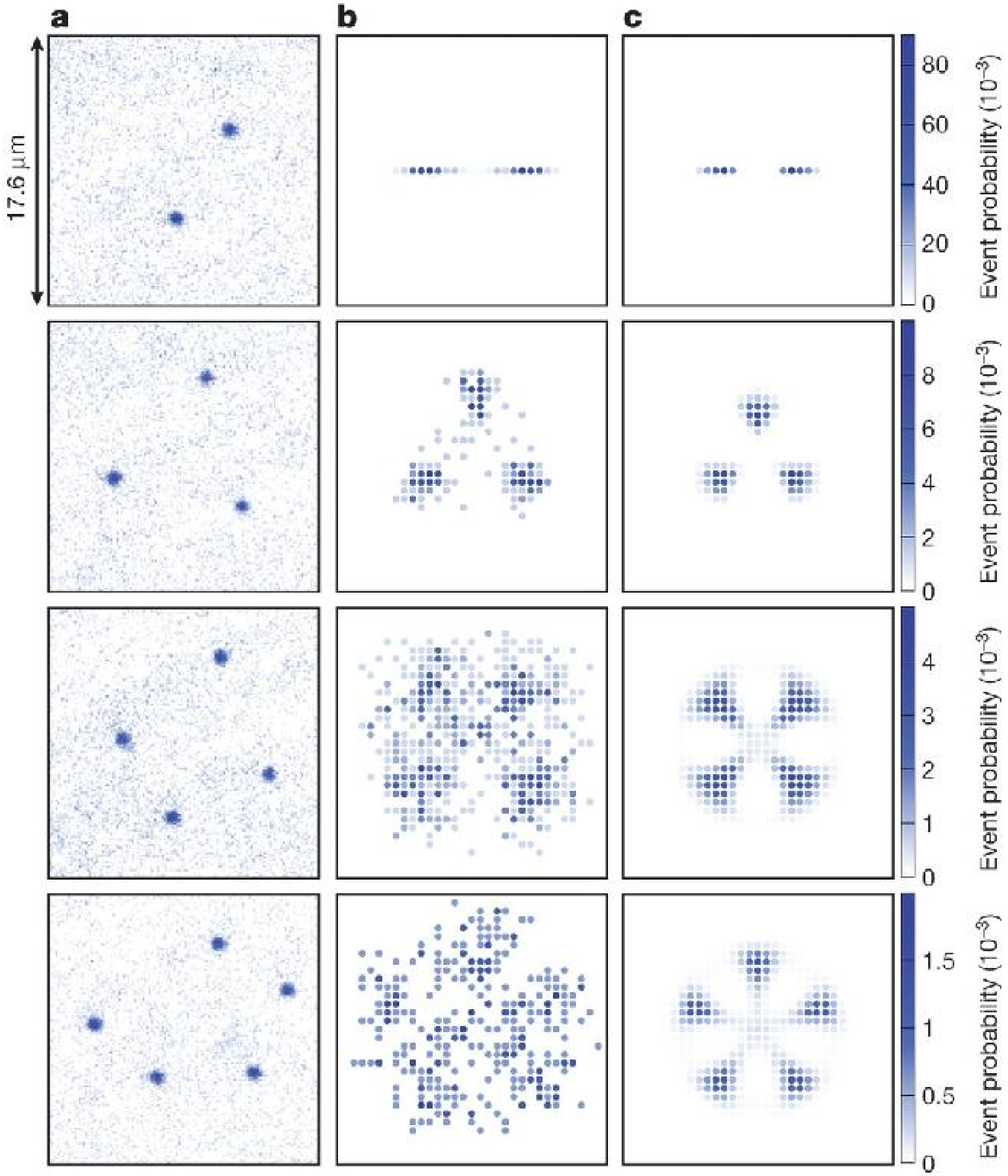
[J. Schachenmayer et al., arXiv/1003.5858]

Special Issue in J. Phys. B, **44**, 18

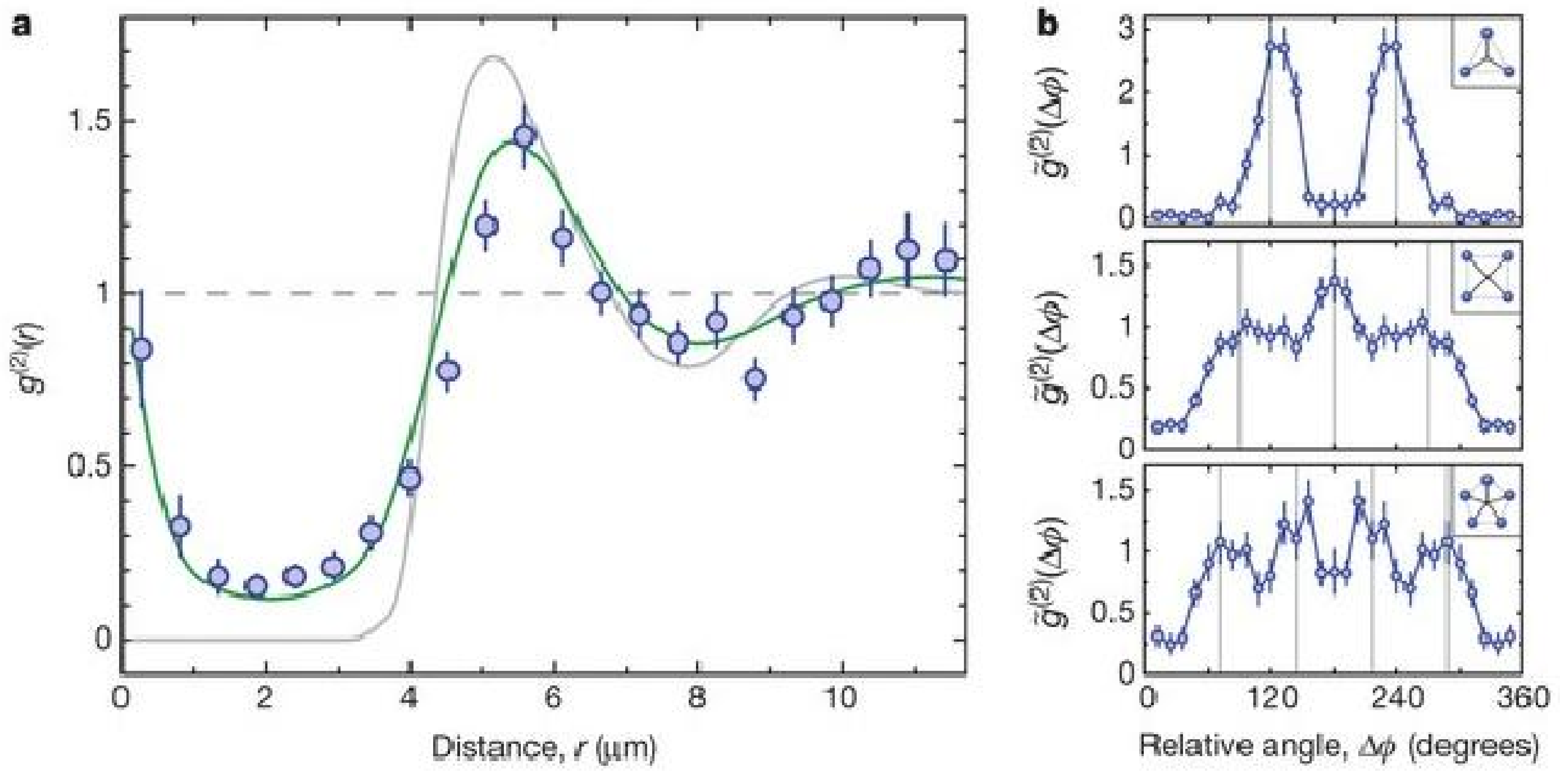


G. Pupillo, et al, Phys. Rev. Lett. **104**, 223002 (2010)

Recent experimental observation



Rydberg atoms created at Blockade radius

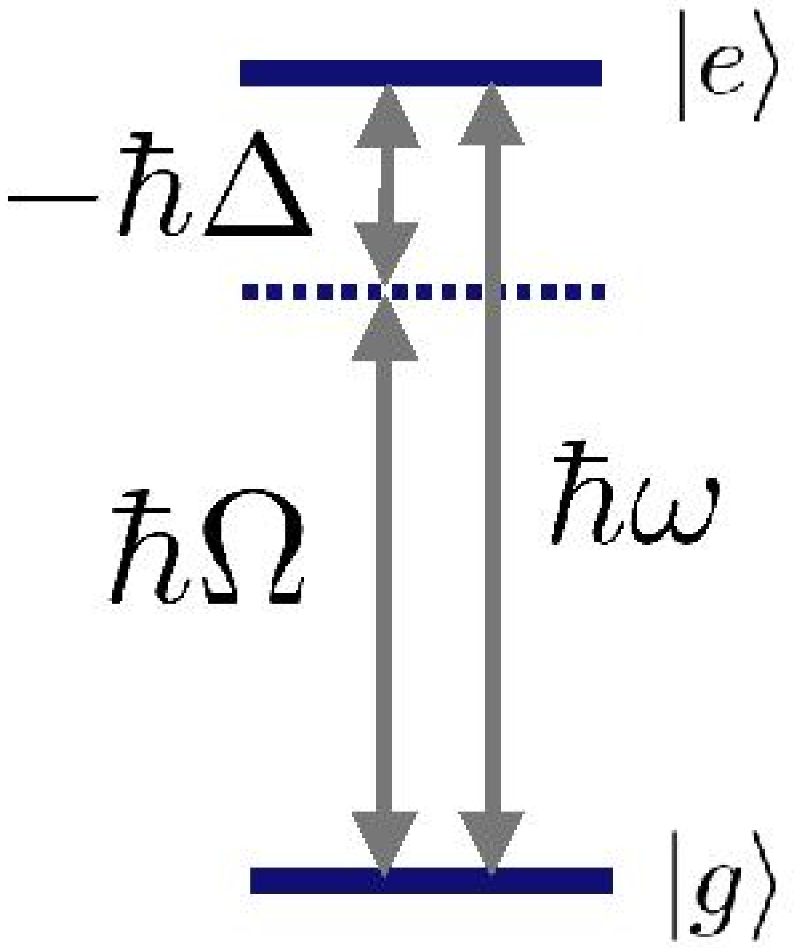


Peter Schauß et al, Nature 491, 87 (2012)

