ISSP Workshop
25 July 2006

Magnetism in Nano-Scale Systems

University of Tokyo

Seiji Miyashita

Energy level structure

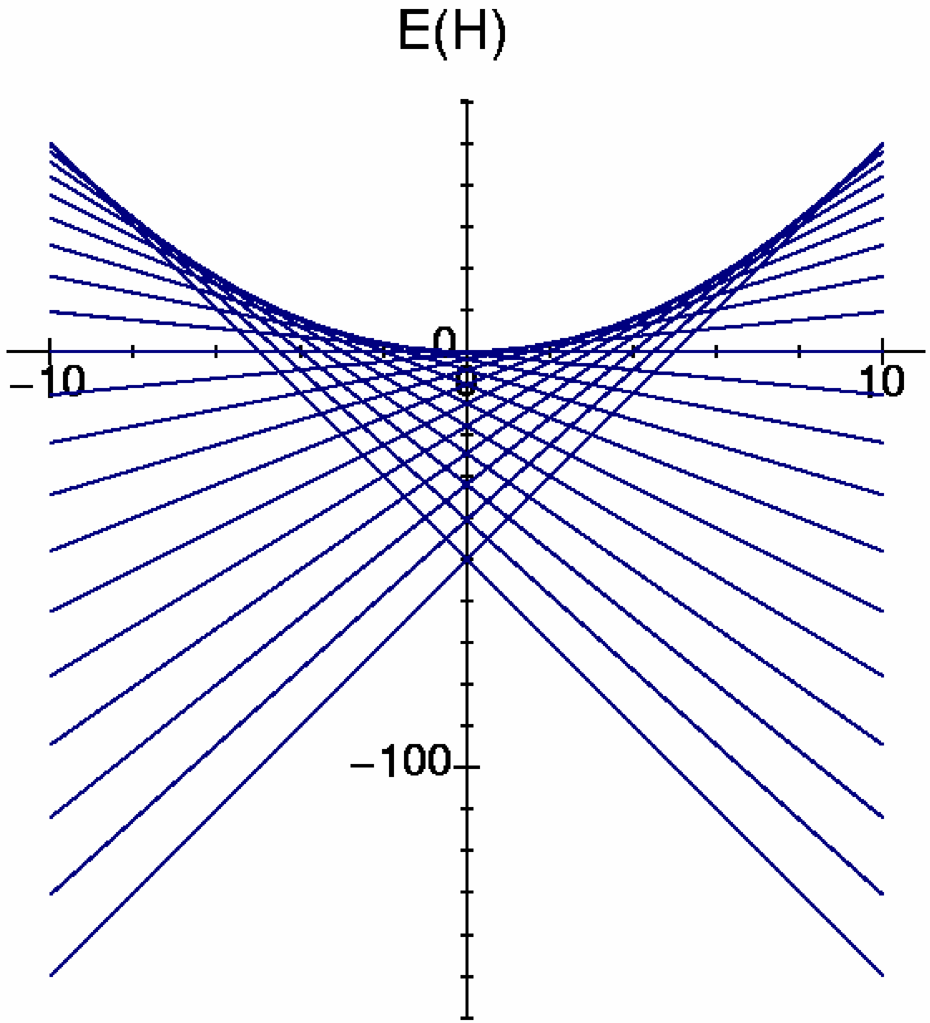
$$H|\Psi\rangle_i = E_i|\Psi\rangle_i$$

spin operator : (S_x, S_y, S_z) $[S_x, S_y] = iS_z$, etc

$$H = -DS_z^2 - HS_z$$

total spin S $\quad \vec{S}\cdot\vec{S} = S_x^2 + S_y^2 + S_z^2$

$$\begin{cases} \vec{S}\cdot\vec{S}|S,M\rangle = S(S+1)|S,M\rangle \\ S_z|S,M\rangle = M|S,M\rangle, \\ M = -S, -S+1, \cdots, S \end{cases}$$



$$H = -DS_z^2 - hS_z$$

$$H|S,M\rangle_i = -DM^2 - hM|S,M\rangle_i$$

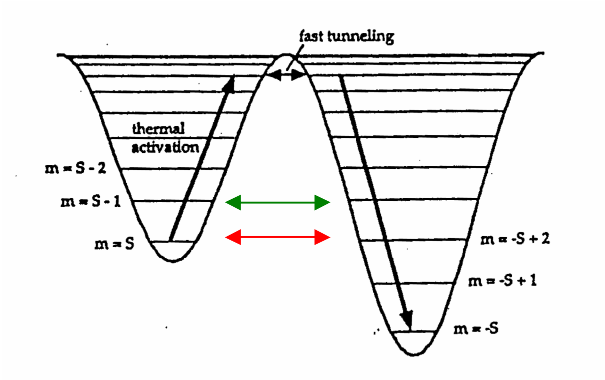
Level crossing

$h = 0 \quad M = \pm S$

$$-DM^2 - hM = -D(M+m)^2 - h(M+m)$$

$$h = -D(2M+m)$$

Resonance tunneling



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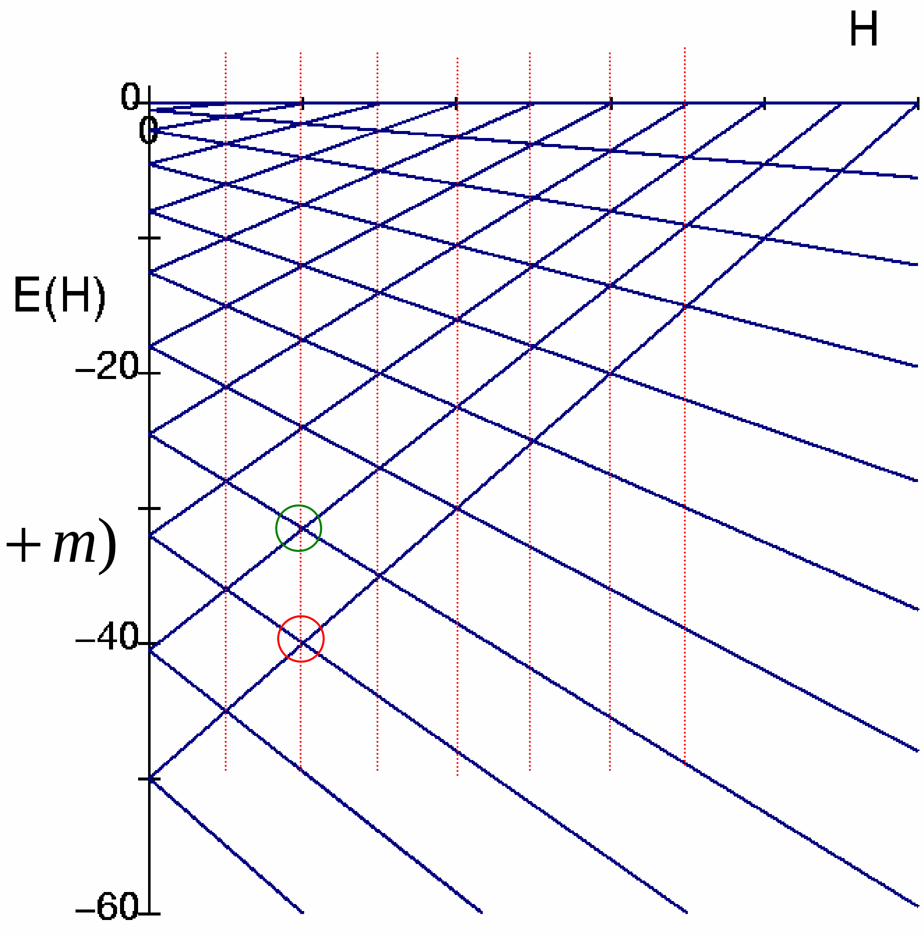
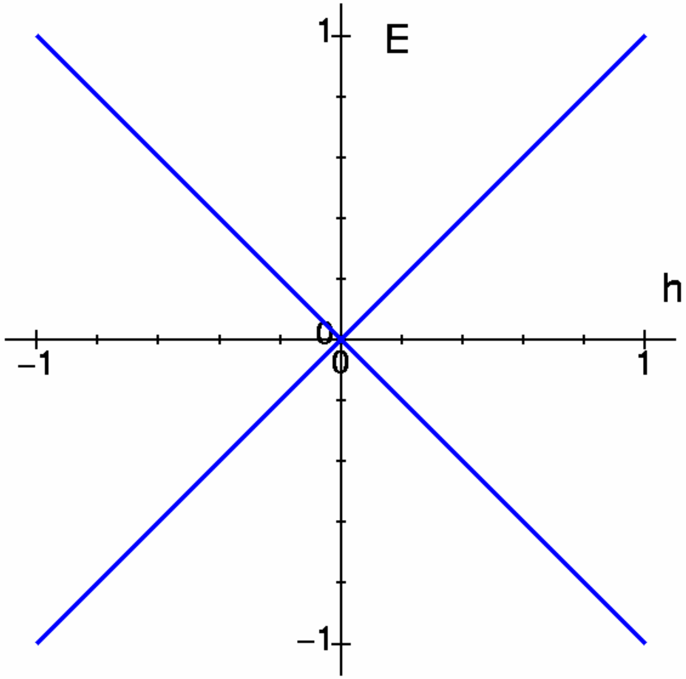


