Magnetic-field Kondo-spectroscopy of single molecule magnets

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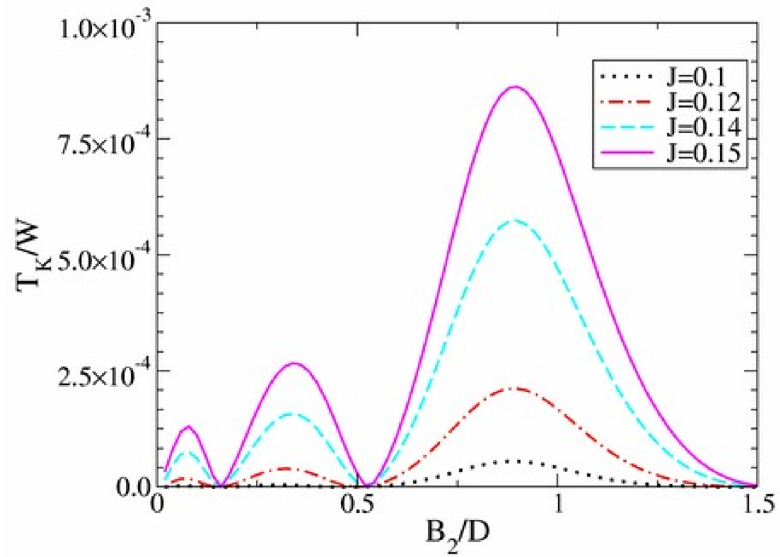
European Commision Research Training Networks Spintronics

Zero-field: Phys. Rev. Lett. 96, 196601 (2006)
Magnetic field: cond-mat/0605514.

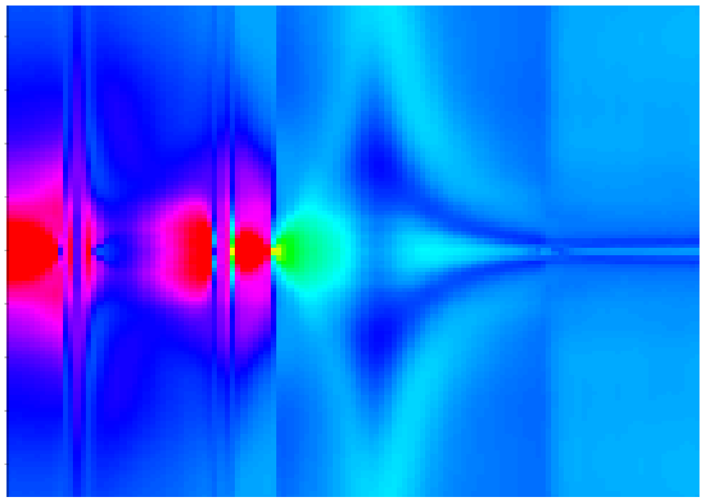
- **Single molecule magnets (SMM)**
  - Magnetic anisotropy:

    barrier + spin-tunneling (QTM)
- **QTM-induced Kondo effect**
  - “Weak” Kondo effect
  - “Strong” Kondo effect



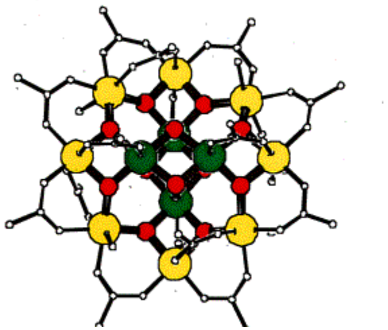
- **Magnetic field controlled Kondo effect**

Kondo effect
~ mixing of magnetic states *M*
induced by QTM
controlled by magnetic field

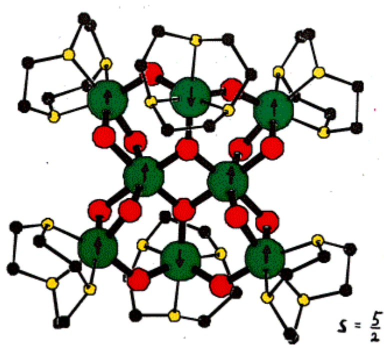


Single molecule magnets (SMM)

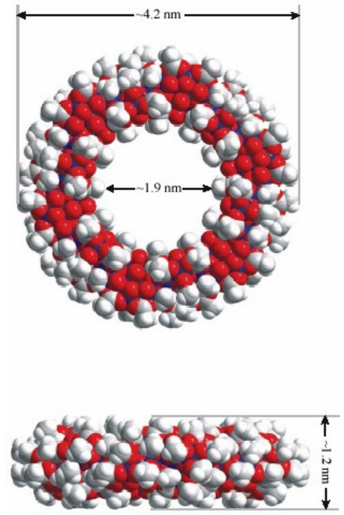
Mn12, *S*=10



Fe8, *S*=10



Mn84, *S*=6

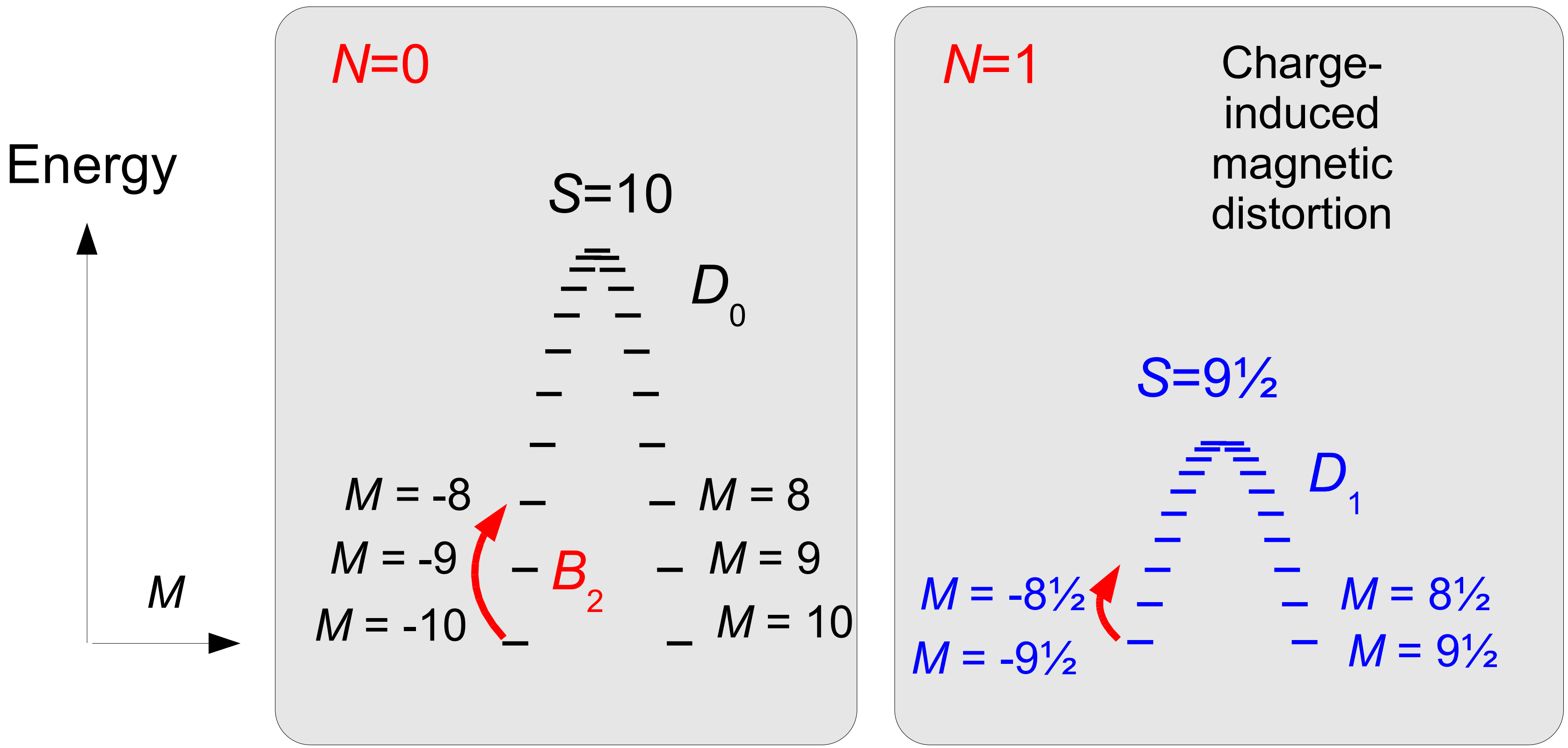


8 (*S*=2) – 4 (*S*=3/2) = 10

Sessoli et. al. J. Am. Chem. Soc 115, 1804 (1993)
Thomas et. al. Nature 383, 145 (1996)
Friedman et al. Phys. Rev. Lett. 76, 3830 (1996)

- Big, finite spin S >> 1/2 ~ intramolecular exchange
- Magnetic anisotropy ~ intramolecular spin-orbit coupling
- Discrete magnetic symmetry ~ geometry of magn. core

Magnetic anisotropy



$$H_{MAB} = -D_N S_z^2$$ Magnetic anisotropy barrier

$$H_{QTM} = -\frac{1}{2} B_2 (S_+^2 + S_-^2)$$ Quantum tunneling of magnetization (**QTM**)