Crystal Creation by changing Laser Parameters

2 Laser Parameters

- Detuning Δ
- Intensity (Rabi frequency) Ω



Single Particle Hamiltonian

$$\hat{H} = \hbar \begin{pmatrix} 0 & \Omega/2 \\ \Omega/2 & -\Delta \end{pmatrix}$$

Idea based on

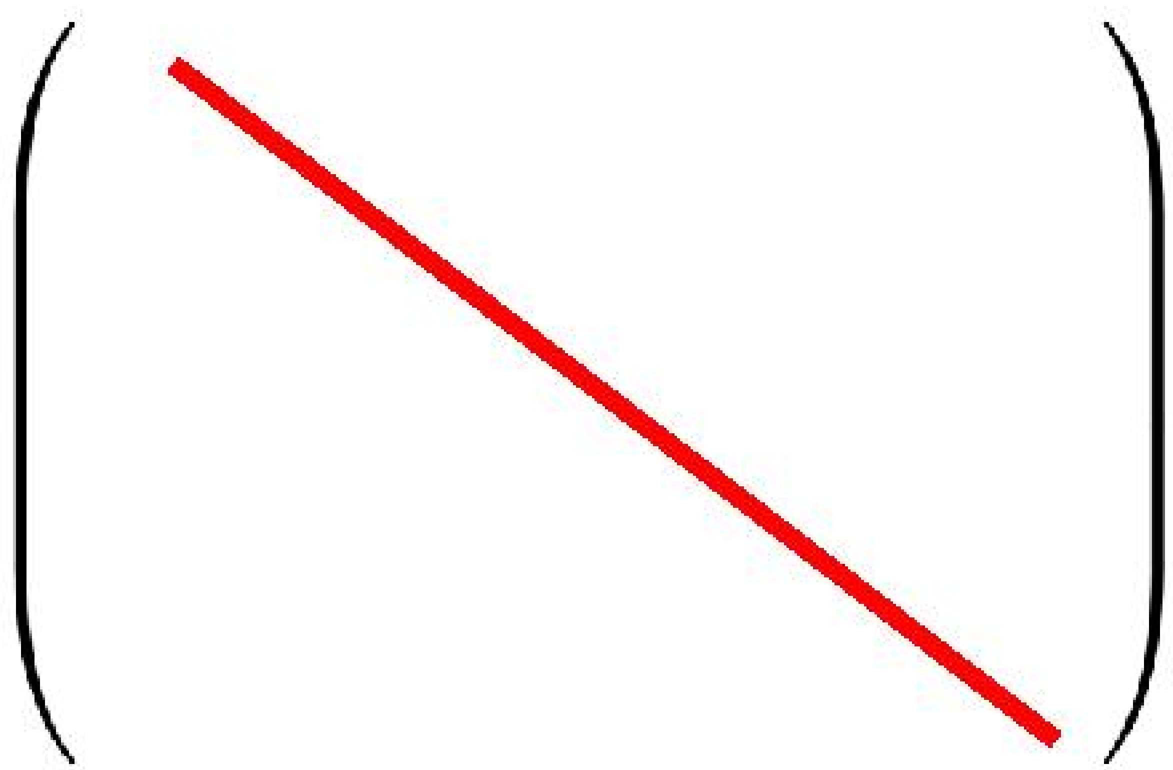
T. Pohl, E. Demler, and M. D. Lukin Phys. Rev. Lett. 104, 043002, (2010)

Crystal Creation by changing Laser Parameters

$$\hat{H} = -\hbar\Delta \sum_i |g_i\rangle\langle g_i| + \sum_{i<j} \frac{C_6}{|\mathbf{r}_i - \mathbf{r}_j|^6} |e_i e_j\rangle\langle e_i e_j|$$

$$\hat{H} = -\hbar\Delta \sum_i |g_i\rangle\langle g_i| + \frac{\hbar\Omega}{2} \sum_i \left(|e_i\rangle\langle g_i| + |g_i\rangle\langle e_i|\right) + \sum_{i<j} \frac{C_6}{|\mathbf{r}_i - \mathbf{r}_j|^6} |e_i e_j\rangle\langle e_i e_j|$$