Level cross at h=Dn

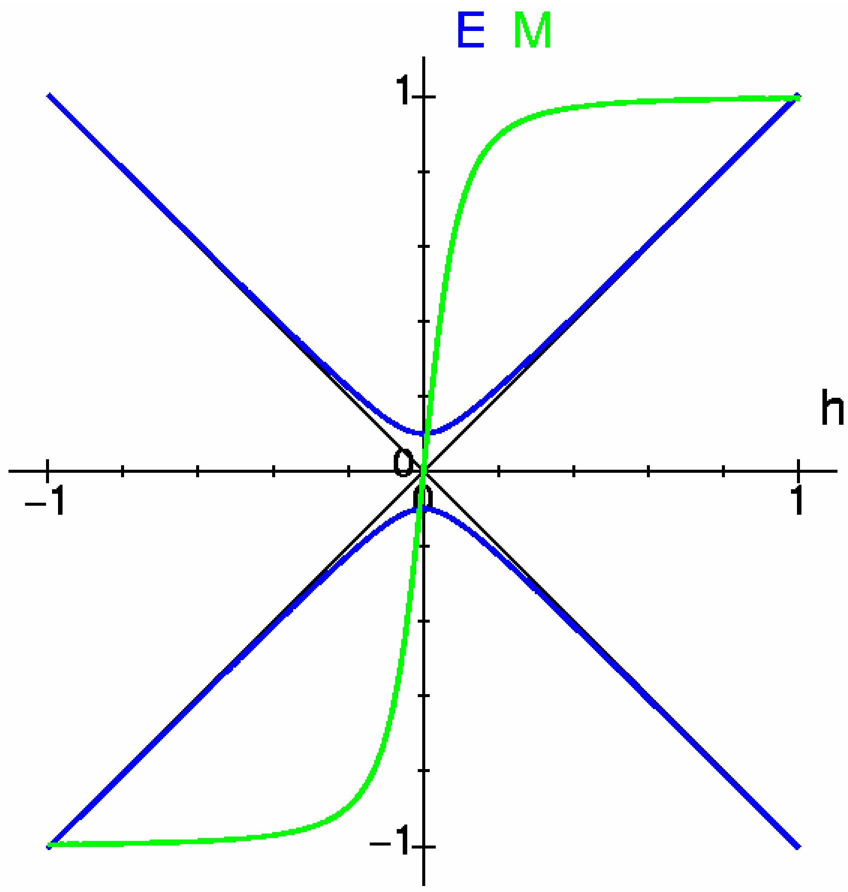
Avoided level crossing



$$[H, S_Z] \neq 0$$

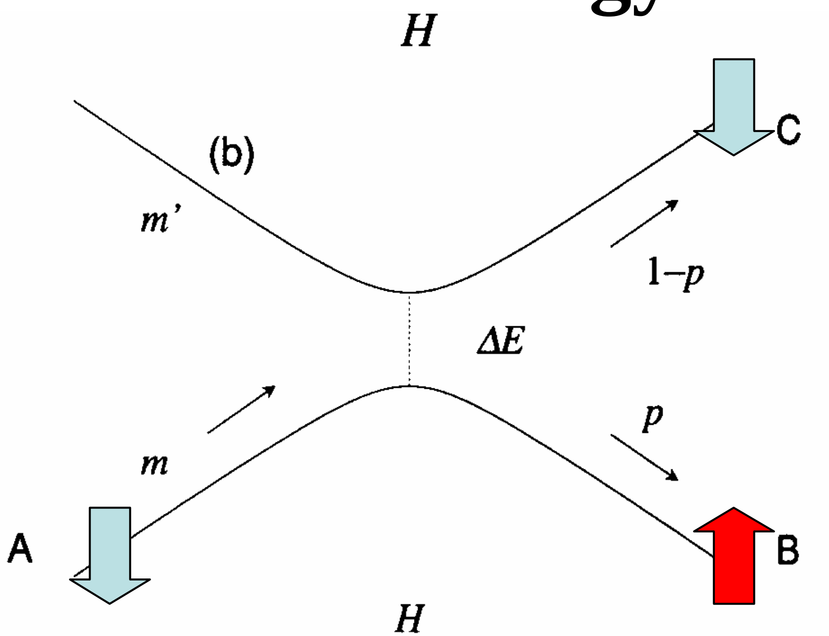
$$H = -hS_z + \Gamma S_x = \frac{1}{2}\begin{pmatrix} -h & \Gamma \\ \Gamma & h \end{pmatrix}$$

$$\begin{pmatrix} -h & \Gamma \\ \Gamma & h \end{pmatrix}\begin{pmatrix} \alpha_\pm \\ \beta_\pm \end{pmatrix} = \pm\sqrt{h^2 + \Gamma^2}\begin{pmatrix} \alpha_\pm \\ \beta_\pm \end{pmatrix}$$



Nonadiabatic Transition

Quantum Dynamics in Discrete Energy Levels



Change in Sweeping Field

SM, JPSJ 64(1995) 3207, 65(1996) 2734.
H. De Raedt et al, PRB56 (1997) 2734

$$H = -\infty \quad |\Psi\rangle = |\downarrow\rangle$$

$$H(t) = ct - h_0, \quad h_0 \to -\infty$$

$$|\Psi(t)\rangle = e_{\to}^{-i\int_0^t H(s)ds/\hbar}|\downarrow\rangle$$

$$\lim_{t\to\infty}|\Psi(t)\rangle = \alpha|\downarrow\rangle + \beta|\uparrow\rangle$$

adiabatic change $\quad p = |\beta|^2$

non-adiabatic transition $\quad 1-p$

Landau-Zener-Stueckelberg Mechanism

C. Zener, Proc. R. Soc. (London) Ser. A137 (1932) 696.

Resonant Tunneling

$$p = 1 - \exp\left(-\frac{\pi(\Delta E)^2}{2|M_{\text{in}} - M_{\text{out}}|v}\right)$$