Symmetry: 2-fold rotation axis

$$B_2 / D_N < 1$$

$D \sim 5.10^{-2}$ meV

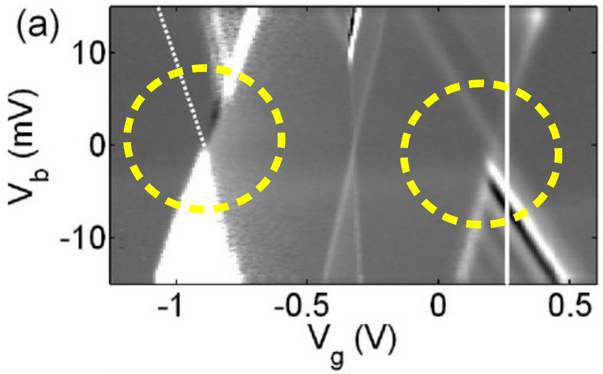
Experiment

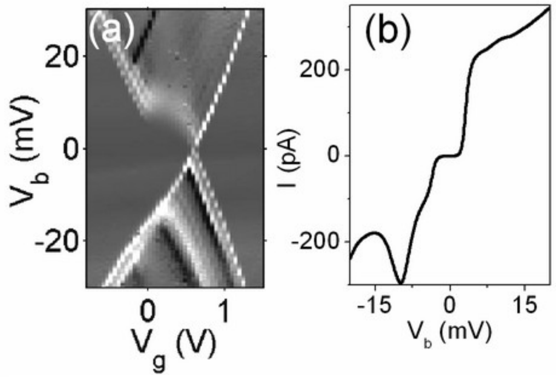
H. van der Zant *Delft*
A. Cornia *Modena*

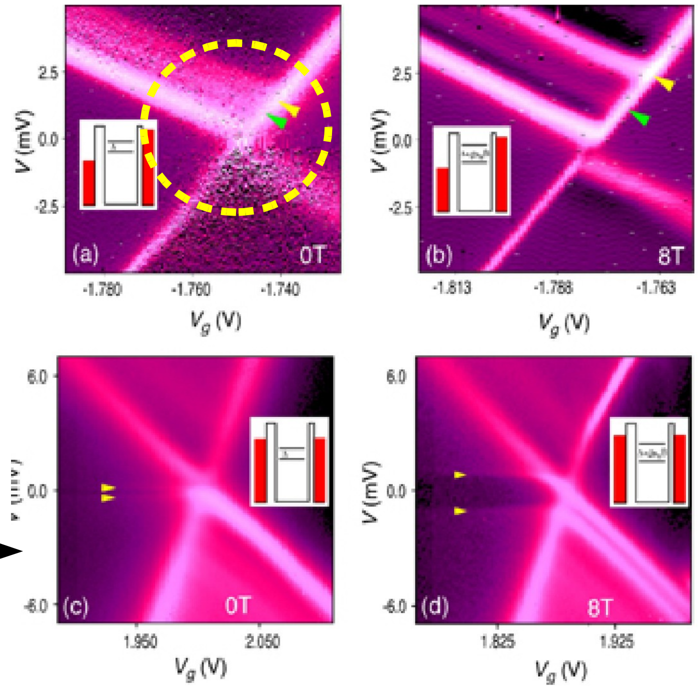
Phys. Rev. Lett. 96, 206801 (2006)

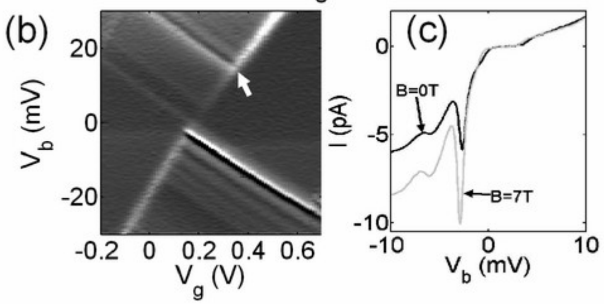
M.-H. Jo, H. Park, J. Long
Harvard

cond-mat/0603276









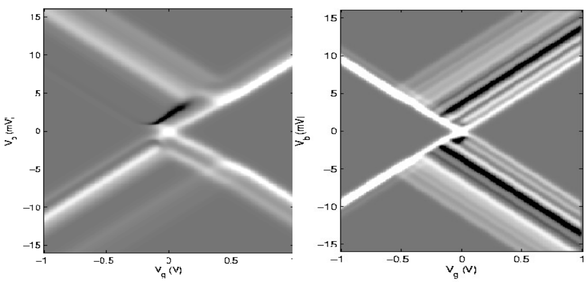
Mn12 with
different ligands

Theory

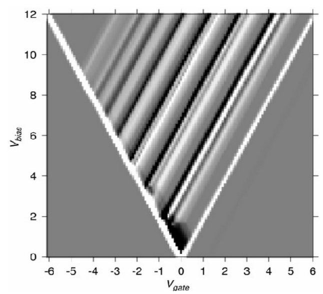
C. Romeike, M. R. Wegewijs, H. Schoeller
Aachen

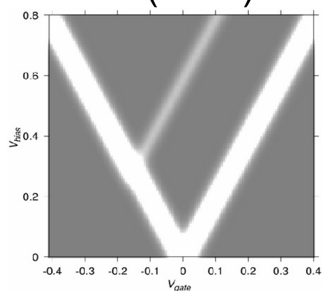
Phys. Rev. Lett. 96, 196805 (2006).

Kim & Kim PRL04
Elastic cotunneling
& QTM



Magnetic-blockade
& full suppresiion
Seveal spin multiplets
per charge state





Yes: QTM visible!

No: ~ 1 out of 100
transitions visible....

Charge-dependent
magnetic parameters?

F. Elste, C. Timm
Berlin

Magnetic read/write
cond-mat/0601294

Spin-amplification
cond-mat/0511291
cond-mat/0410641

S= ½ Kondo effect

S =molecular spin

$$s=\sum_{kk'\sigma\sigma'} a^{\dagger}_{k\sigma}\sigma_{\sigma,\sigma'}a_{k\sigma}/2$$ **= reservoir spin**

$$H_K = J\,\mathbf{S}\cdot\mathbf{s} + \sum_{k\sigma}\epsilon_k a^{\dagger}_{k\sigma}a_{k\sigma}$$

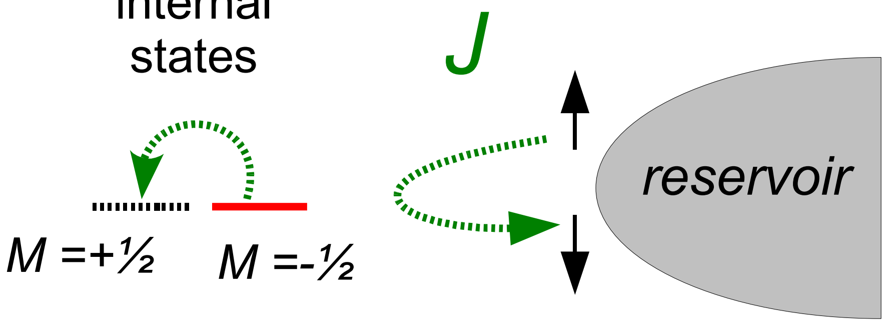
AF exchange scattering — reservoir

$$\rho J \sim \frac{\Gamma}{E_{add}(V_g)}$$ STM / 3-terminal

$$W = 1/\rho$$ bandwidth

degenerate internal states

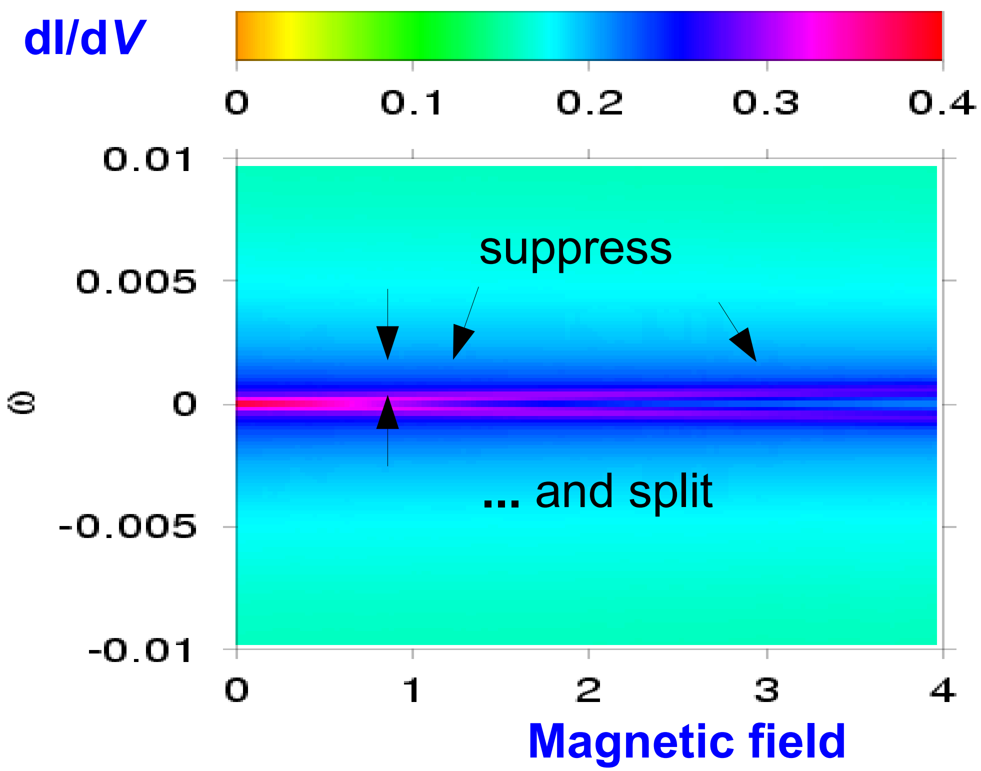
S=1/2



Spin-binding energy ~ T_K << J

Numerical Renormalization Group (NRG)