$\hat{H} =$



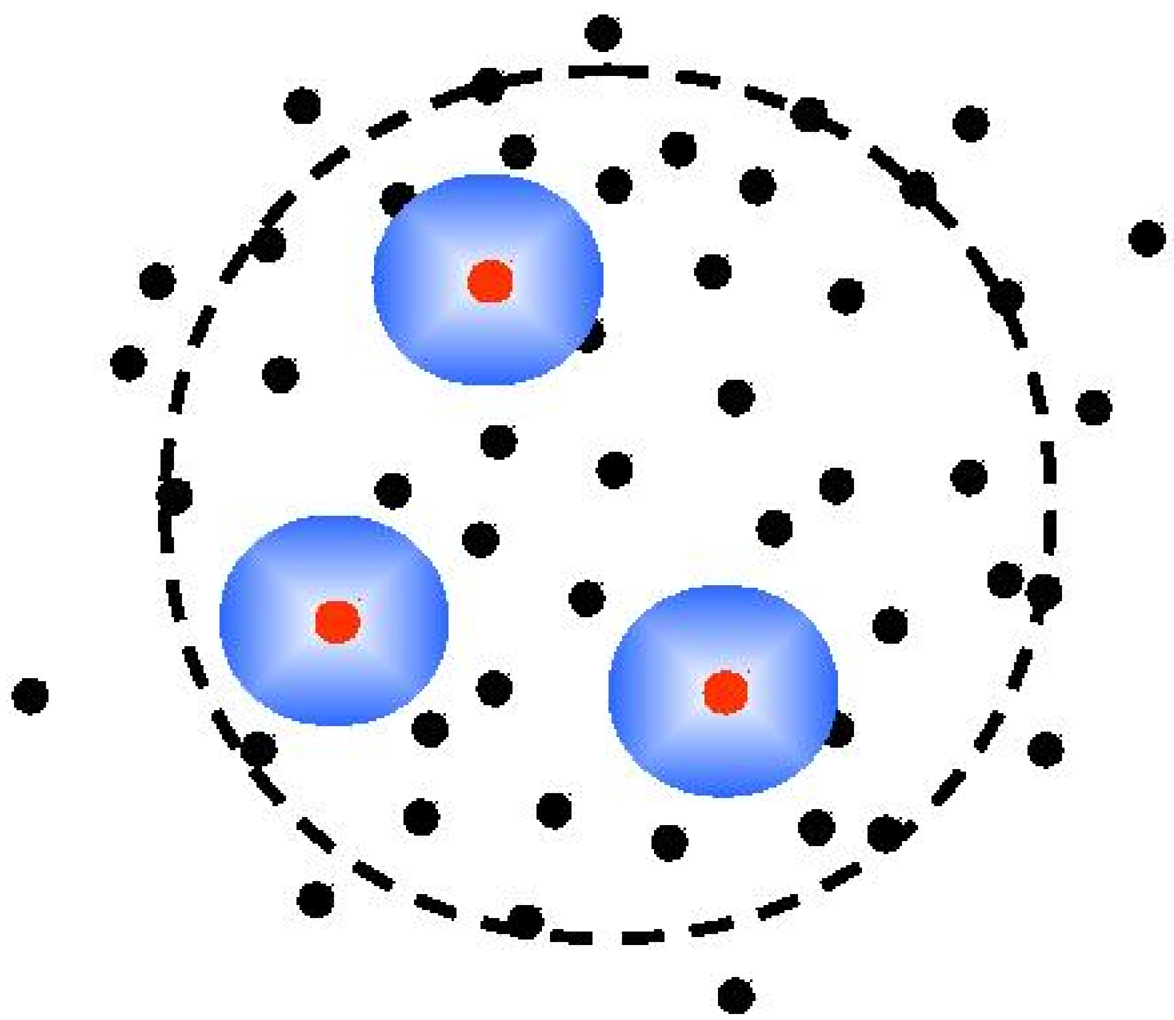
$|gggggg..\rangle$

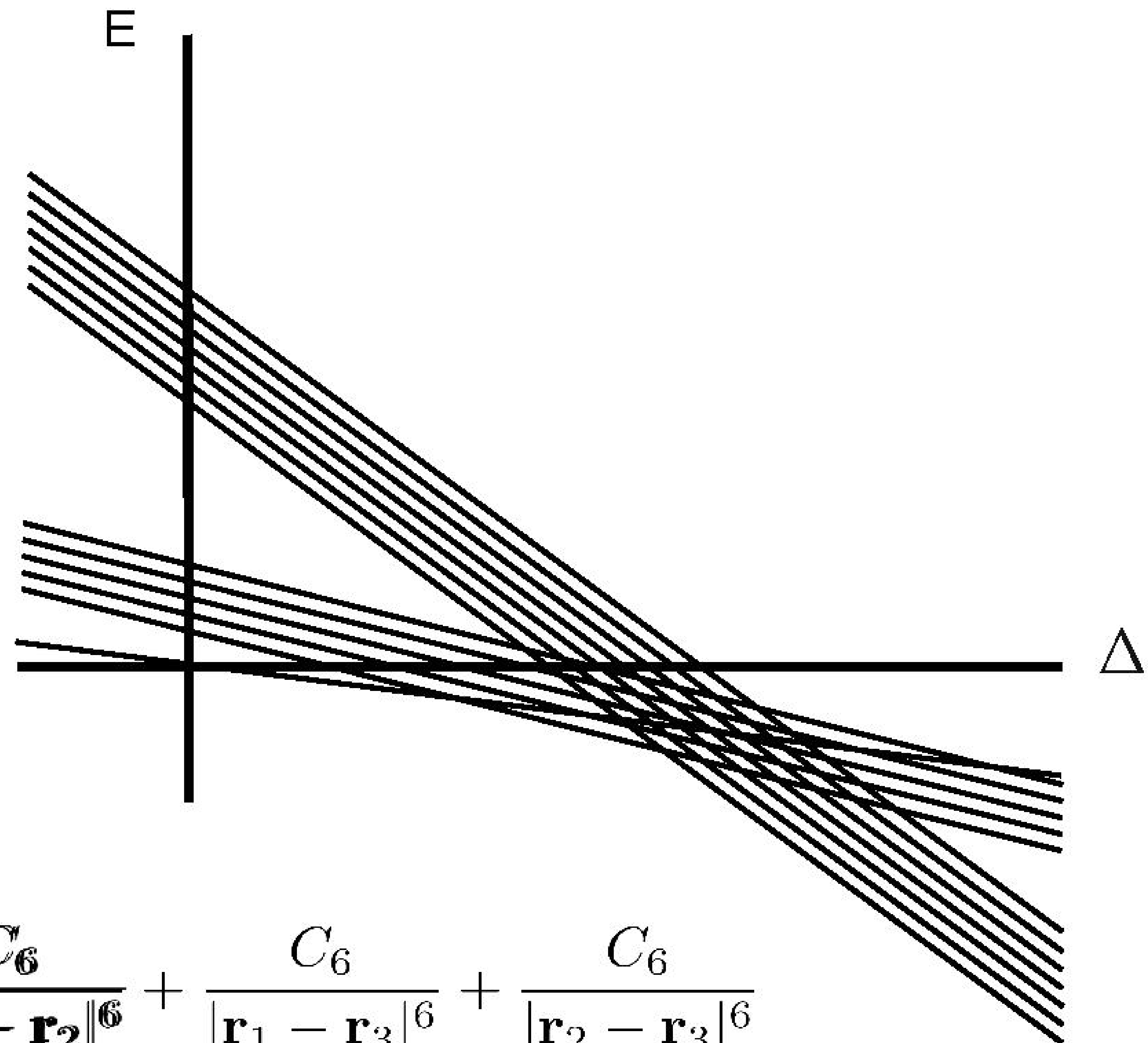
$|eegggeg..\rangle$

$|gggegeg..\rangle$

Energy as function of detuning Δ

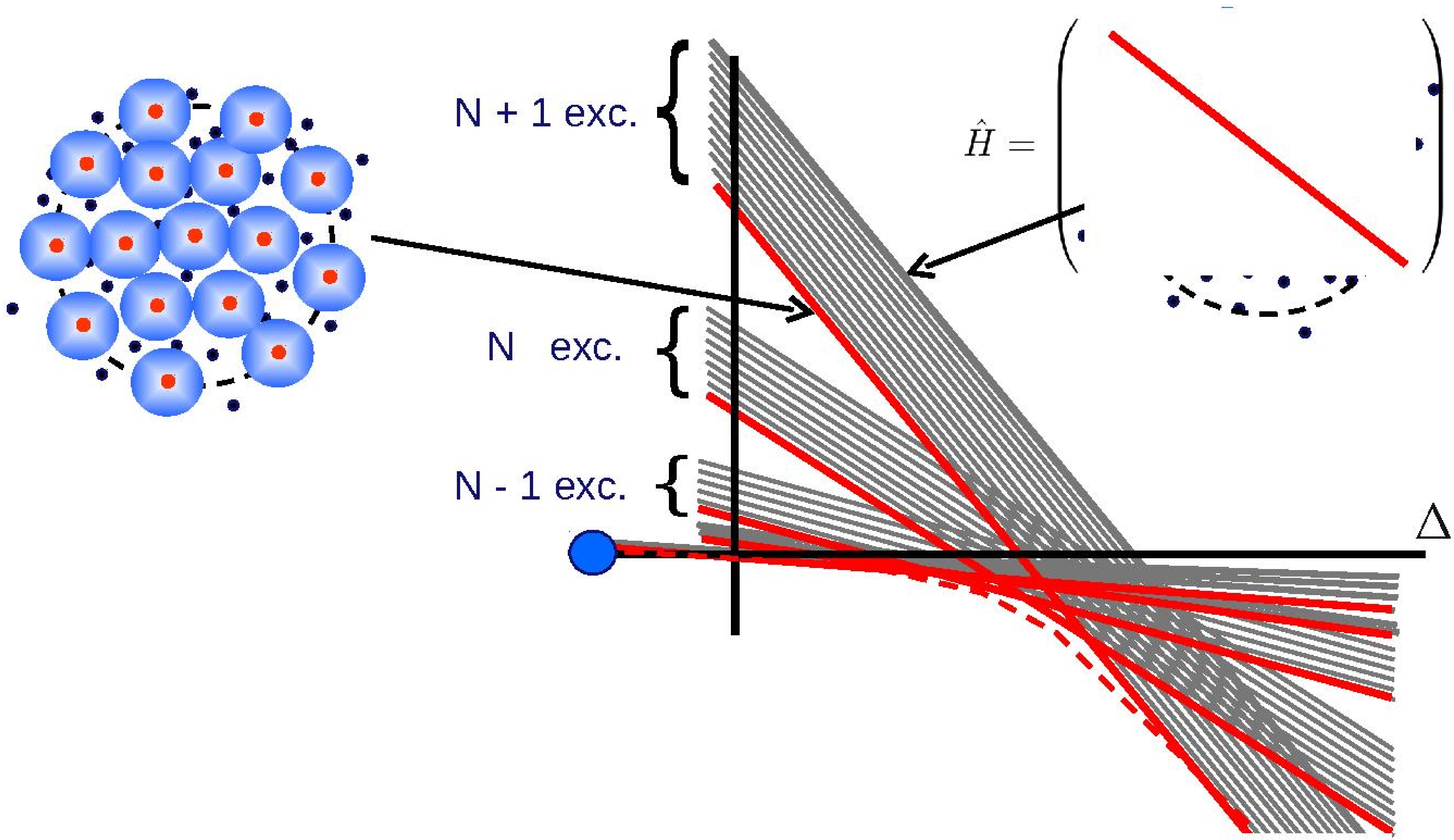
$|gggggggg\text{--}\rangle$





$$E = 3\hbar\Delta + \frac{C_6}{|\mathbf{r}_1 - \mathbf{r}_2|^6} + \frac{C_6}{|\mathbf{r}_1 - \mathbf{r}_3|^6} + \frac{C_6}{|\mathbf{r}_2 - \mathbf{r}_3|^6}$$

Energy as function of detuning Δ



Simulating the experiment (1D)

1D excitation volume

MOT densities	10^{10}, 10^{11} cm^{-3}:	~ 30	particles
Dipole trap densities	~10^{14} cm^{-3}:	~ 2000	particles
BEC densities	~10^{14} cm^{-3}:	~ 40.000	particles