Control of quantum states

- Field sweep LZS mechanism
- Quantum oscillation (tunneling resonance)
- Alternating field

Sx Rabi oscillation

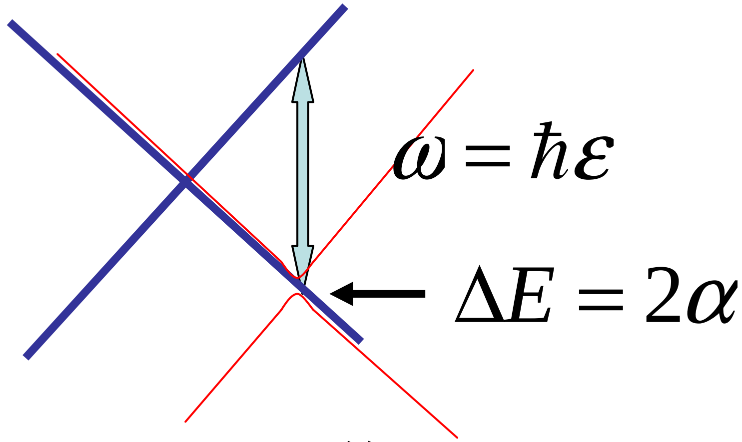
(e.g. π /2 pulse NMR)

Sz Floquet phenomena

(Nontrivial resonance)

Spin-rotation pulse

Transverse oscillating filed



$$i\hbar\frac{\partial}{\partial t}\left|\psi\right\rangle = H(t)\left|\psi\right\rangle$$

$$= \left(-\hbar\varepsilon S^{z} + \alpha\left(S^{+}\mathrm{e}^{\mathrm{i}\omega t} + S^{-}\mathrm{e}^{-\mathrm{i}\omega t}\right)\right)\left|\psi\right\rangle$$

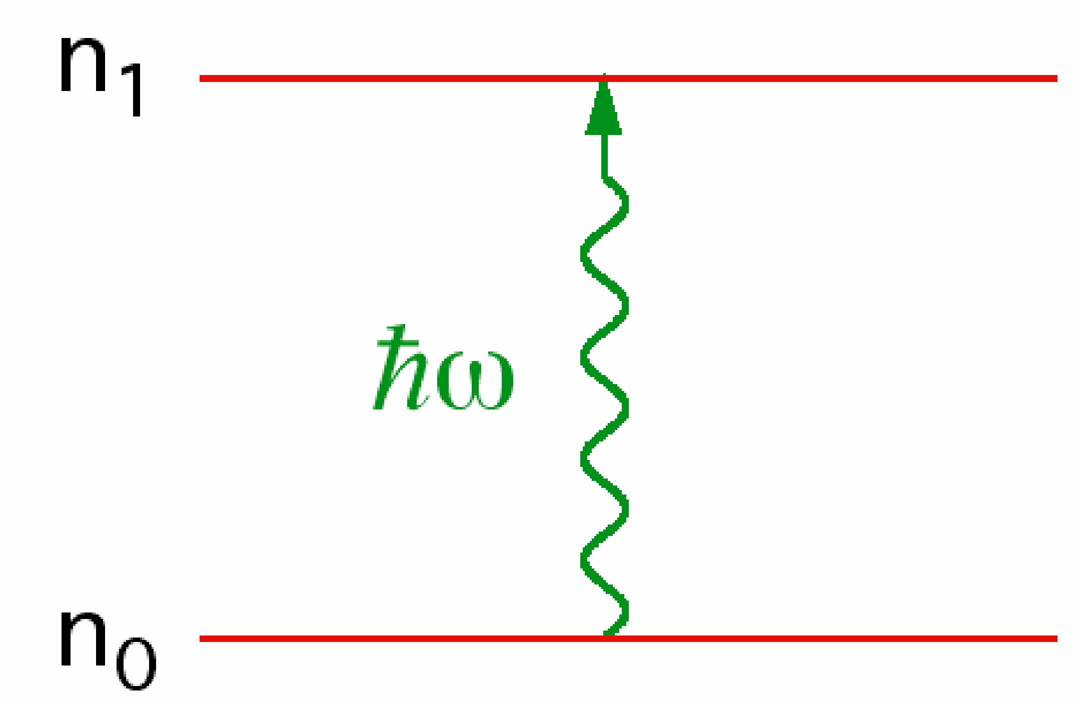
$$\left|\phi\right\rangle = \mathrm{e}^{-\mathrm{i}\omega S^{z}t}\left|\psi\right\rangle$$

$$i\hbar\frac{\partial}{\partial t}\left|\phi\right\rangle = \left(-\hbar(\varepsilon-\omega)S^{z} + 2\alpha S^{x}\right)\left|\phi\right\rangle$$

Pulse duration τ

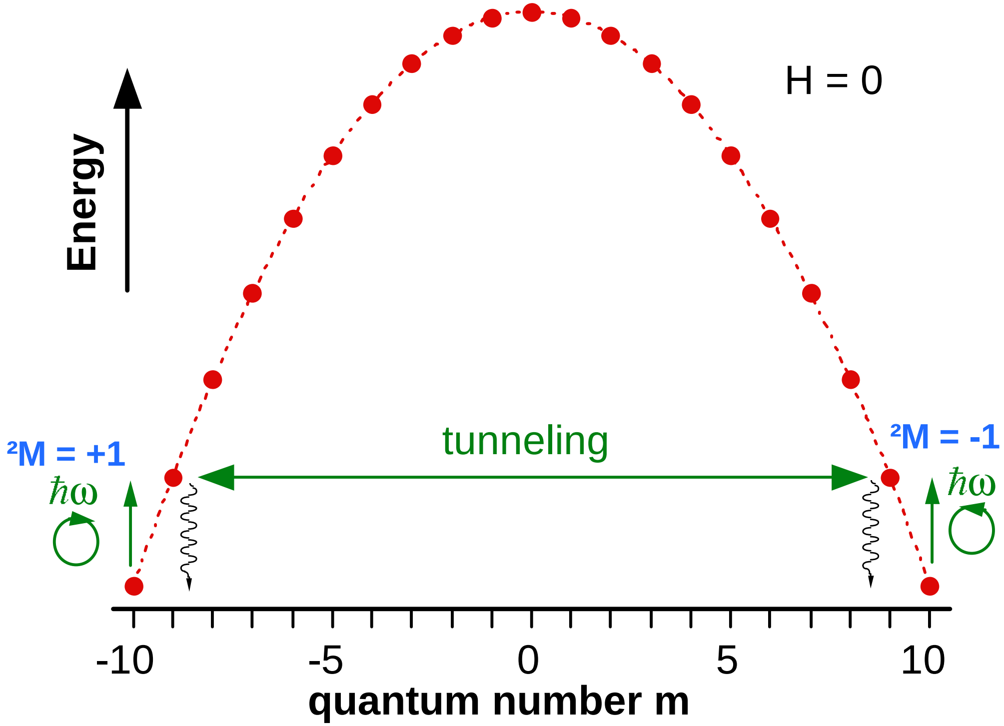
$2\alpha\tau = \frac{\pi}{2}$ Rotation around the x - axis : 90°