Kondo dI/dV-peak Width ~ T_K >>T



S > ½ Kondo effect *with anisotropy*

$H_{MAB} = -DS_z^2$

suppresses
underscreened Kondo

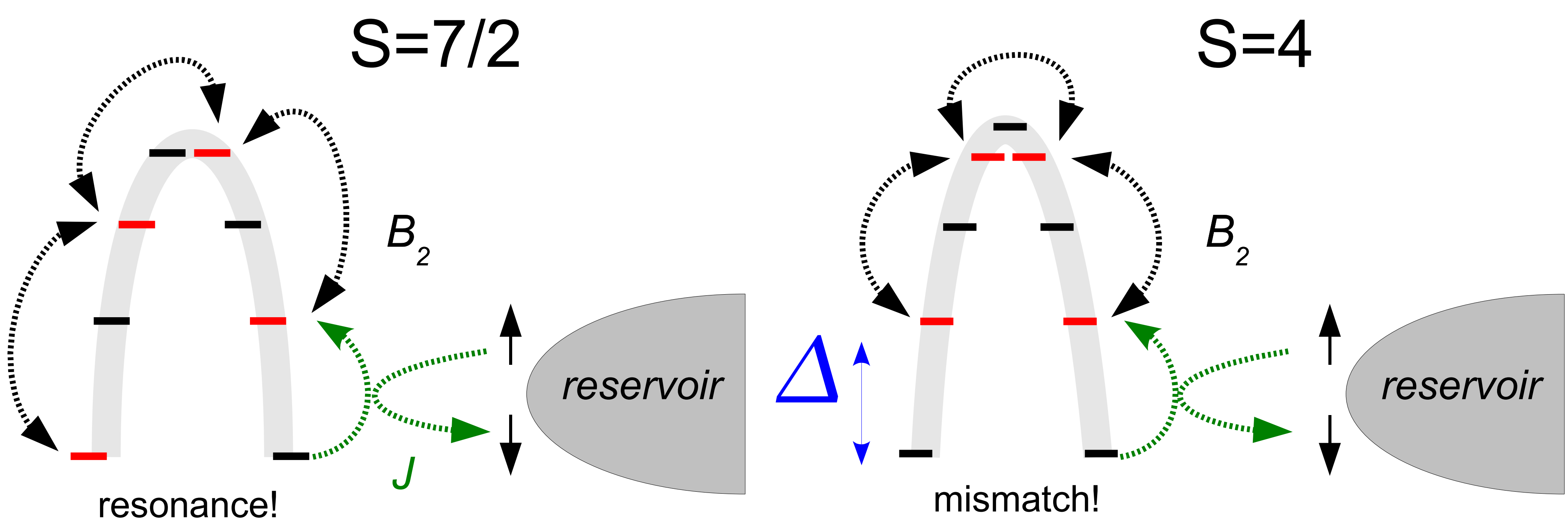
$H_{QTM} = -\frac{1}{2} B_2 (S_+^2 + S_-^2)$

assists spin reversal
pseudo-spin Kondo

$H_K = J\,\mathbf{S}\cdot\mathbf{s} + \sum_{k\sigma} \epsilon_k a_{k\sigma}^\dagger a_{k\sigma}$

completes
spin reversal

reservoir



“Weak” vs “Strong” Kondo coupling
$T_K \gtrless \Delta \sim 2SD \sim 1$ meV (magnetic scale)

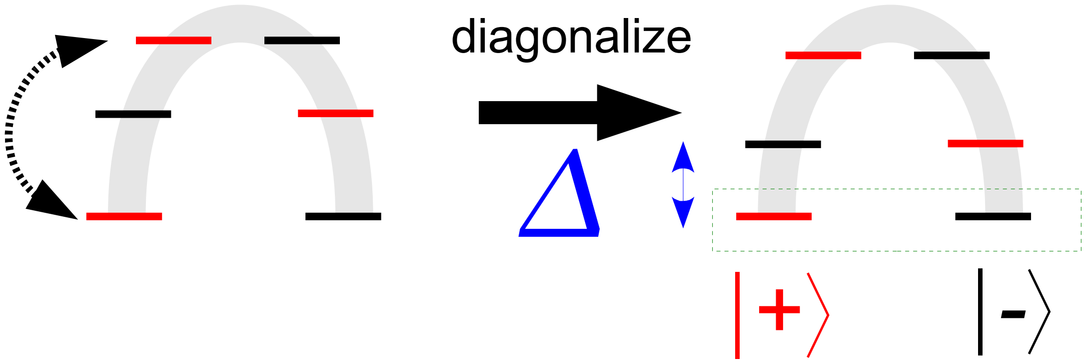
$B_2 << D$

Weak Kondo effect in SMMs

Phys. Rev. Lett. 96, 196601 (2006).

$$T_K(J,\Delta) \ll \Delta$$

$$H_K = \sum_{i=xyz} J^i \boldsymbol{P}^i \cdot \boldsymbol{s}^i + \sum_{k\sigma} \epsilon_k a^\dagger_{k\sigma} a_{k\sigma}$$



truncate: pseudo-spin-½ ***P***

$$J^z = 2J\langle + | S_z | + \rangle$$

$$J^{x,y} = J\langle + | S_+ \pm S_- | - \rangle$$

Cut-off:

$$W_{eff} \sim \Delta(B_2, D)$$

$$\frac{dJ^\alpha}{d\ln W} = -\rho J^\beta J^\gamma$$

$$\alpha, \beta, \gamma \in \{x, y, z\}$$

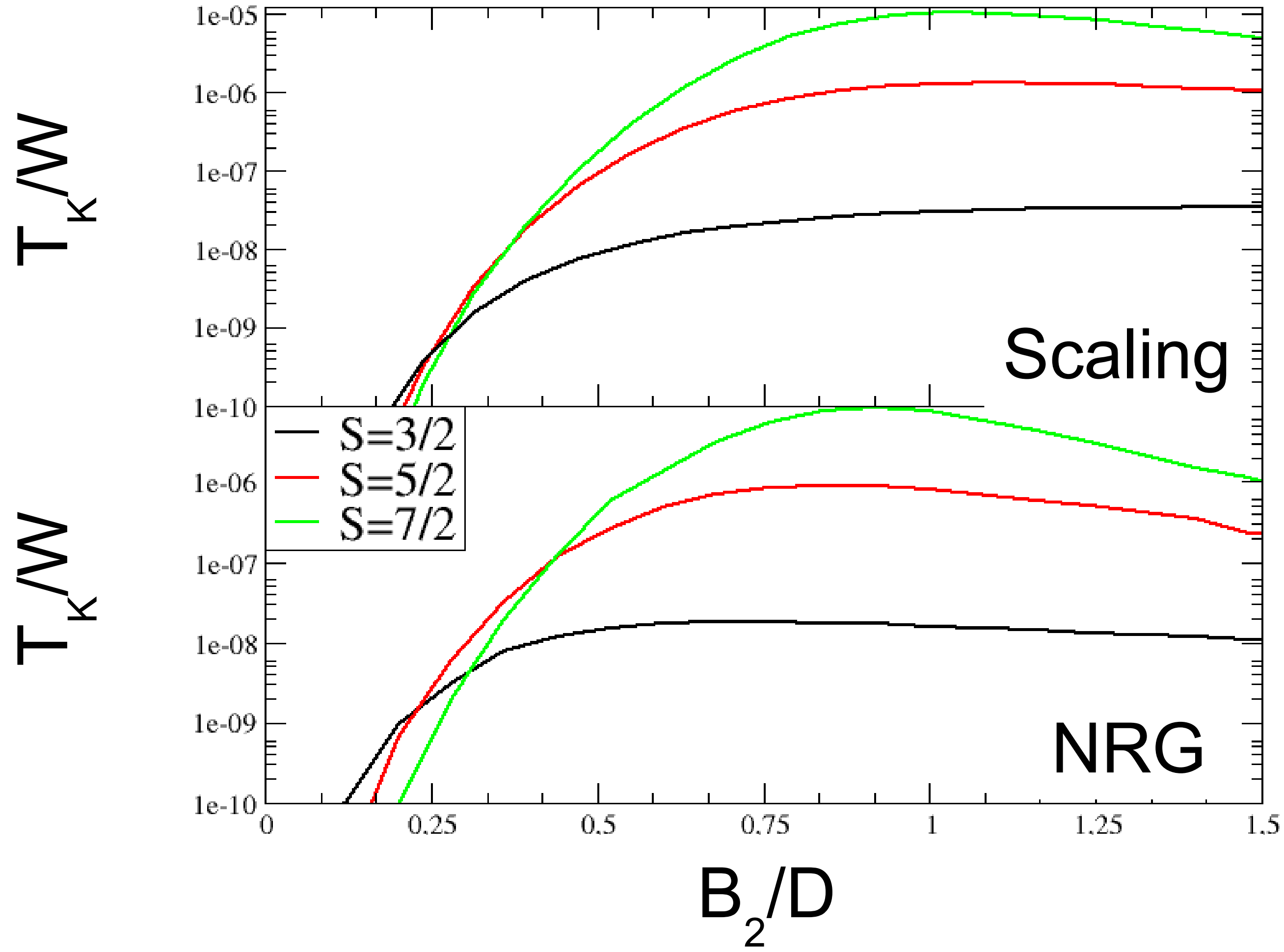
Poor-man scaling:
fully anisotropic exchange