Goal: create 5 or 6 excitations

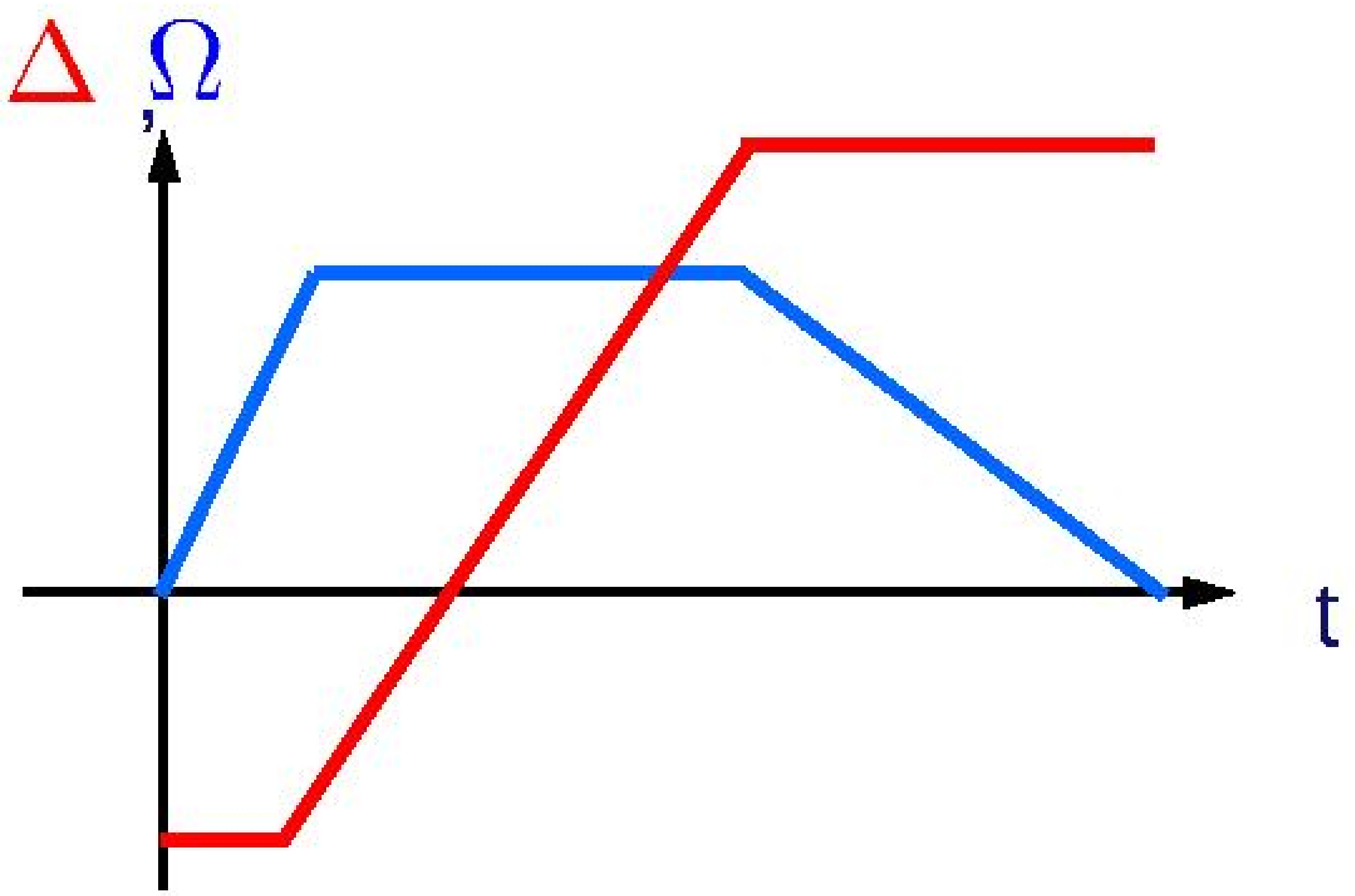
System

Superatomic Hamiltonian, time dependent detuning and intensity

$$\hat{H}(t) = -\hbar\Delta(t)\sum_i |g_i\rangle\langle g_i| + \frac{\hbar}{2}\sqrt{M_i}\Omega(t)\sum_i \left(|e_i\rangle\langle g_i| + |g_i\rangle\langle e_i|\right) + \sum_{i<j} |e_i e_j\rangle \frac{C_6}{|\mathbf{r}_i - \mathbf{r}_j|^6} \langle e_i e_j|$$

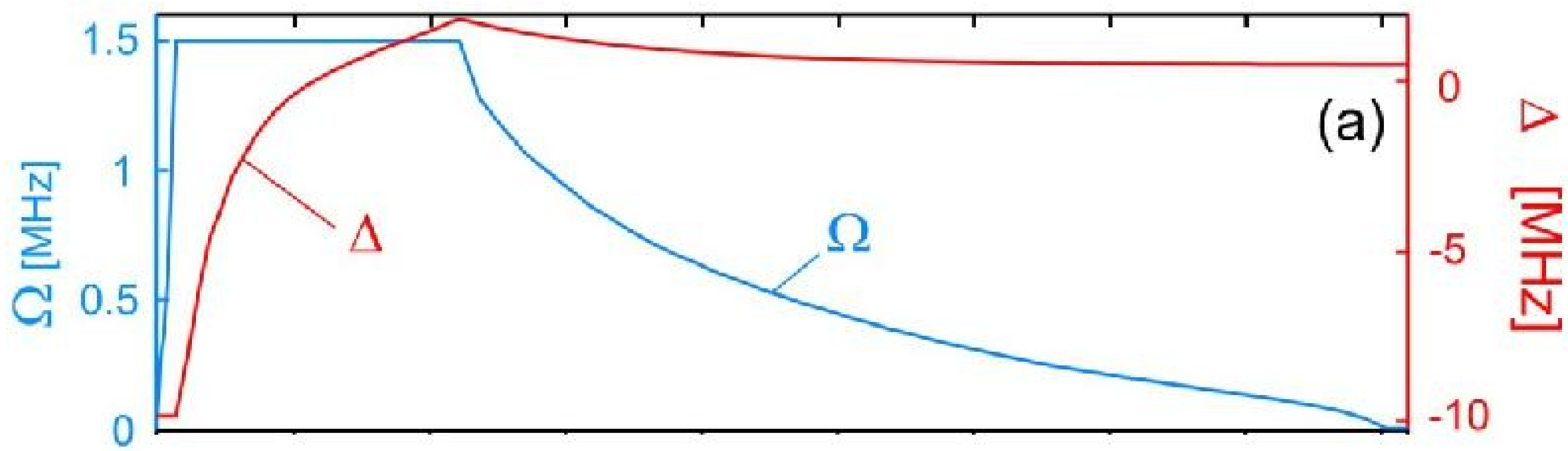
Start: $|\Psi(0)\rangle = |gggggg...\rangle$

Solve: $i\hbar \frac{\mathrm{d}\,|\Psi(t)\rangle}{\mathrm{d}t} = \hat{H}(t)\,|\Psi(t)\rangle$



Results MOT

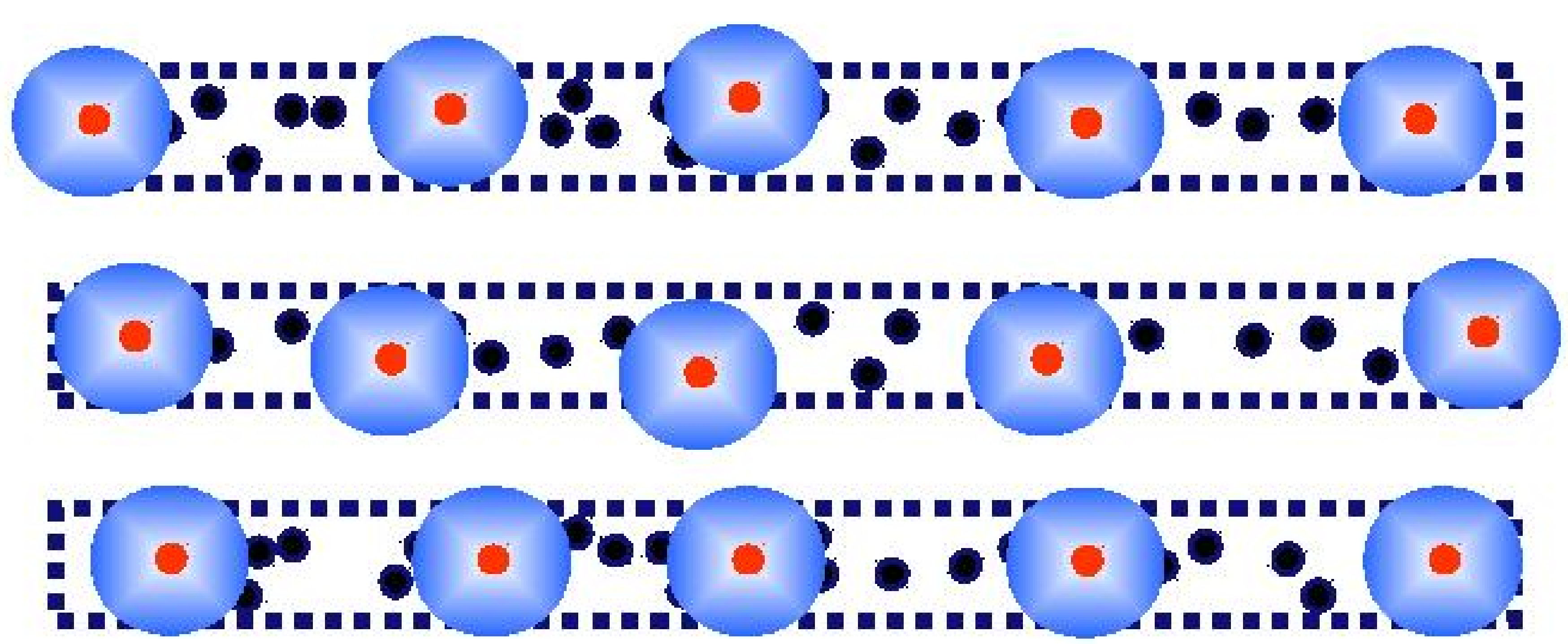
10^{11} cm^{-3}: ~30 particles in exc. vol.



E

N + 1 exc.