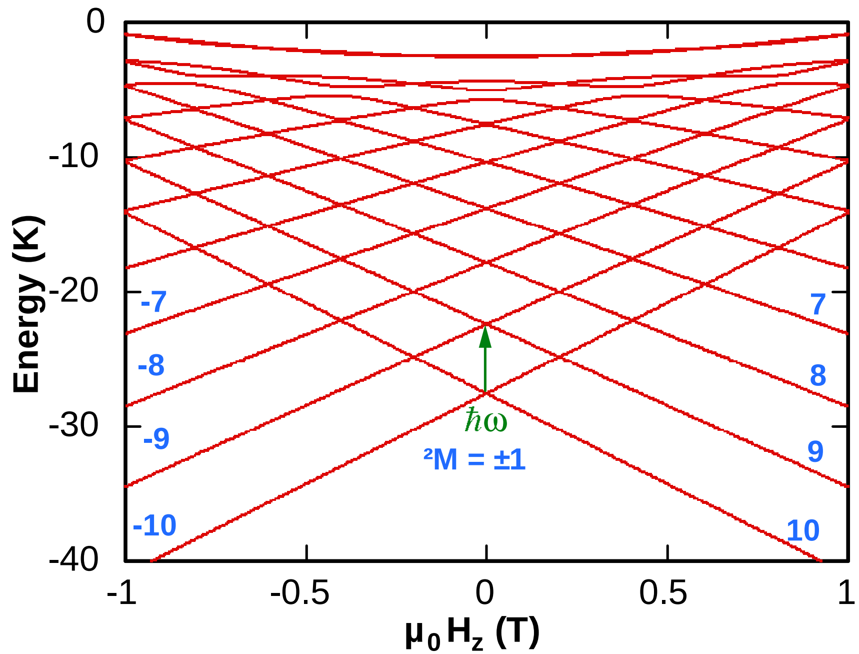
Interaction with photons
(microwaves: 1 to 115 GHz)



Photon assisted tunneling

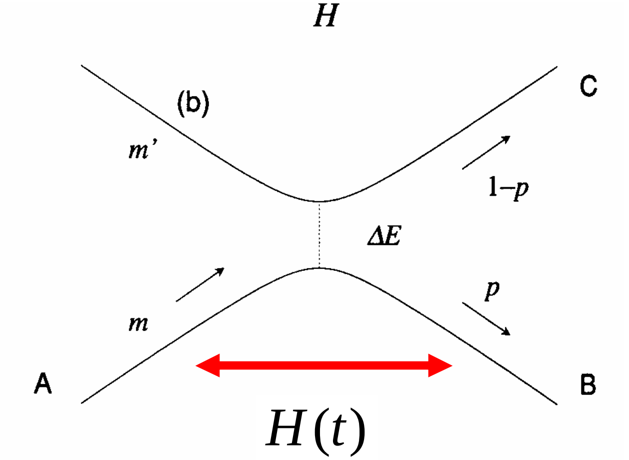
Absorption of circular polarized microwaves





Resonance on the AC field
Non-trivial Resonance

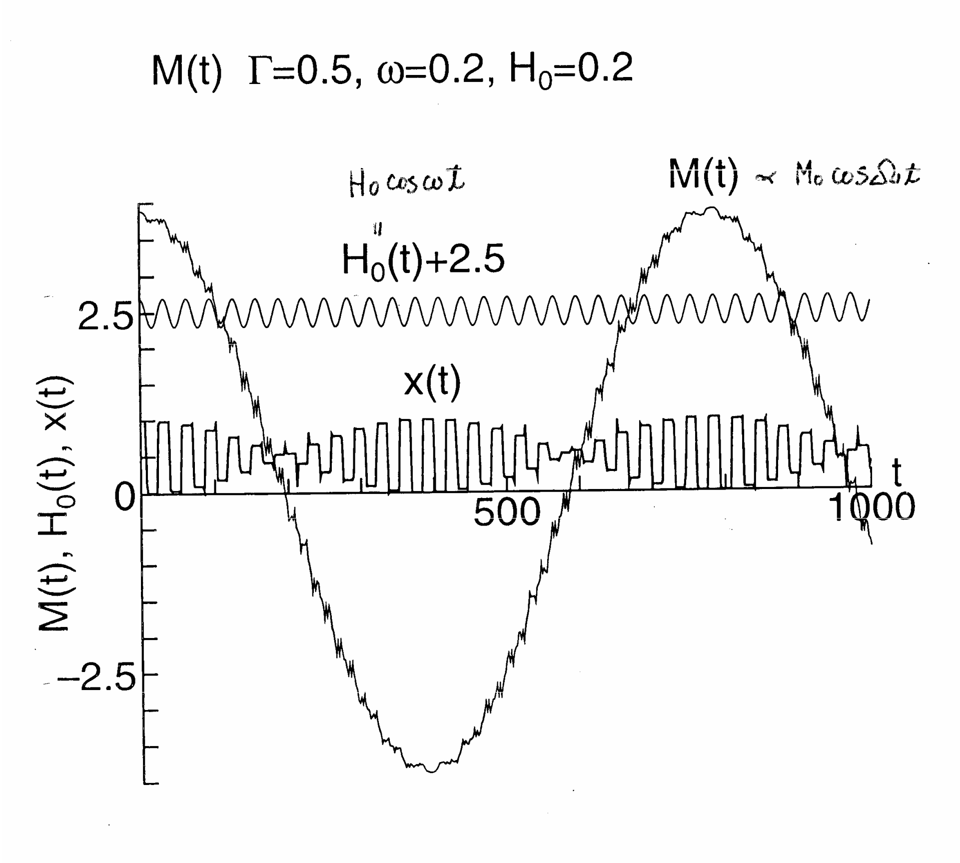
$$H(t) = -h_W \cos(\omega t) \sum_i S_i^z$$



$$M(t) = M_0 \cos(\Omega t + \delta),$$

$$\Omega = \frac{\omega}{\pi} \sqrt{2p(1 - \cos\alpha)}$$

$$p = 1 - \exp\left(-\frac{\pi(\Delta E)^2}{4c\Delta M} \right)$$



Y. Kayamuma, PRB 47 (1993) 9940
SM, K. Saito, H. De Daedt,
Phys. Rev. Lett. 80 (1998) 1525.

Hz resonance

$$H(t) = -\Gamma S^x - \left(h + A\sin(\omega t)\right)S^z, \quad \vartheta(t) = \int_0^t \left(h + A\sin(\omega s)\right)ds$$

$$F = T\mathrm{e}^{\int_0^{2\pi/\omega} H(u)du}$$

$$= T\mathrm{e}^{\int_0^{2\pi/\omega} -\frac{i}{2}\Gamma\begin{pmatrix} 0 & \mathrm{e}^{i\vartheta(u)} \\ \mathrm{e}^{-i\vartheta(u)} & 0 \end{pmatrix}du} \approx \exp\left(-\frac{i}{2}\Gamma\begin{pmatrix} 0 & \varepsilon \\ \varepsilon^* & 0 \end{pmatrix}\right)$$

$$\varepsilon \equiv \int_0^{2\pi/\omega} \mathrm{e}^{i\vartheta(u)}du = \mathrm{e}^{iA/\omega}\sum_{n=-\infty}^{\infty}(-i)^n J_n\left(\frac{A}{\omega}\right)\int_0^{2\pi/\omega}\mathrm{e}^{i(h-n\omega)u}du$$

at $h = n\omega$

$$M(t) = \cos(\Omega t), \quad \Omega = \Gamma J_n\left(\frac{A}{\omega}\right)$$