QTM modulates magnetic *state mixing*

Modifies exchange: *strength* & *anisotropy*

$$J^{x,y,z}(B_2/D)$$

T_K tuned by QTM



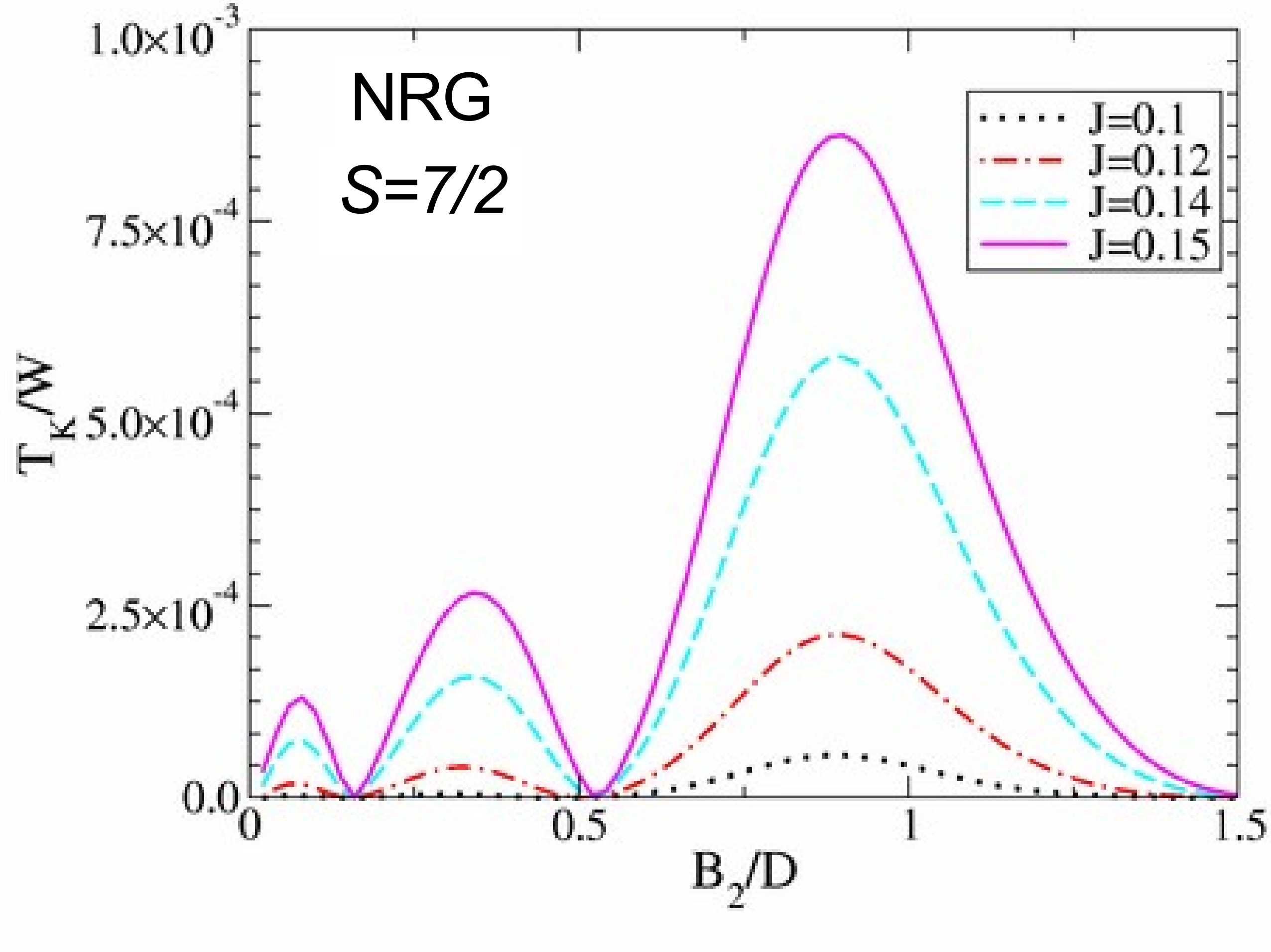
truncation

Full model
no truncation

Parameters: *W=1, D=0.005W, J=0.1W*

Strong Kondo effect in SMMs

C. Romeike, M. Wegewijs, H. Schoeller, W. Hofstetter, cond-mat/0605514.

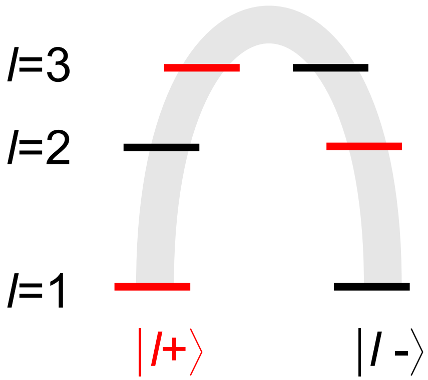


S -1/2 peaks & dips
*intra*molecular origin

$$T_K(J,\Delta) \gg \Delta$$

Excited state spin scattering

S +1/2 Kramers doublets



$(S+1/2)^2$ pairs l, l'

Pseudo-spin vectors:

$(P^x_{ll'}, P^y_{ll'}, P^z_{ll'})$

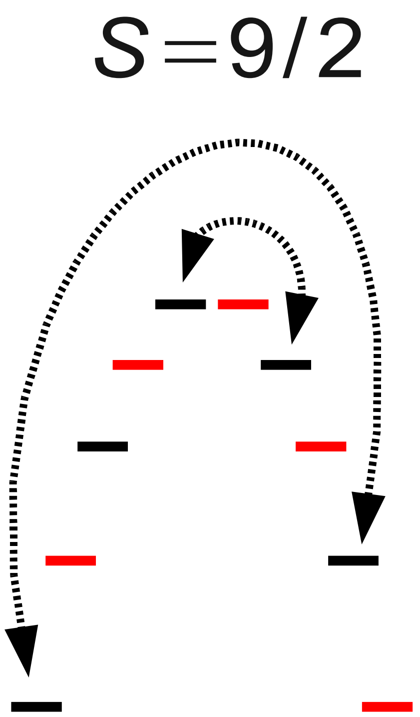
$$H_K = \sum_{l\,l'} \sum_{i=xyz} J^i_{ll'} P^i_{ll'} s^i + \sum_l E_l N_{ll} + \sum_{k\,\sigma} \epsilon_k a^\dagger_{k\sigma} a_{k\sigma}$$

$$J^{x,y}_{ll'} = J \left\langle +l \middle| S_+ \pm S_- \middle| -l' \right\rangle \qquad J^z_{ll'} = 2J \left\langle +l \middle| S_z \middle| +l' \right\rangle$$

anticrossing
many exchange couplings change

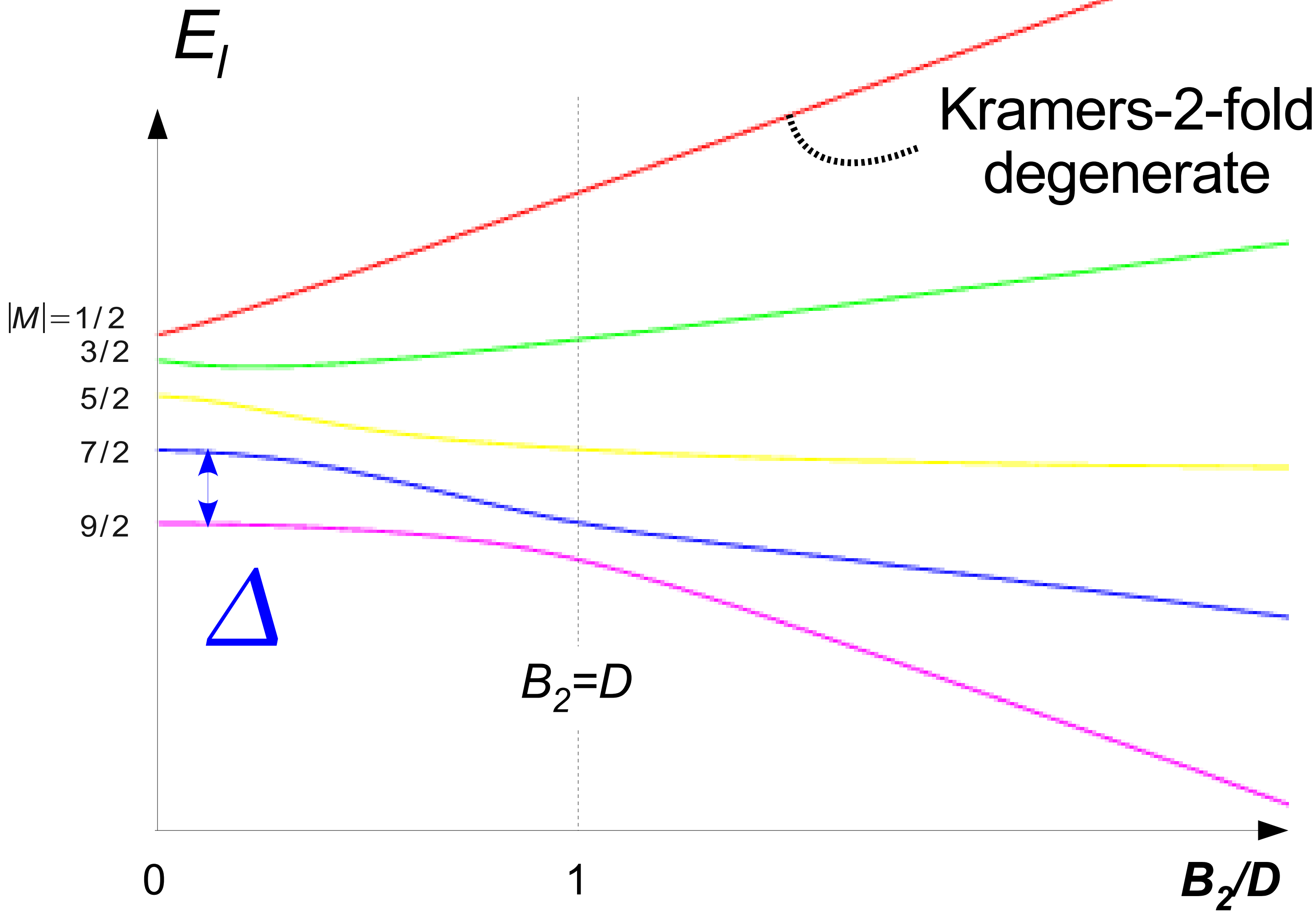
QTM induced anticrossings

Large M / low energy:
- Large initial splitting
- Coupled in high order of B_2



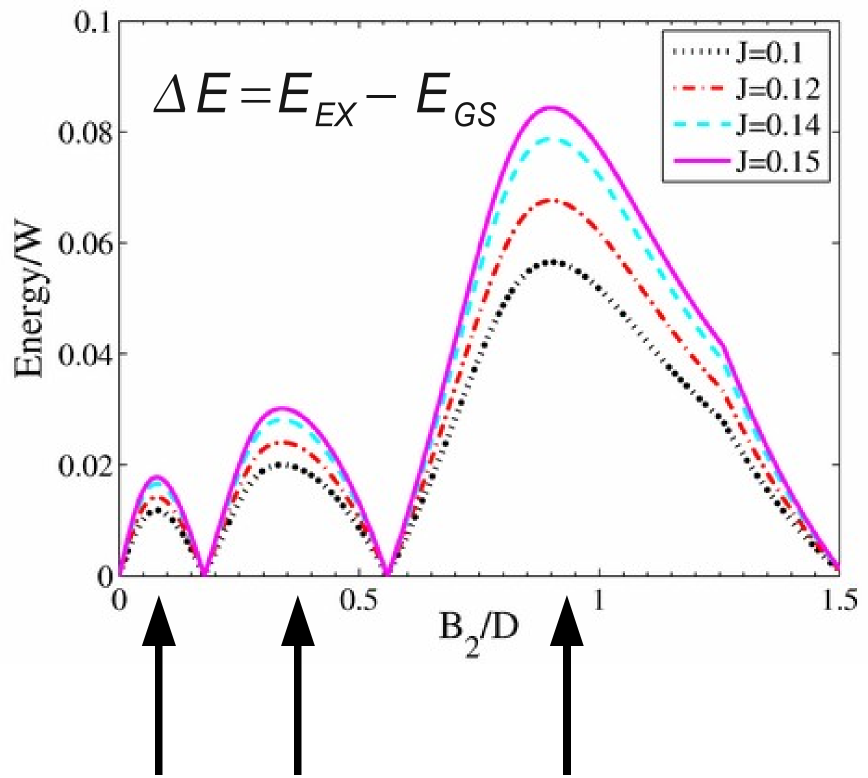
Progressive mixing of wavefunctions with *low M*