Monte Carlo integration:



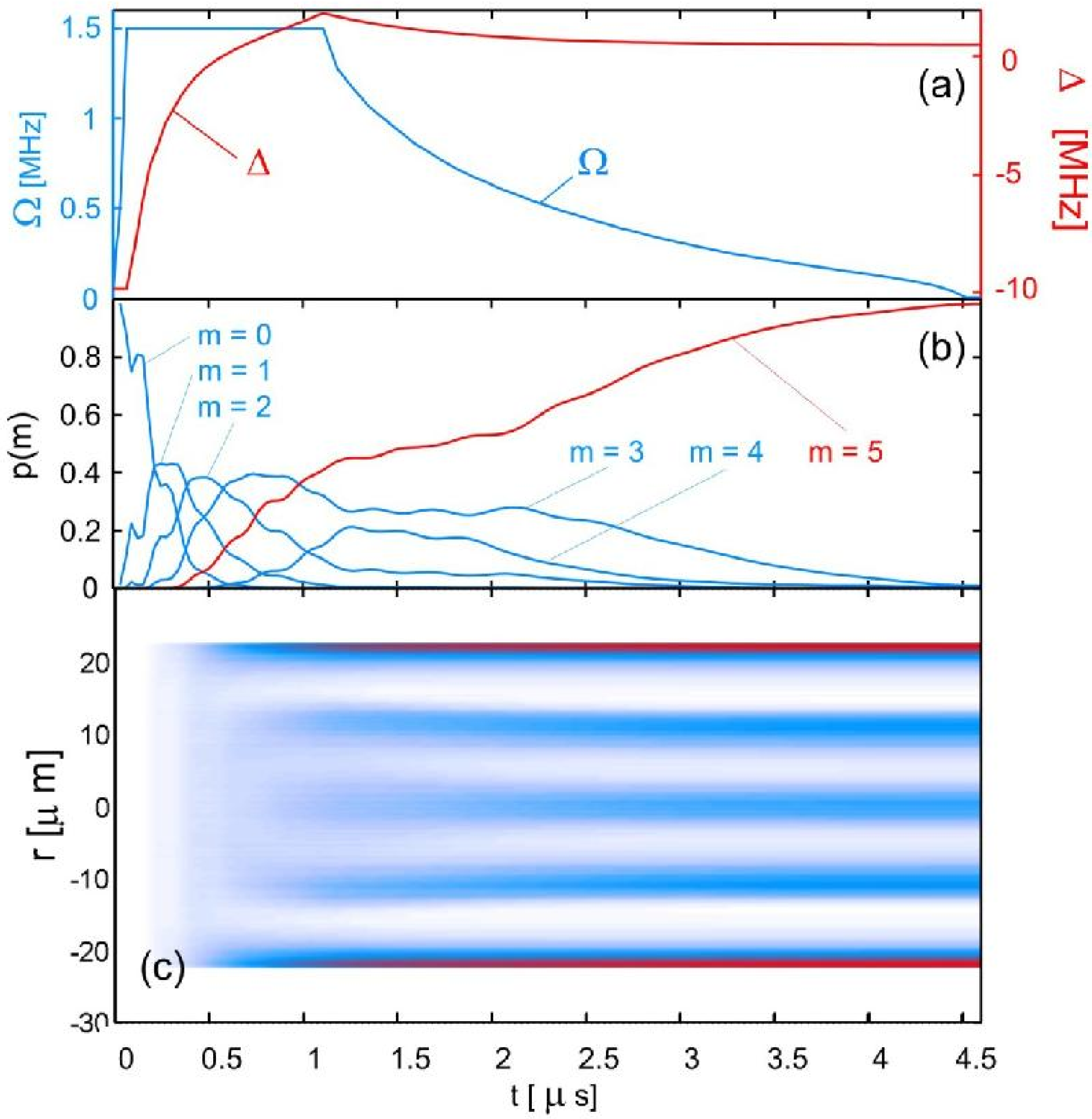
N exc.

N - 1 exc.

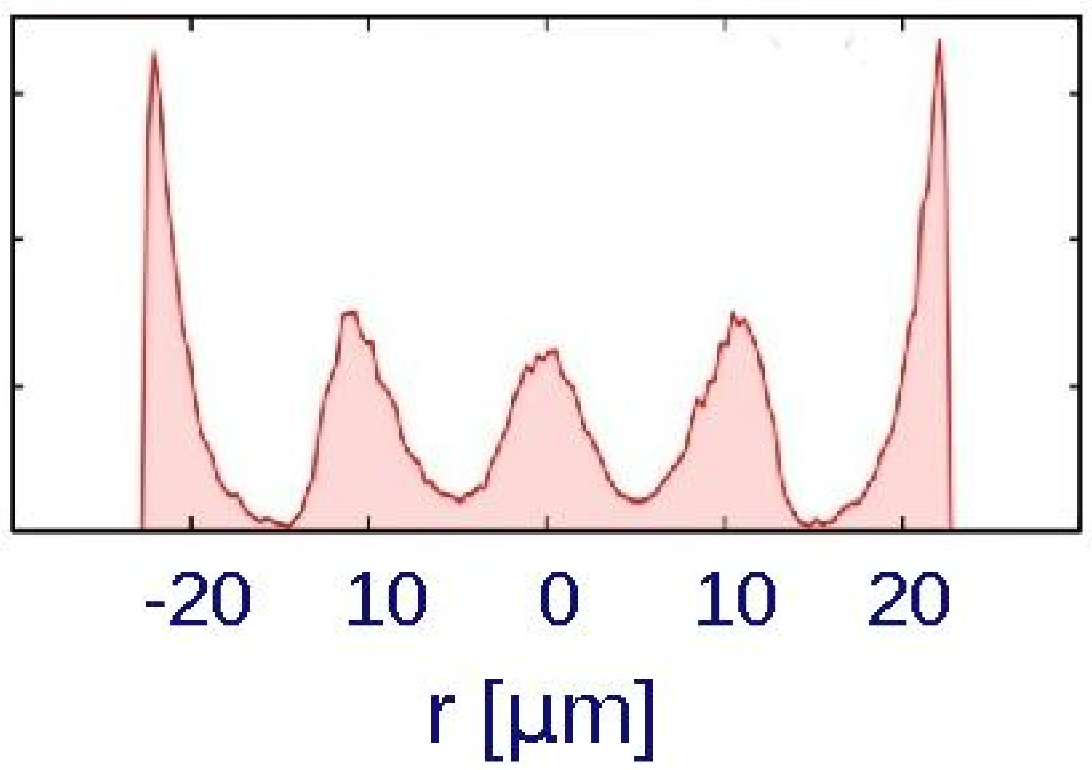
….. etc.

Results MOT

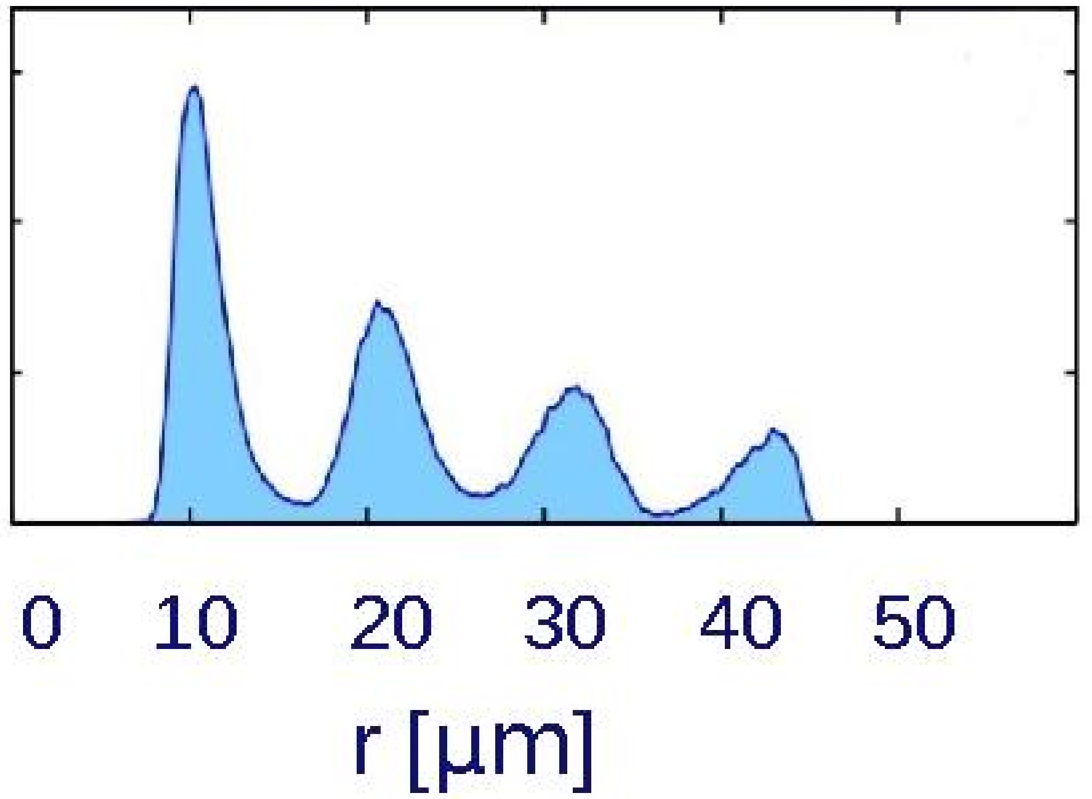
10^{11} cm^{-3}: ~30 particles in exc. vol.