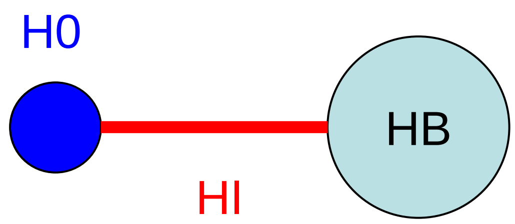
Quantum Master Equation

$$\frac{\partial}{\partial t}\rho = iL\rho = \frac{1}{i\hbar}[H_0 + H_\mathrm{I} + H_\mathrm{B}, \rho]$$

$$H = H_0 + H_\mathrm{I} + H_\mathrm{B},$$

$$H_\mathrm{I} = \sum_k \lambda_k \left(b_k^+ + b_k\right)X,$$

$$H_\mathrm{B} = \sum_k \omega_\kappa b_k^+ b_k$$



Reduction of environment

$$\sigma = \mathrm{p}\rho = \rho_\mathrm{eq}\mathrm{Tr_B}\rho, \quad \rho_\mathrm{eq} = \mathrm{e}^{-\beta H_0} / \mathrm{Tr_B e}^{-\beta H_0}$$

$$\frac{\partial}{\partial t}\sigma = i\mathrm{p}L\sigma + \mathrm{p}iL\int_0^t \mathrm{e}^{(t-s)(1-\mathrm{p})\mathrm{iL}}(1-\mathrm{p})iL\mathrm{p}\rho(t)ds$$

$$+ \quad \mathrm{p}iL\mathrm{e}^{(t-s)(1-\mathrm{p})\mathrm{iL}}(1-\mathrm{p})\rho(0)$$

e.g. Photon dissipation and pumping :

(SM., H. Ezaki, and E. Hanamura PRA 57 (1998) 2046)

$$\frac{\partial\sigma}{\partial t} = \frac{1}{i\hbar}[H_0, \sigma] - \kappa\left(b^+ b\sigma - 2b\sigma b^+ + \sigma b^+ b\right)$$

Lindblad form ➔ Stochastic Schrodinger Equation (antibunching, squeezing photo emission)

General formulation

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[H,\rho]$$

$$-\frac{\lambda^2}{\hbar^2}\int_0^t ds \int_{-\infty}^{\infty} d\omega e^{i\omega t}\Phi(\omega)\{XX(-s)\rho(t) - e^{\beta\hbar\omega}X\rho(t)X(-s)$$

$$+ e^{\beta\hbar\omega}\rho(t)X(-s)X - X(-s)\rho(t)X \}$$

time correlation function of the reservoie's operators $\Phi(t)$

$$\Phi(\omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t}\Phi(t) = \hbar\gamma(\omega)^2 \frac{D(\omega) - D(-\omega)}{e^{\beta\hbar\omega} - 1}$$

$I(\omega) = \gamma(\omega)^2 D(\omega) = I_0\omega^\alpha \quad \omega > 0:$ the spectral density

Independent phonon bath

$$H = H_0 + H_\mathrm{I} + H_\mathrm{B},$$

$$H_\mathrm{I} = \sum_k \lambda_k \left(b_k^+ + b_k\right)X,$$

$$H_\mathrm{B} = \sum_k \omega_\kappa b_k^+ b_k$$

$$\frac{d\rho}{dt} = -i[H,\rho] - \lambda\left([X,R\rho] + [X,R\rho]^+\right)$$

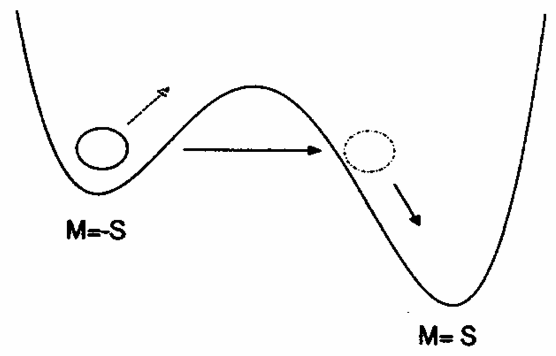
$$\langle k|R|m\rangle = \zeta\left(\frac{E_k - E_m}{\hbar}\right) n_\beta(E_k - E_m)\langle k|X|m\rangle,$$

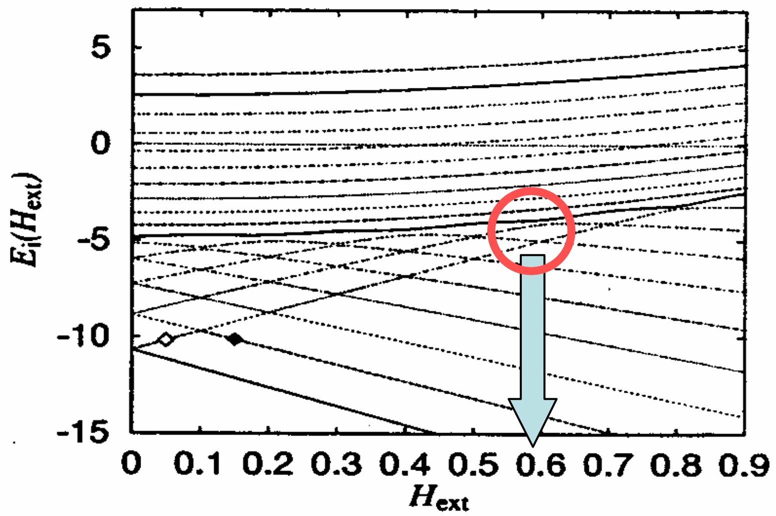
$$\zeta(\omega) = I(\omega) - I(-\omega)$$