Molecular eigenenergies including QTM

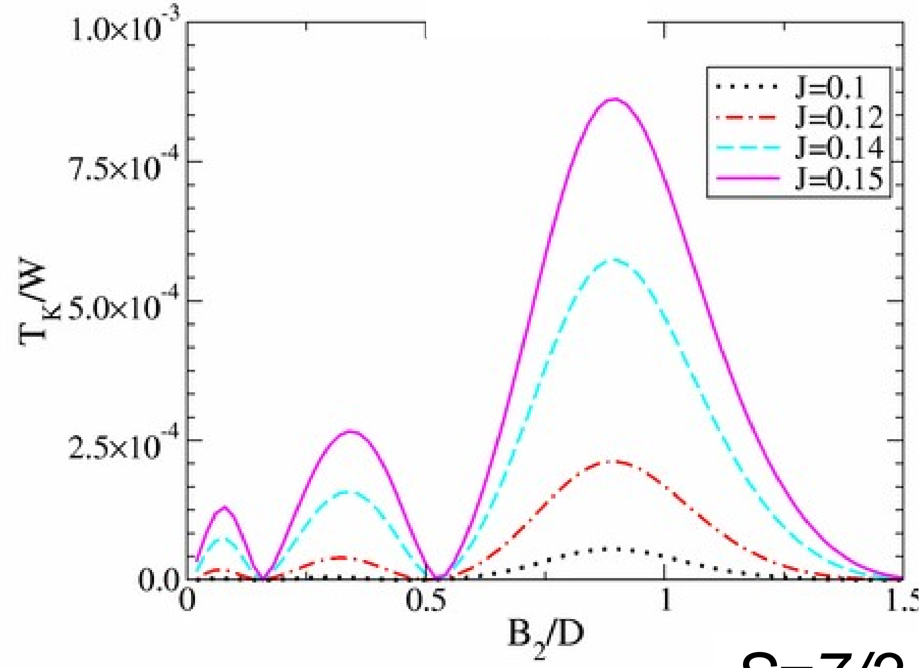


Single spin-binding energy

Zero-bandwidth model



NRG



S=7/2

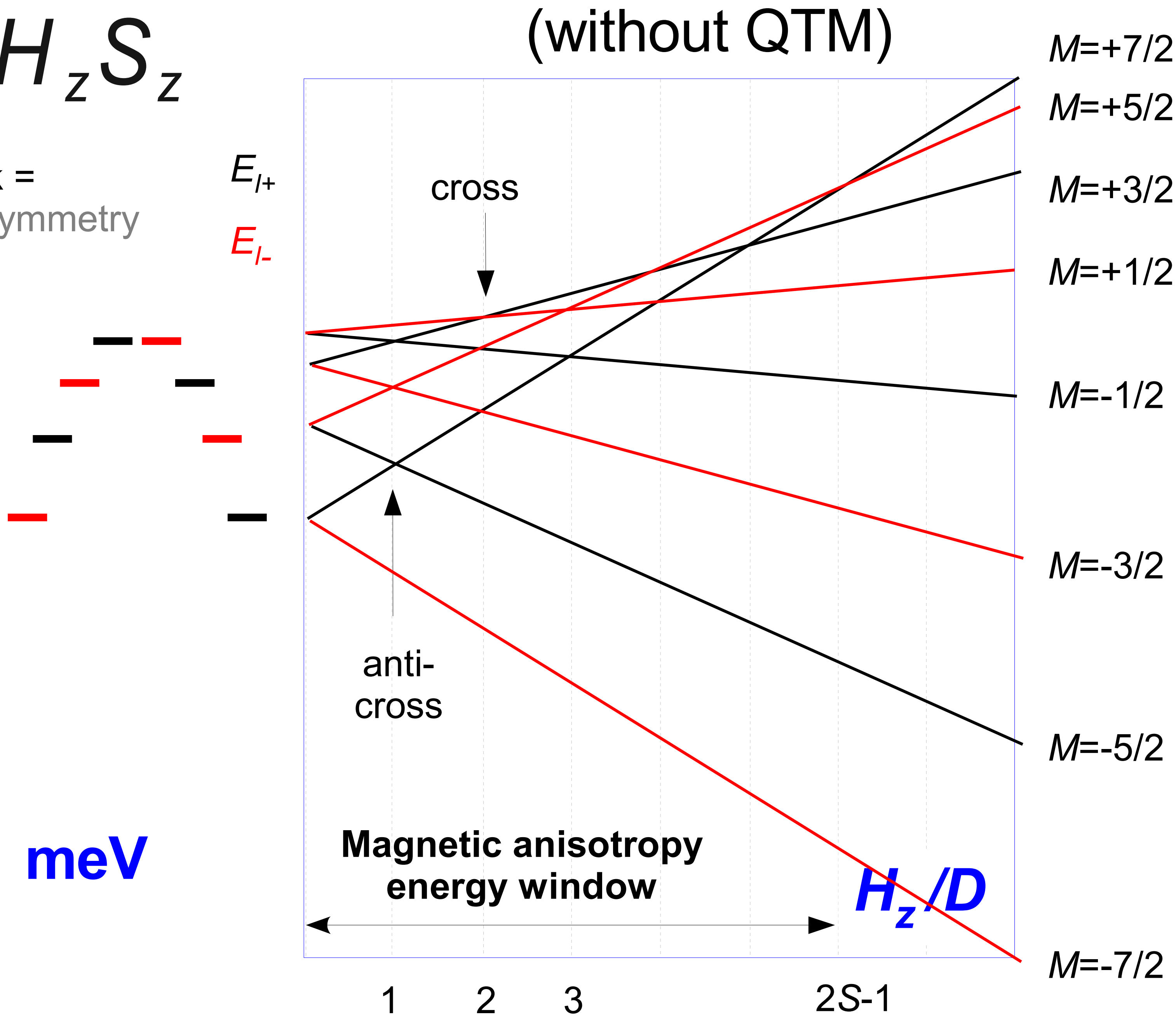
Different magnetic excitations of SMM involved