exc. dens

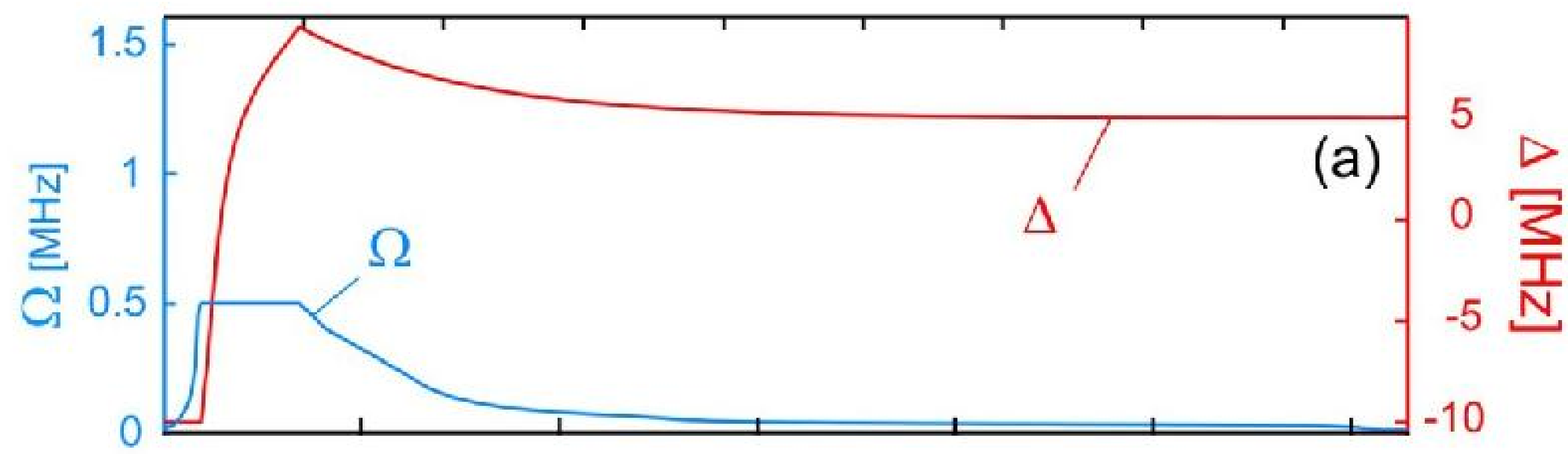


correlation function

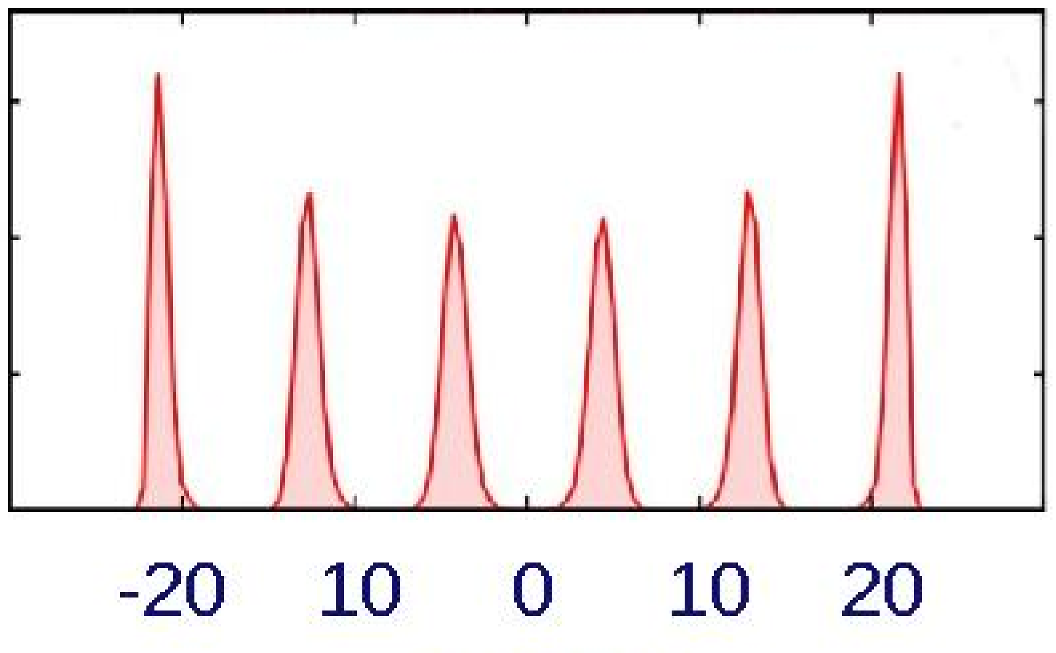


Results BEC

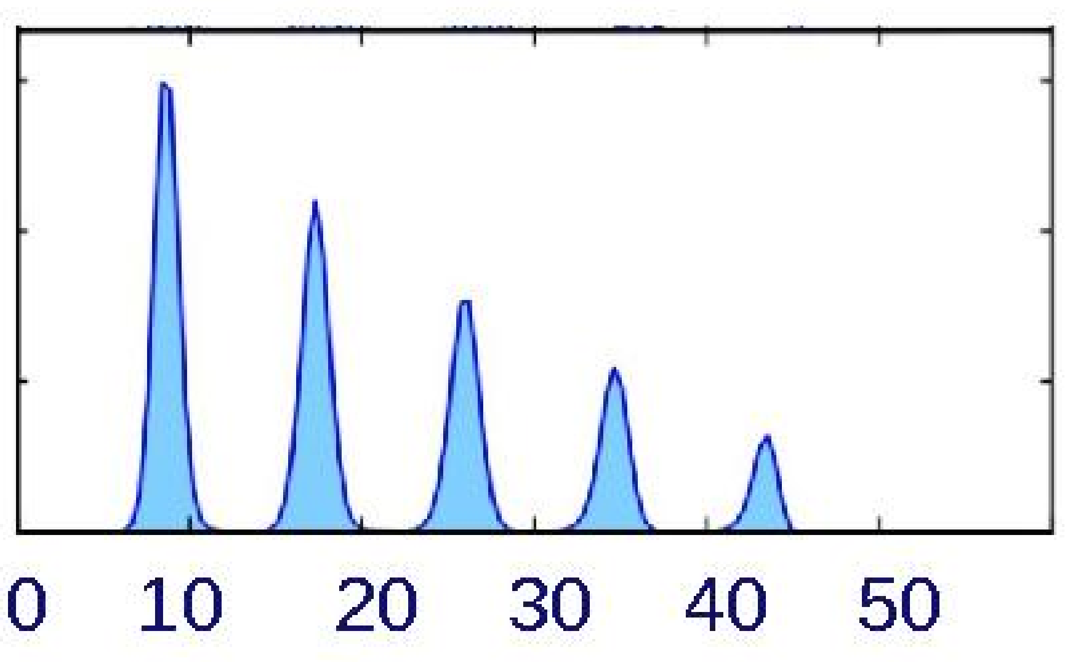
10^{14} cm^{-3}: ~40.000 particles in exc. vol.