K. Saito, S. Takesue and SM. Phys. Rev. B61 (2000) 2397

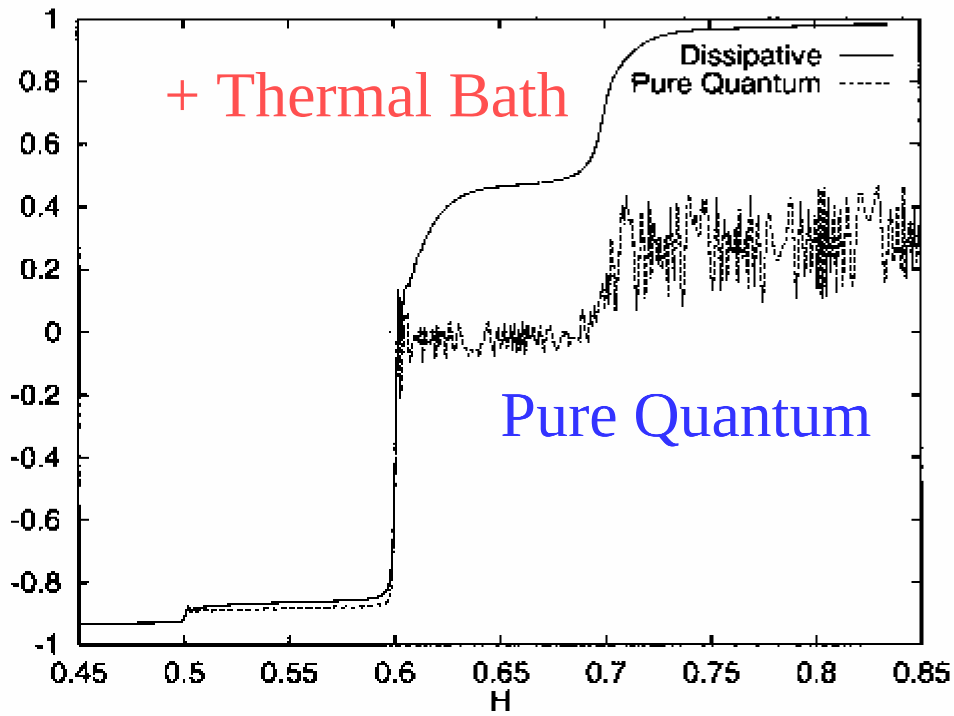
No feedback effects

Adiabatic transition and Relaxation





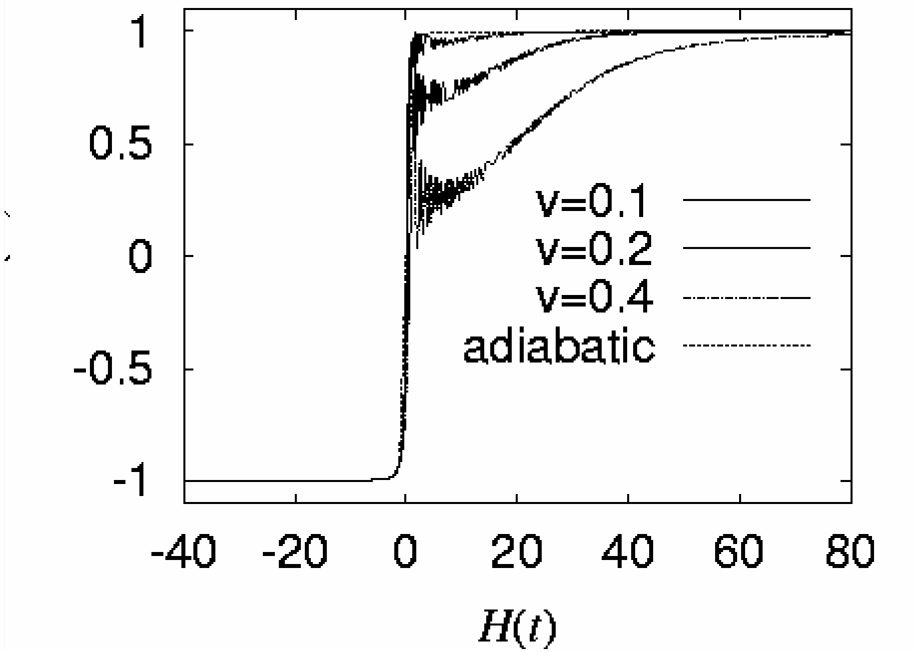
$T \to 0$



K. Saito, SM, H.de Raedt,
Phys. Rev. B60 (1999) 14553

Field sweeping with thermal bath