$T_K(J, \Delta) \gg \Delta$

Magnetic field induced anticrossings

$$H_{Zeeman} = H_z S_z$$

Red / black = magnetic symmetry



2SD ~ 1 meV

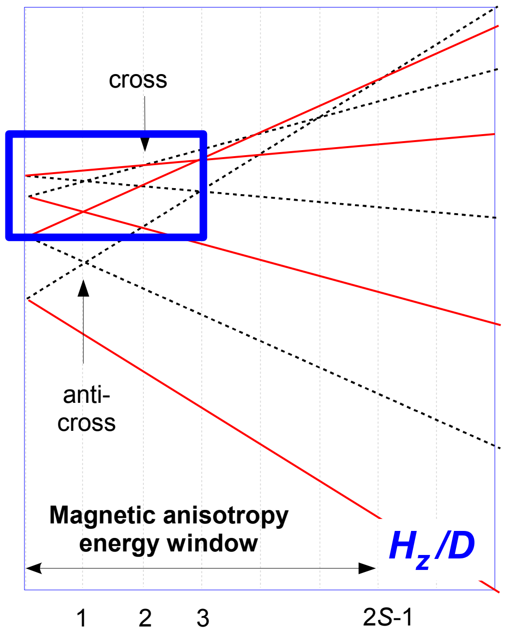
Magnetic field induced anticrossings

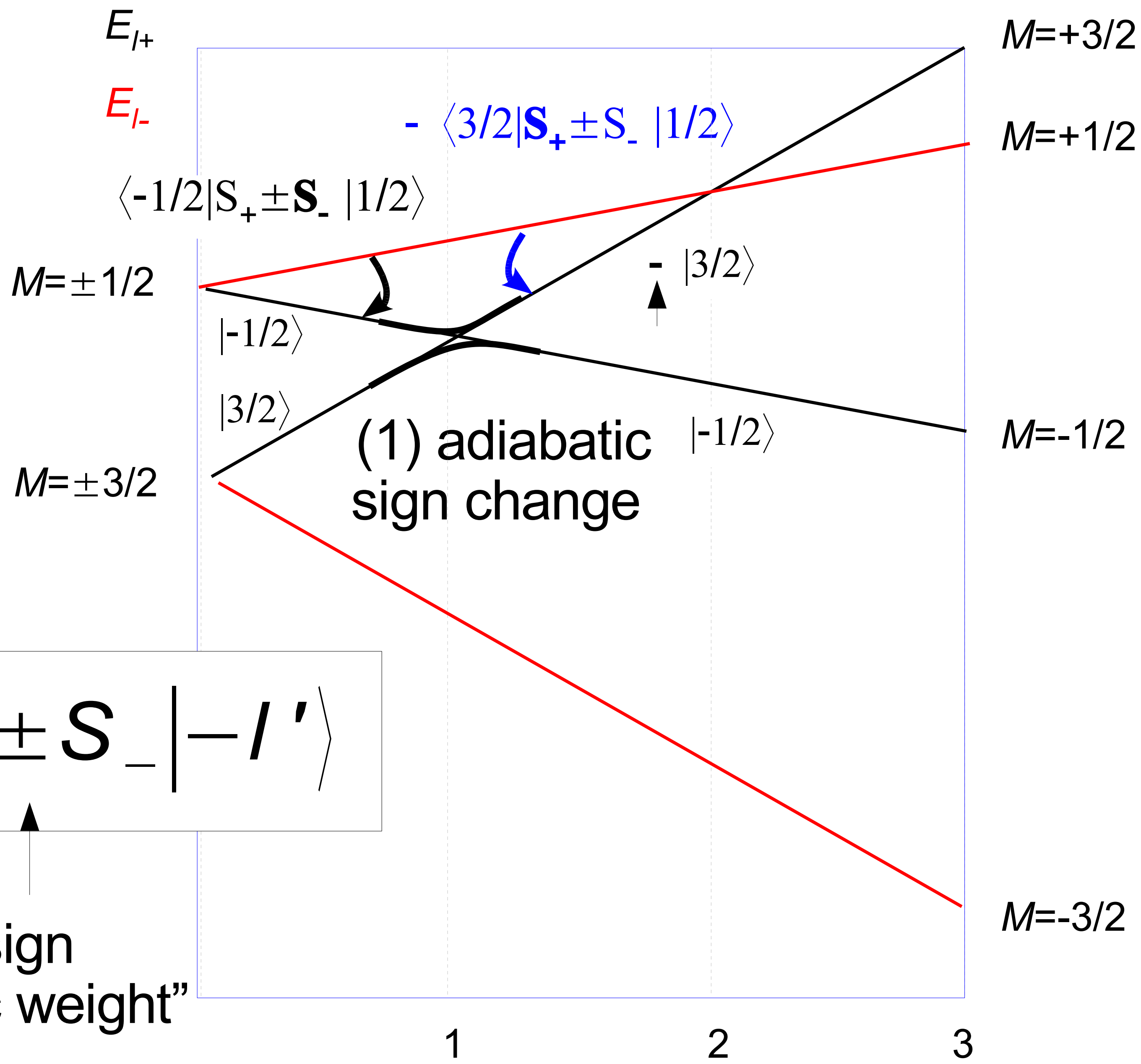
$$H_{ex} = \sum_{ll'} \left(\sum_{i=x,y} J^i_{ll'} P^i_{ll'} s_i + \sum_{\sigma=\pm} J^z_{ll'\sigma} \left| l\,\sigma \right\rangle \left\langle l'\,\sigma \right| s_z \right)$$

↑

Transverse scattering with anticrossing states
quenched at anticrossing

Anticrossing: 2 sign changes

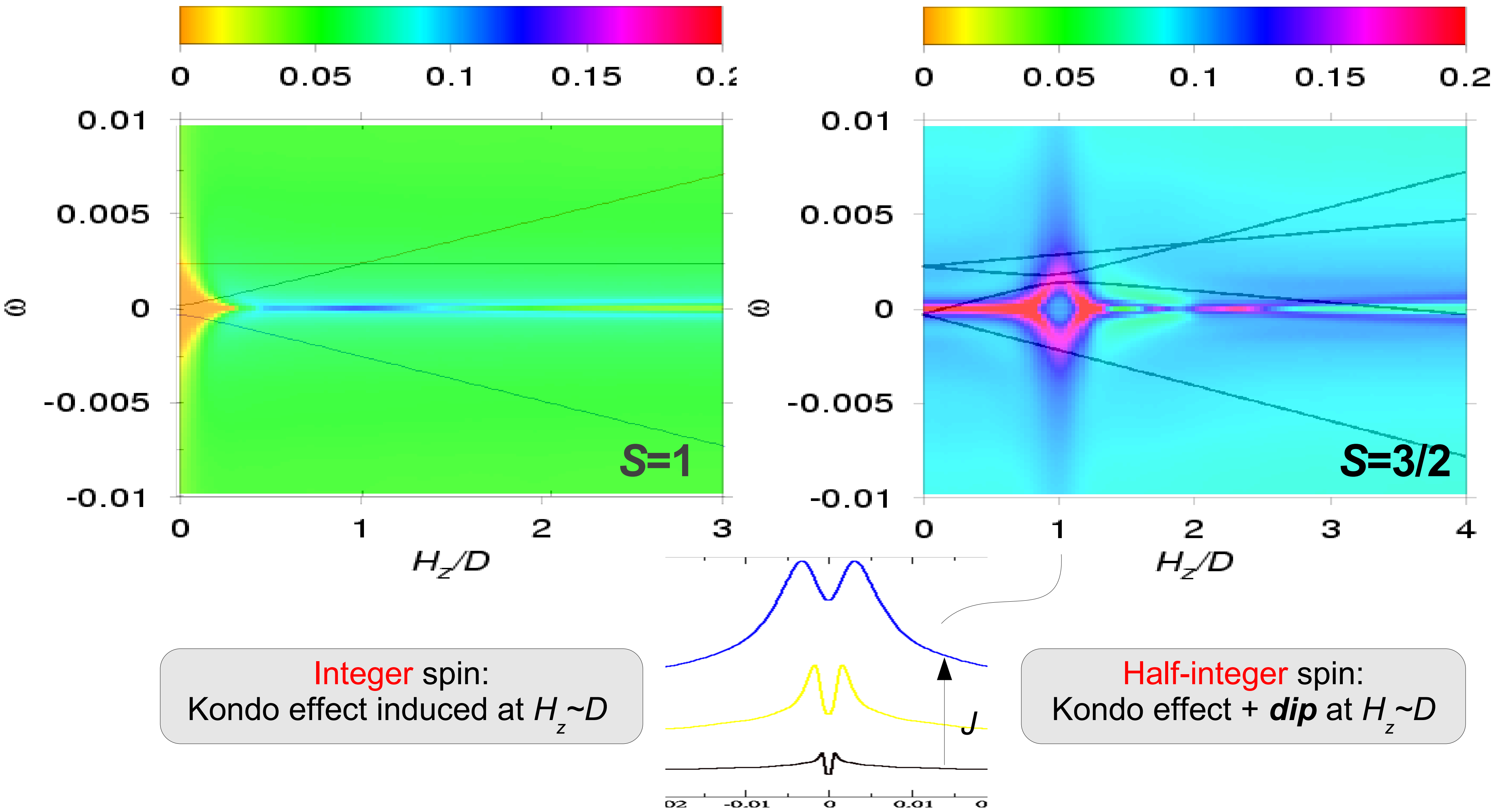




$$J_{l\,l'}^{x,y} = J\left\langle +l \middle| S_+ \pm S_- \middle| -l' \right\rangle$$

(2) sign
“magnetic weight”