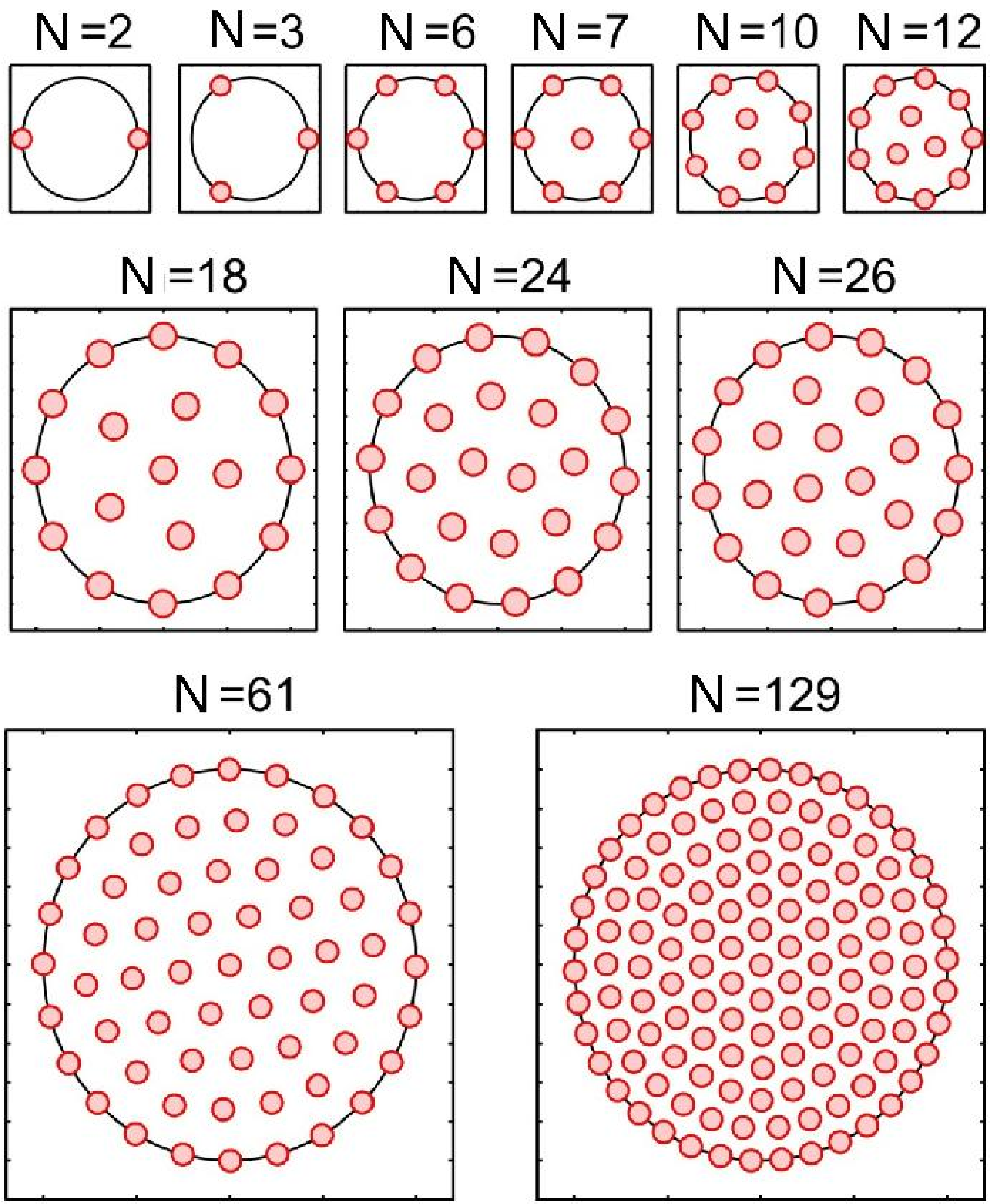
exc. dens



correlation function



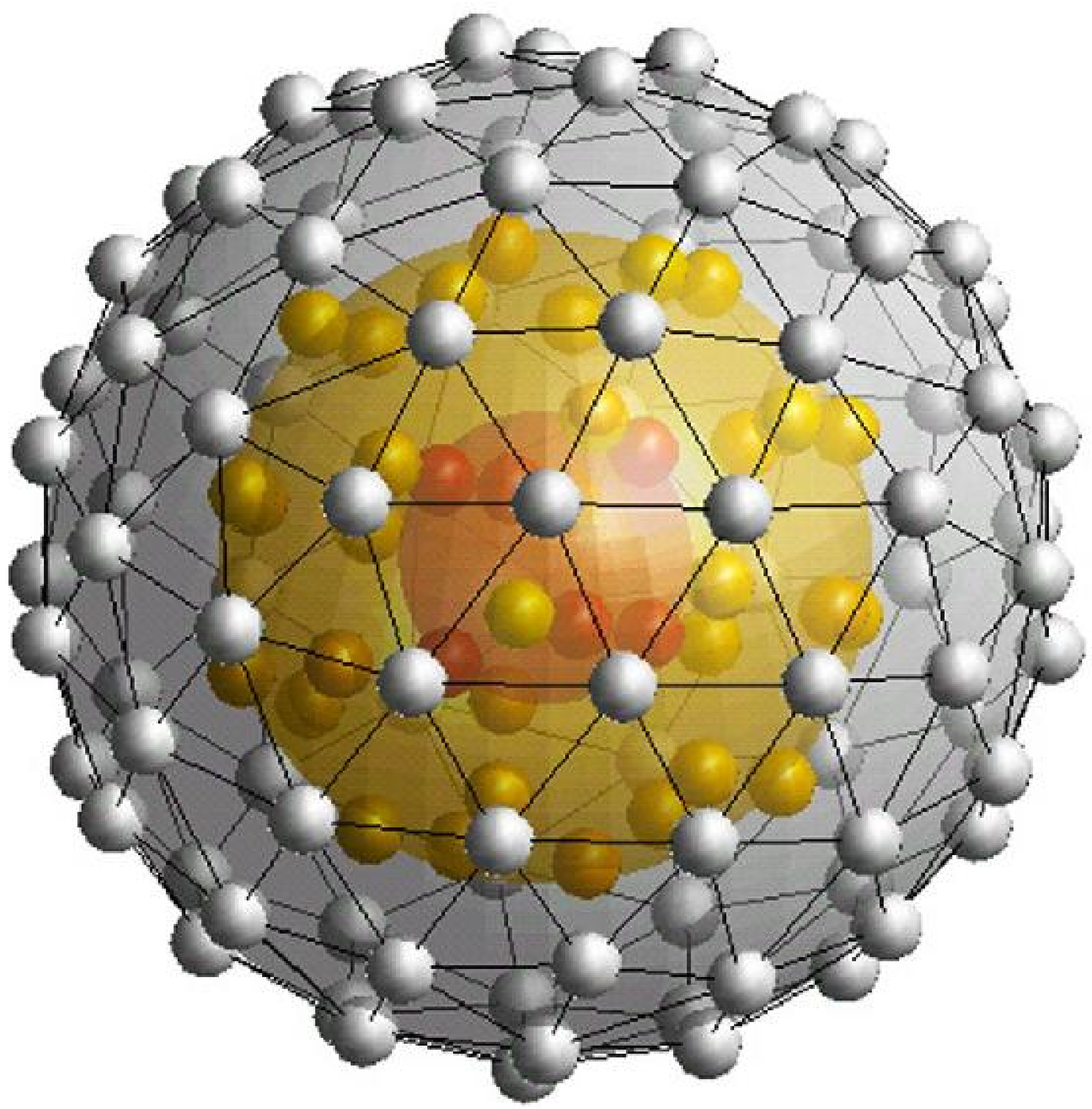
2D Rydberg crystals

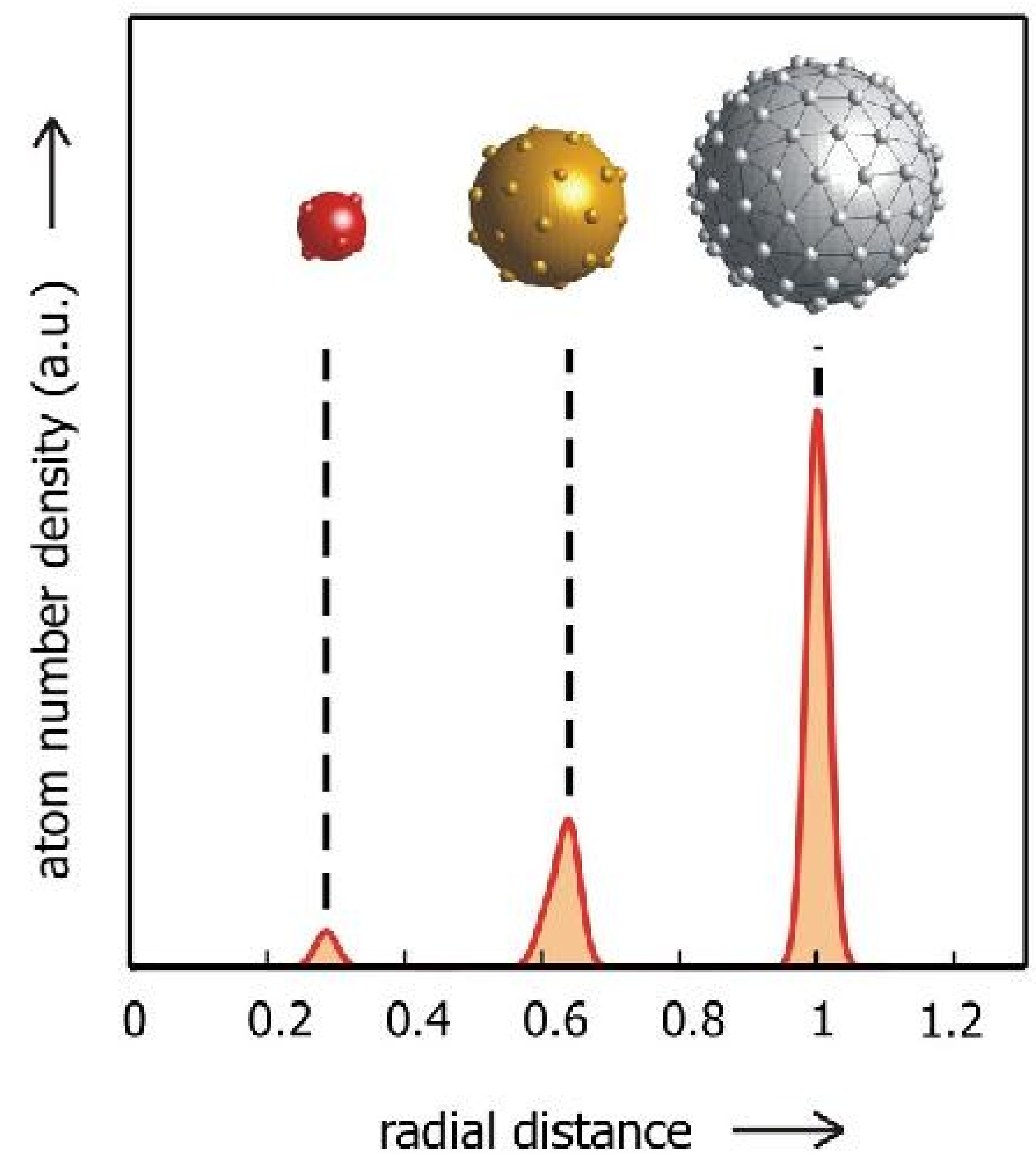


More complex (3D) structures

Experimentally challenging

- Shell structures appear

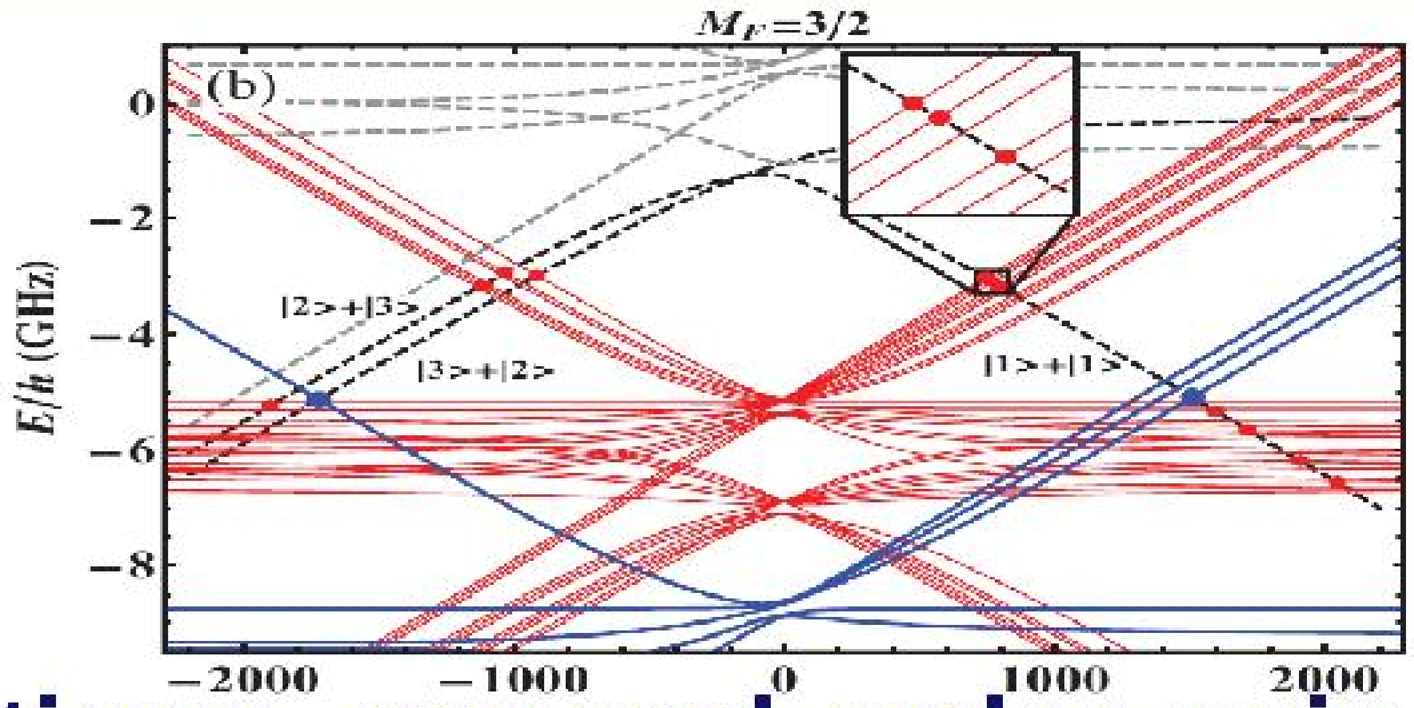




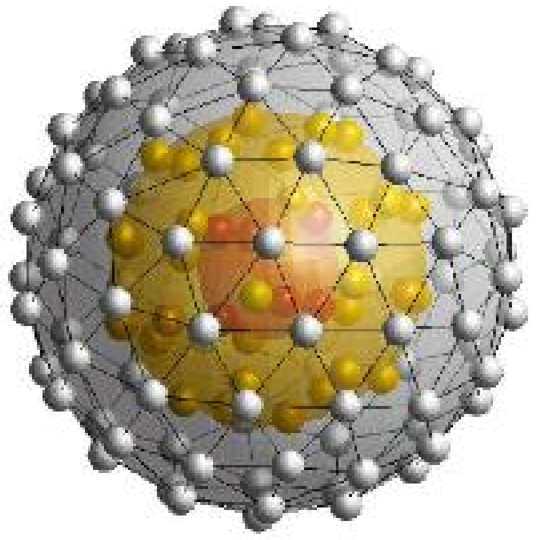
Shell structures also observed in 9-Be^{+} ion clouds [T.B. Mitchell et al, Phys. Plasmas **6**, 1751 (1999).]

Summary

- **Asymptotic Bound-state Model**
  - Initially a two-parameter model
  - complexity controllable:
    magnetic dipole-dipole interactions, second order spin-orbit coupling, higher partial waves, more bound states
  - easy to use (& fast!)