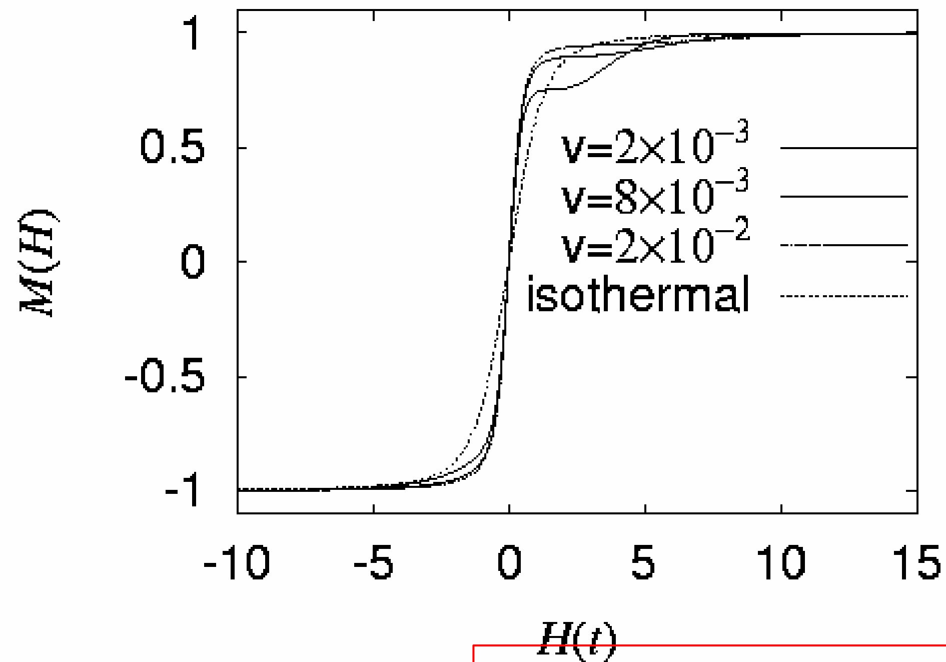
Fast sweeping



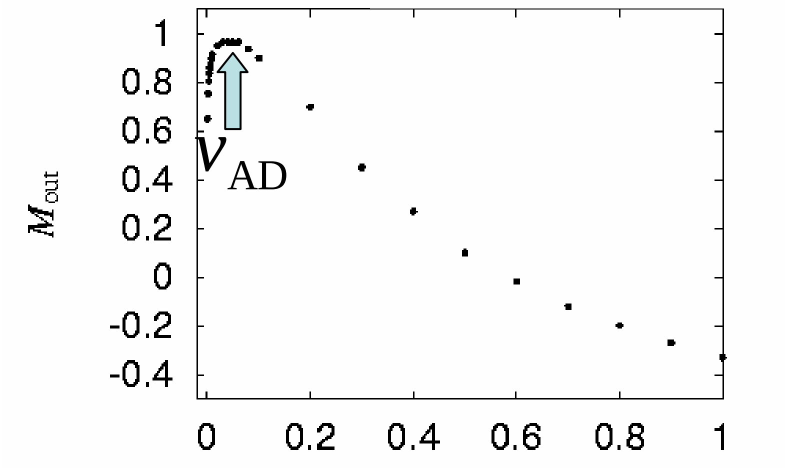
$$v_{AD} < v$$

LZS

Slow sweeping



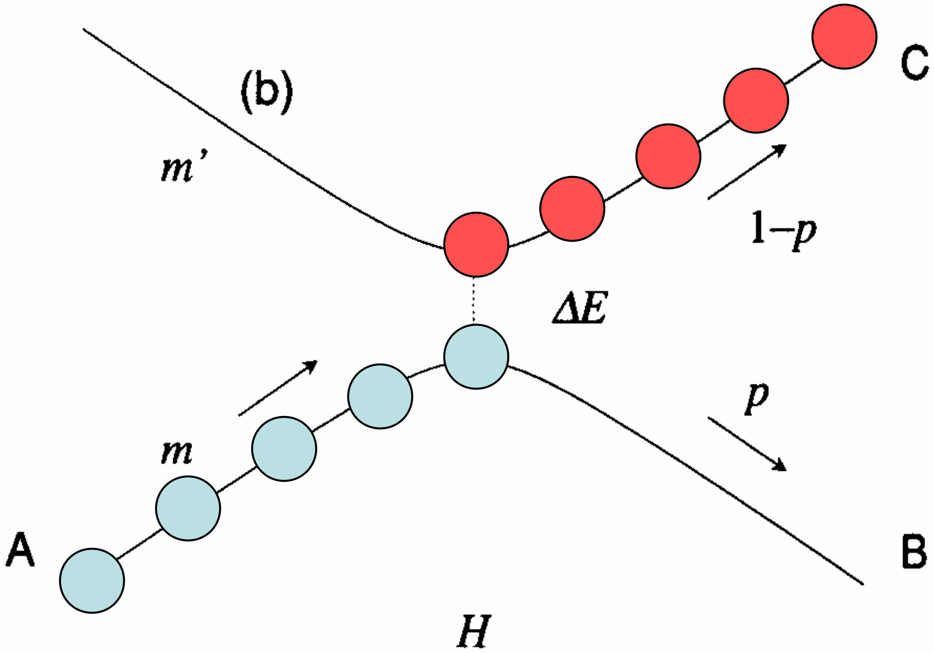
$$v_{TH} < v < v_{AD}$$

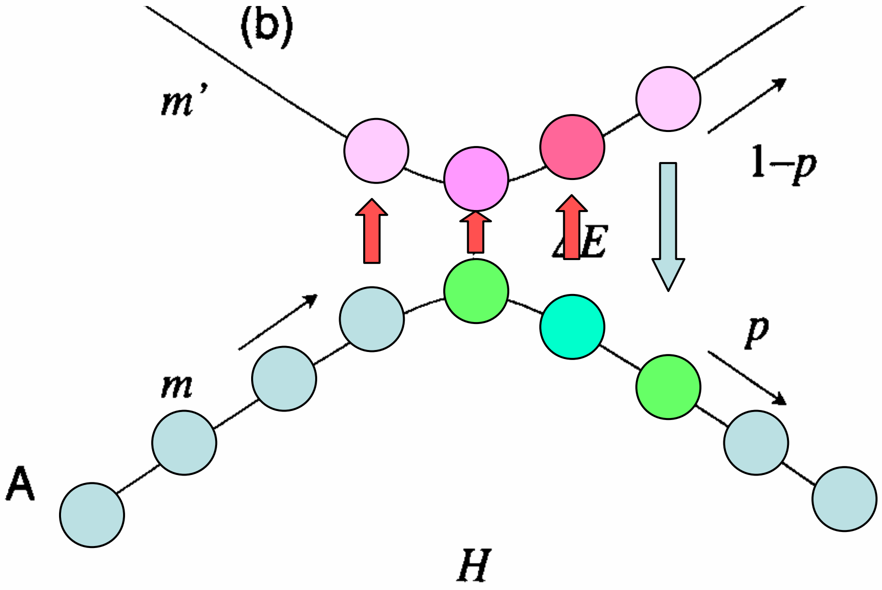


Magnetic FoehnEffect

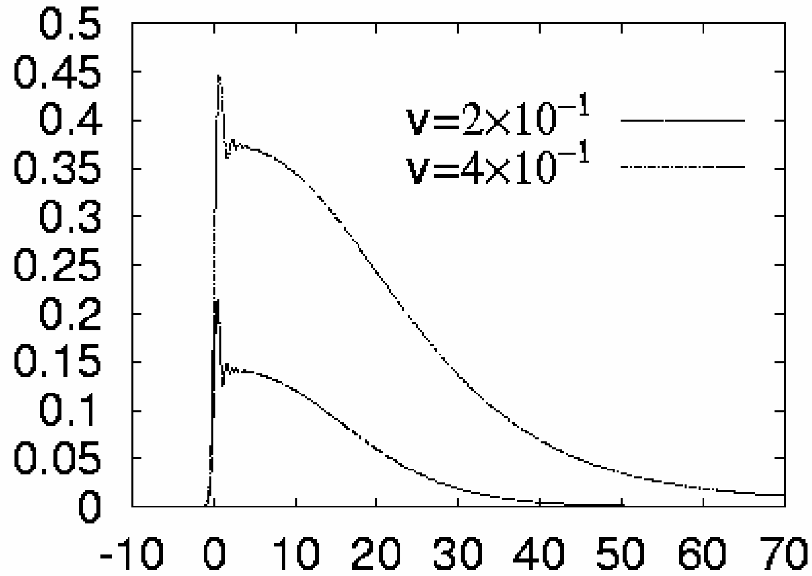
K. Saito & SM. JPSJ (2001) 3385.