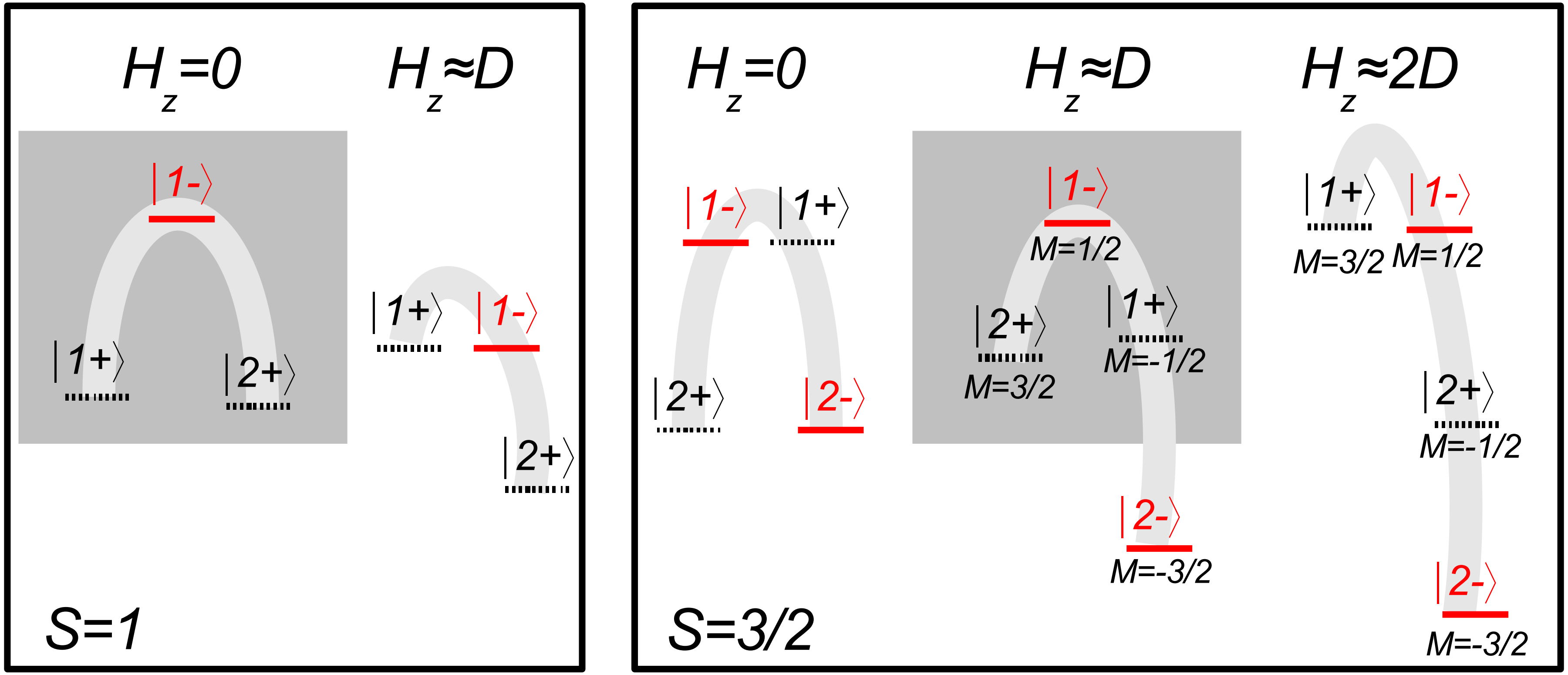
Field induced/modulated Kondo effect



Integer spin:
Kondo effect induced at $H_z \sim D$

Half-integer spin:
Kondo effect + ***dip*** at $H_z \sim D$

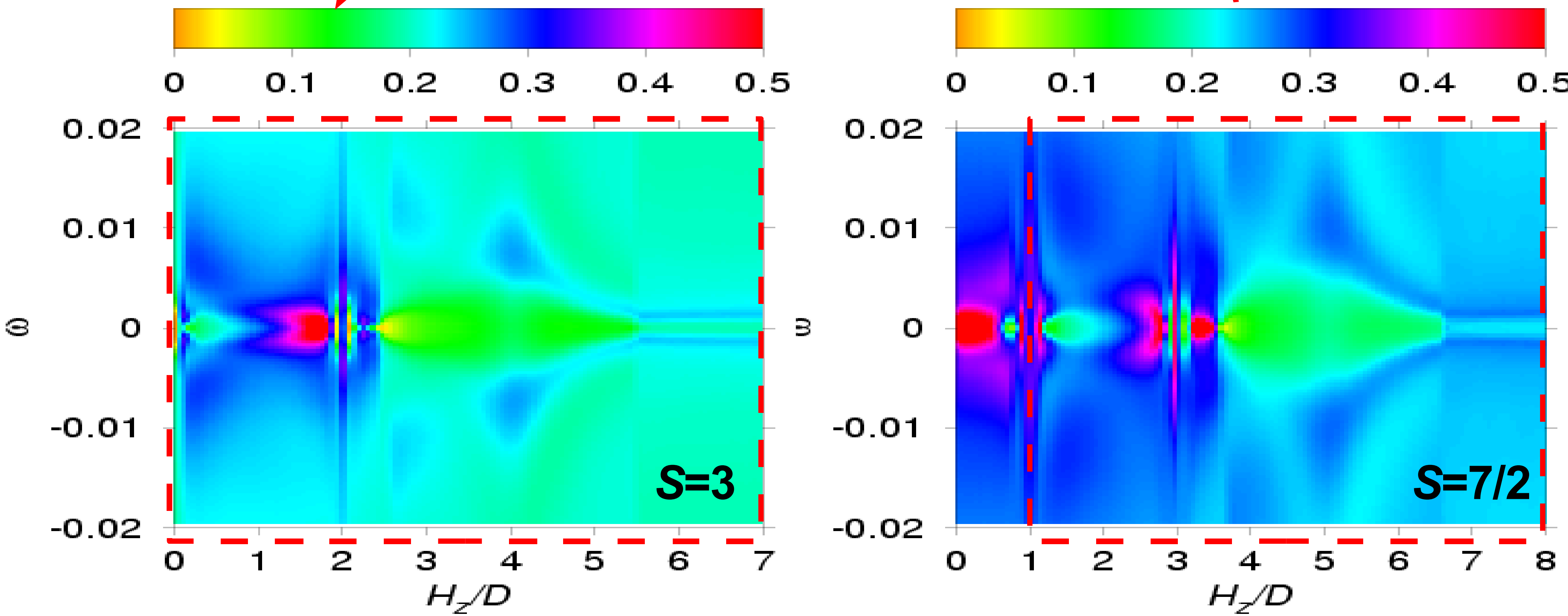
Relation integer / half-integer spin



Relation integer / half-integer spin

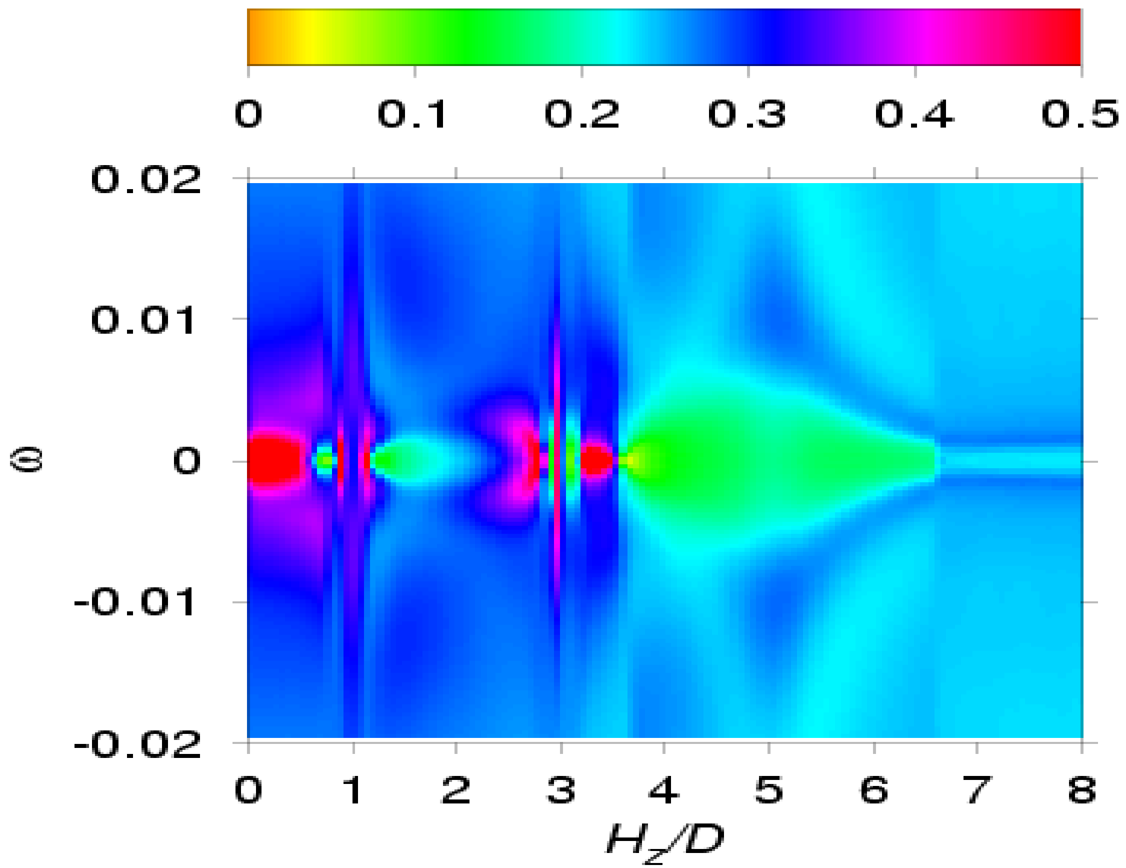
$S\text{-}1/2 \leftarrow S$

$H_z\text{-}D \leftarrow H_z$



Parameters: $W=1,\ D=5*10^{-5}W,\ B_2=0.1D,\ \ J=0.15W$

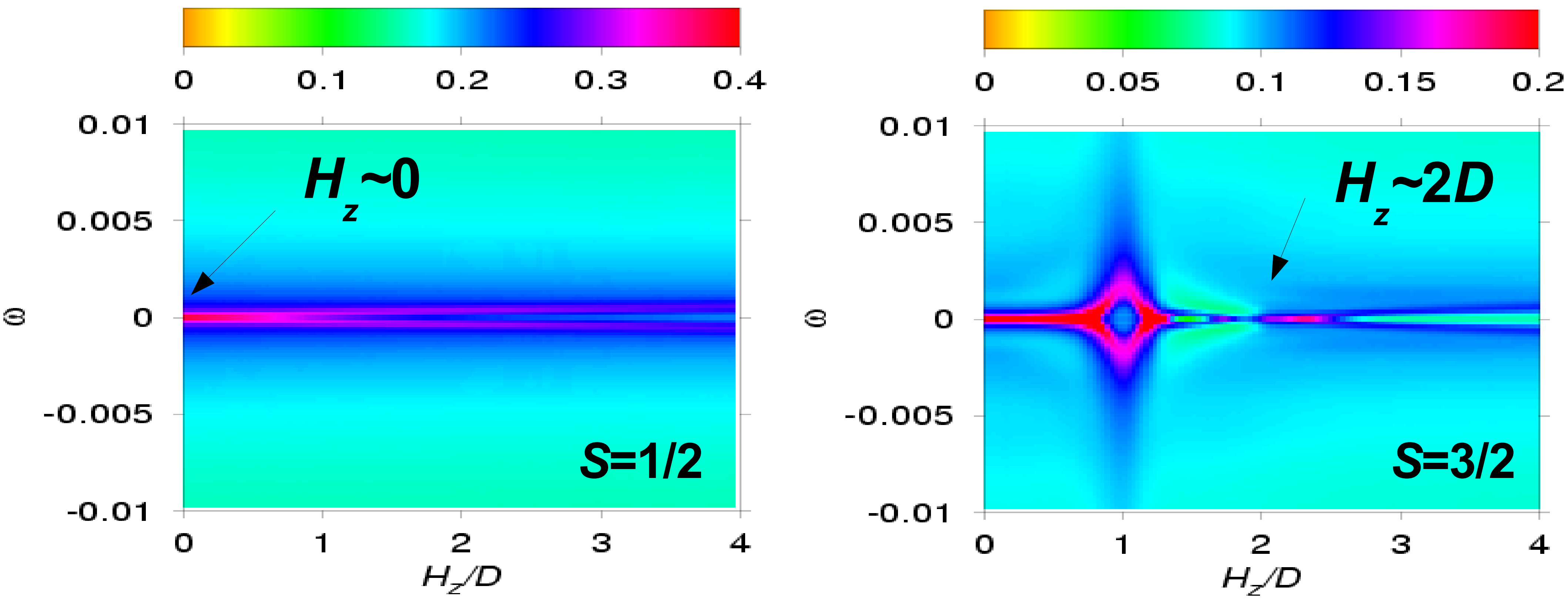
Entertainment break



Relation integer / half-integer spin

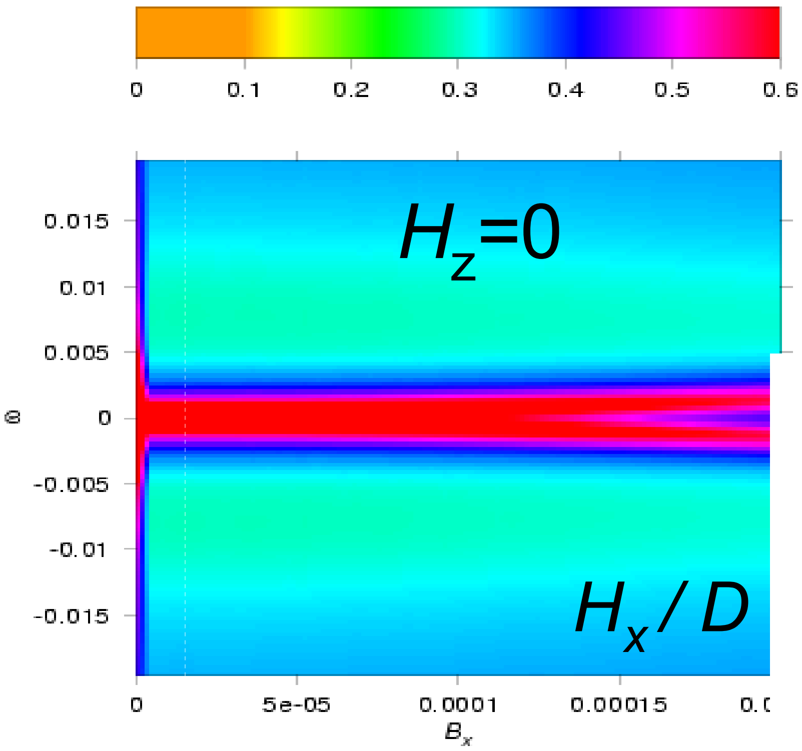
Iteration
$n=1,2,..,2S-1$

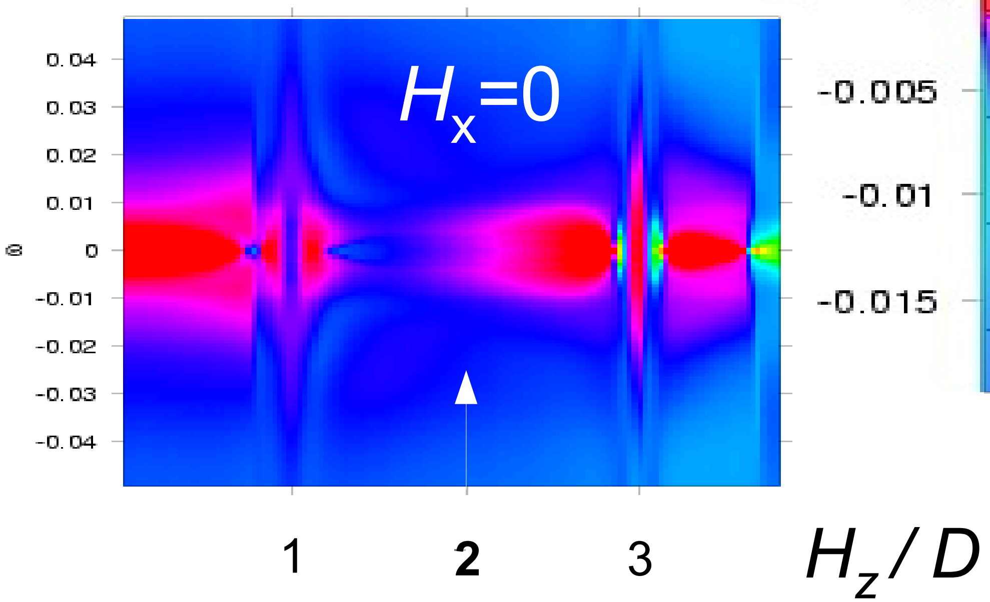
$S-n/2,\ H_z\sim 0 \leftarrow S,\ H_z\sim nD$



Transverse field effect