  - successful for several "unexplored" systems, mainly mixtures as Li-K, Li-Na, etc
  - Feshbach resonances can be systematically assigned

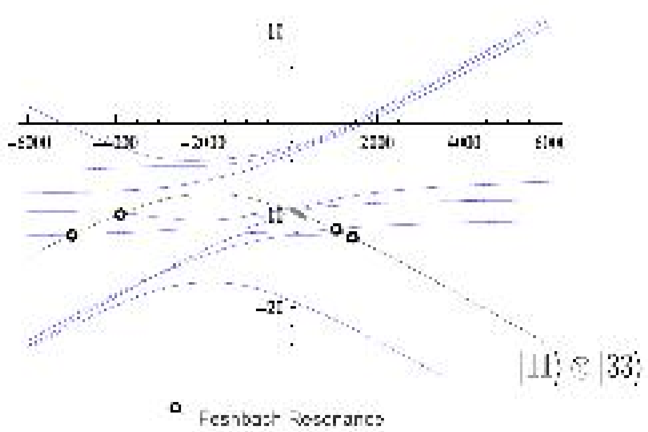


- **Rydberg crystals**
  - Should be possible in current experimental conditions
  - Adiabatic crystal formation

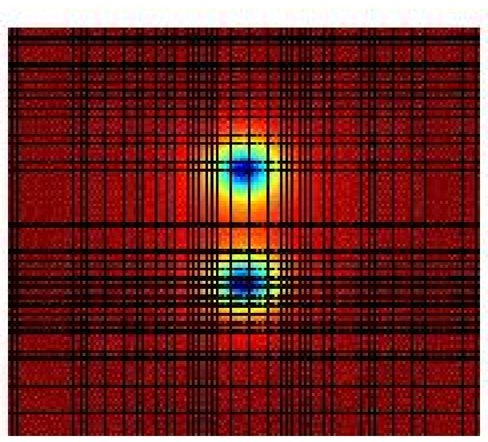


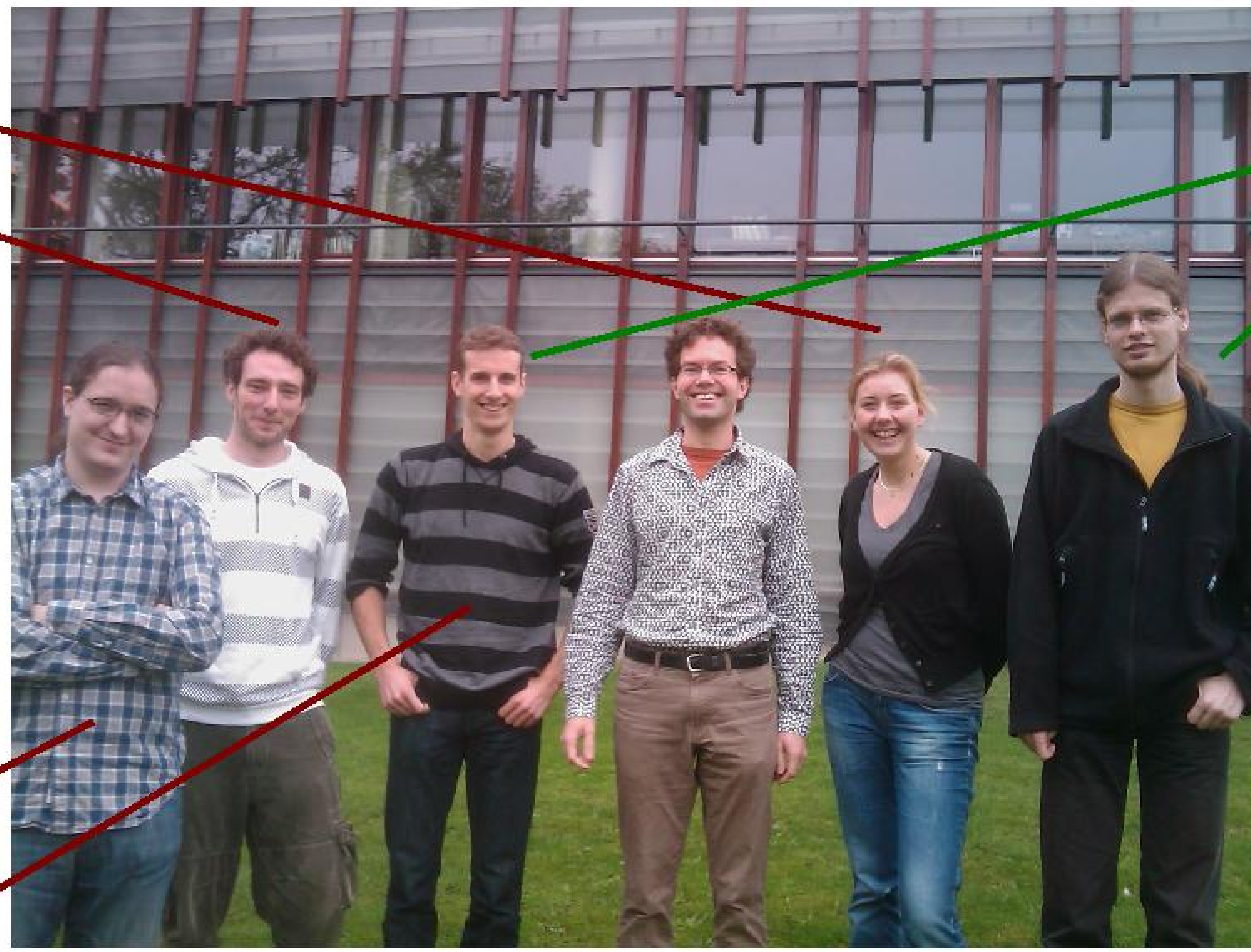
Acknowledgments

Feshbach res.
Marije Haverhals
Maikel Goosen



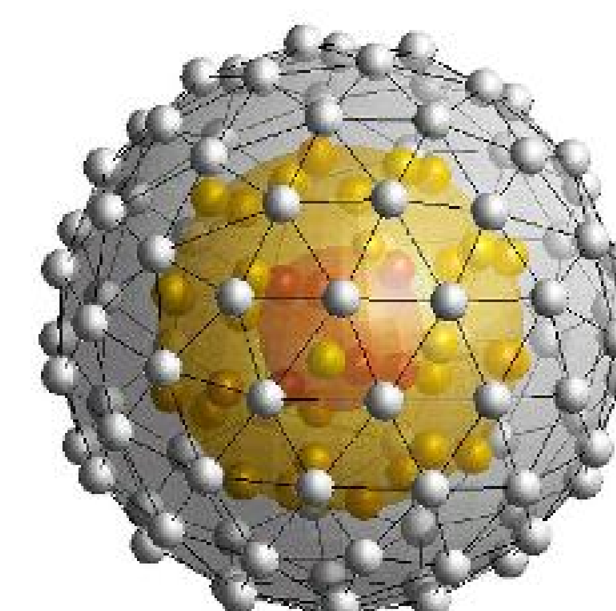
Dipolar BEC
Elmer doggen
Rick van Bijnen





Rydberg lattices
Rick van Bijnen
Sjoerd Smit

Not in picture:
Chris Bradly
Gerwin Dijk
Cornee Ravensbergen



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