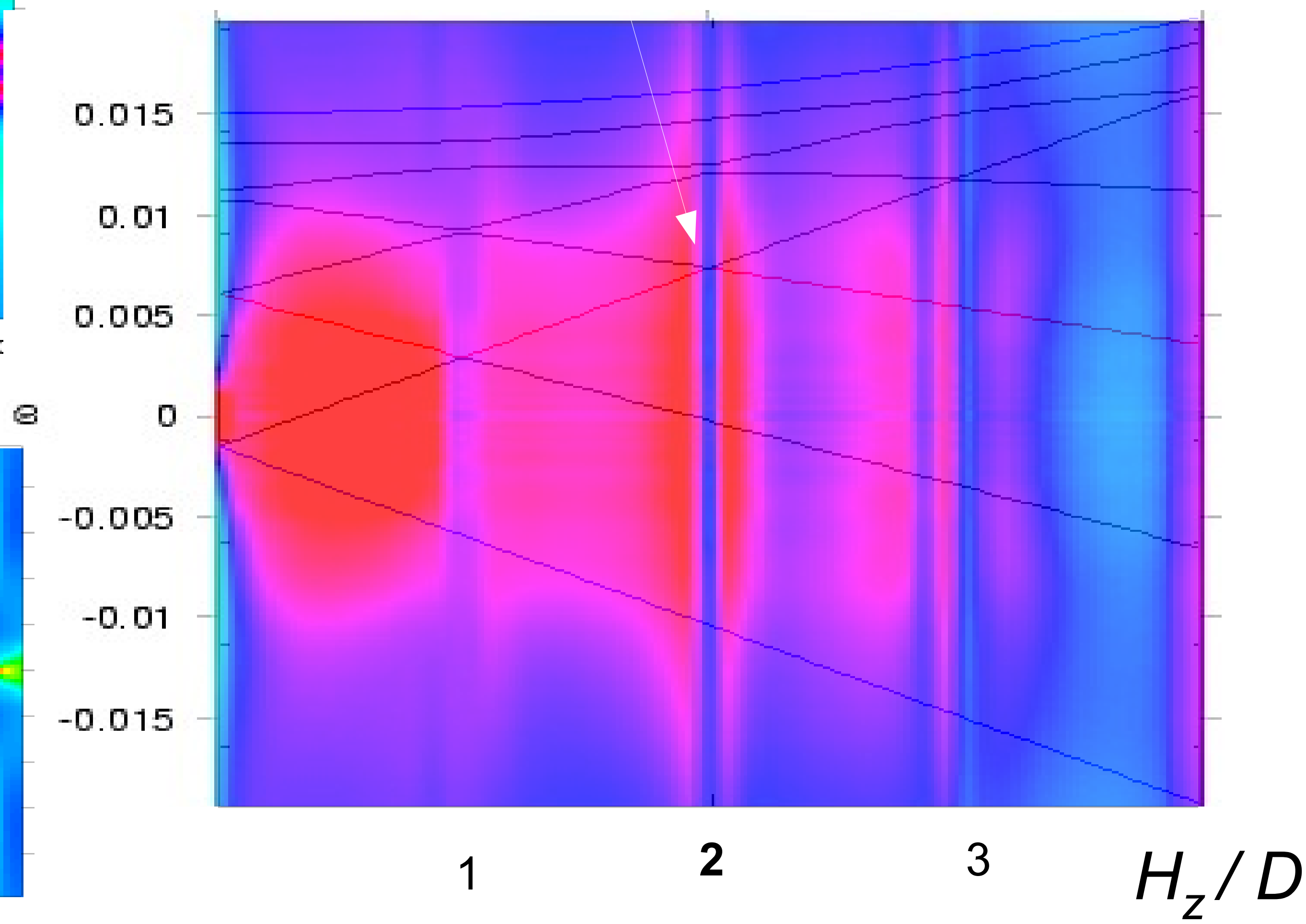
$H_{Zeeman} = H_z S_z + H_x S_x$ *easy* + *hard axis*





Field (H_x) *induced* QTM
detected by sweeping H_z

crossing for H_x=0



D=5e-5=10B2=5Bx, J=0.2

Magnetic field Kondo spectroscopy

- Magnetic parameters *of SMM in electrical circuit*
- Magnetic field energies < 2 D S ~ 1 meV

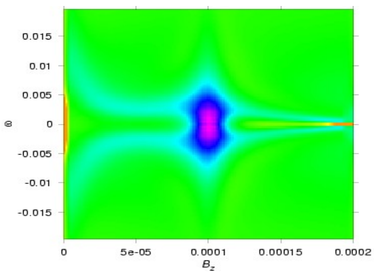
    no complete Zeeman splitting required
- Temperatures ~ T_K >> 2 D S ~ 1 meV

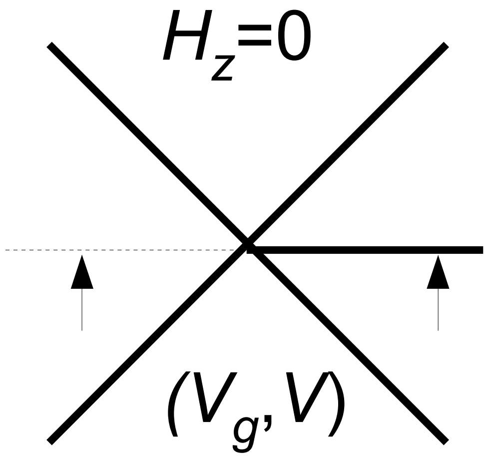
    no mK required
- Control J : tunneling distance (STM) or gate (3-terminal)
- 3-terminal transport:

    "Weak" & "strong" Kondo ~ gate-tunable

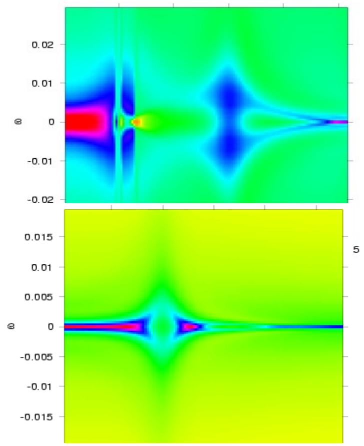
    Kondo in subsequent charge and *spin* states

S=integer





S±1/2=half-integer



S+1/2

S-1/2