Nonadiabatic Tr. & Heat-inflow

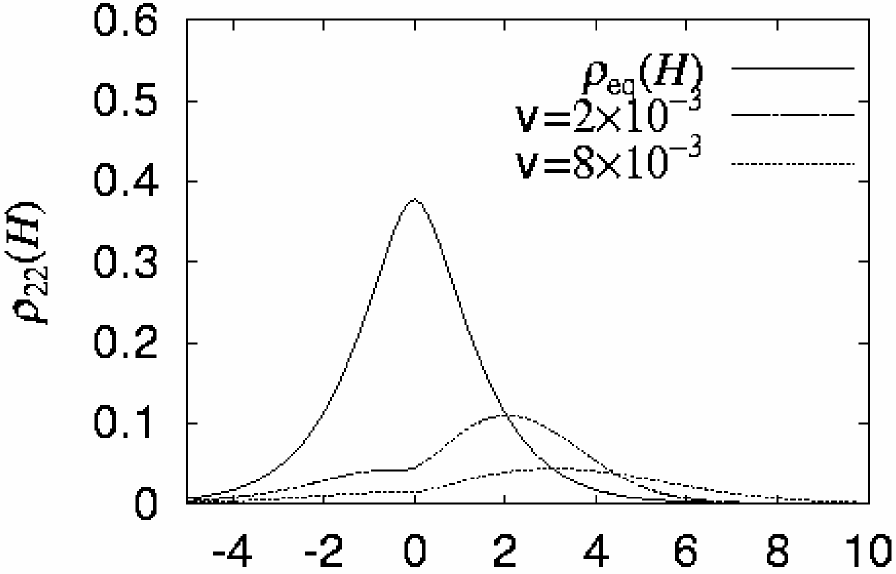




LZ transition

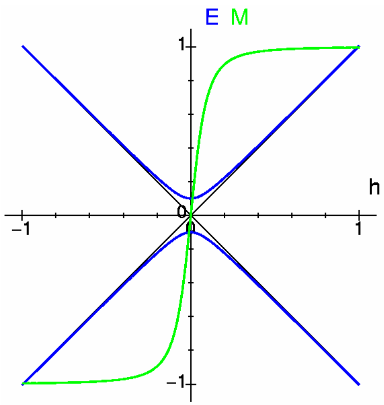


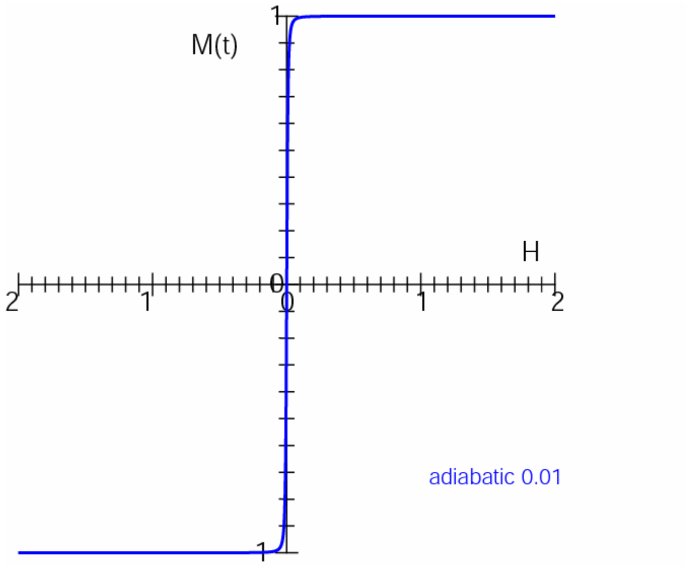
Magnetic Foehn Effect



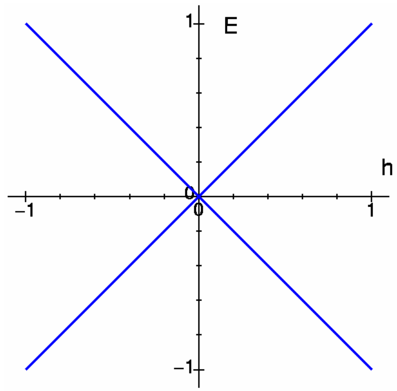
Various types of magnetization process

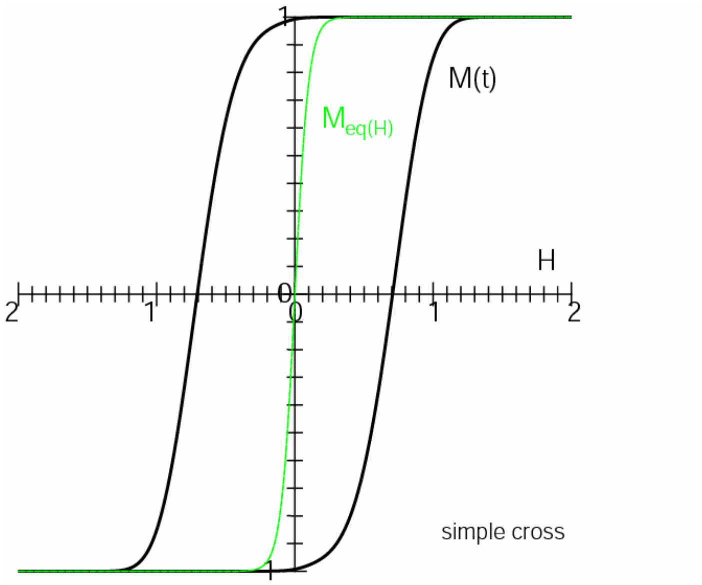
Adiabatic change





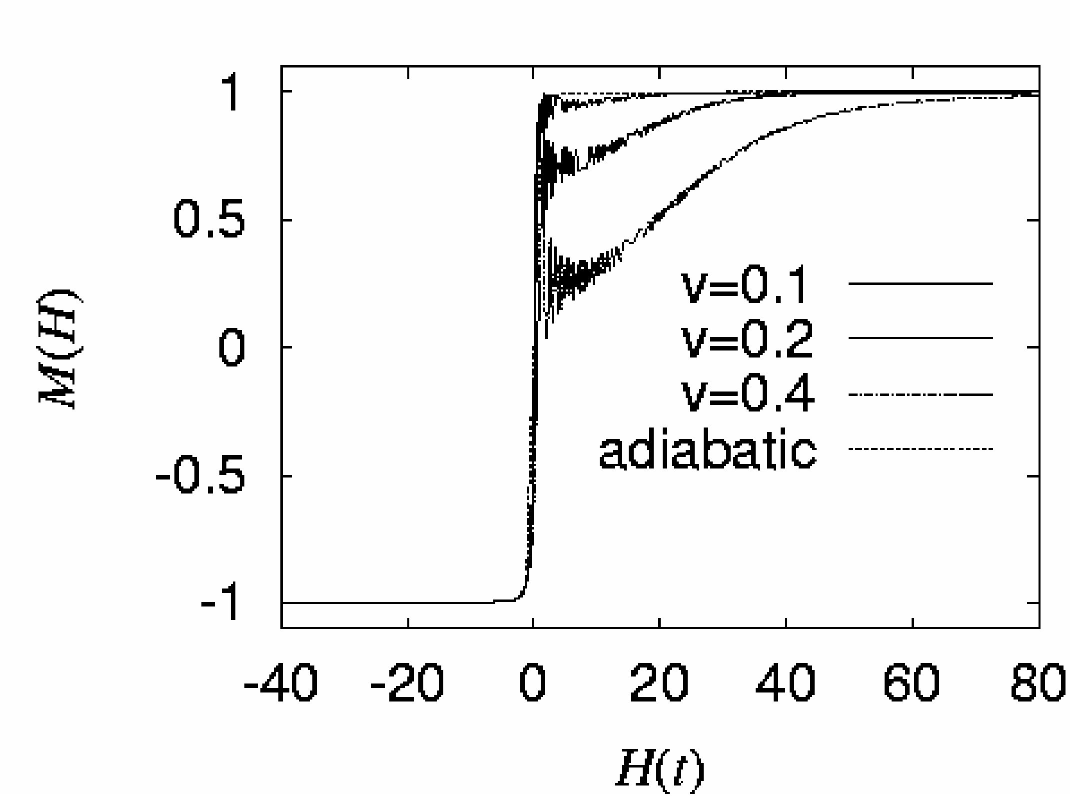
Thermal relaxation (no Gap)

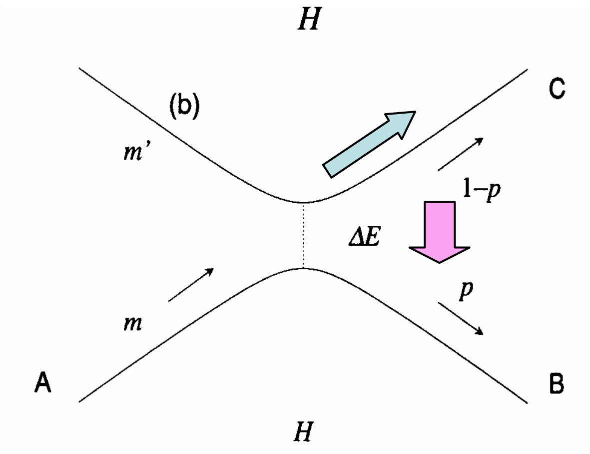




Non adiabatic transition

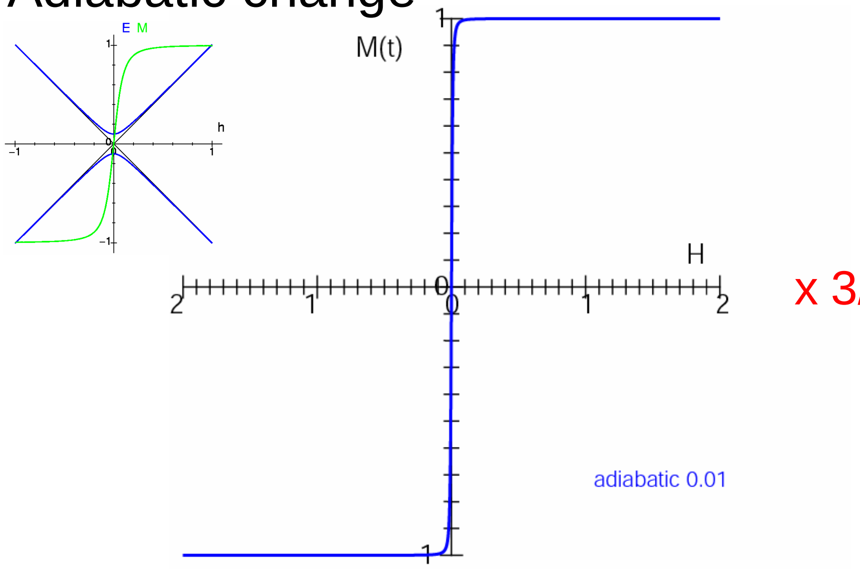
Fast sweeping $v_{AD} < v$





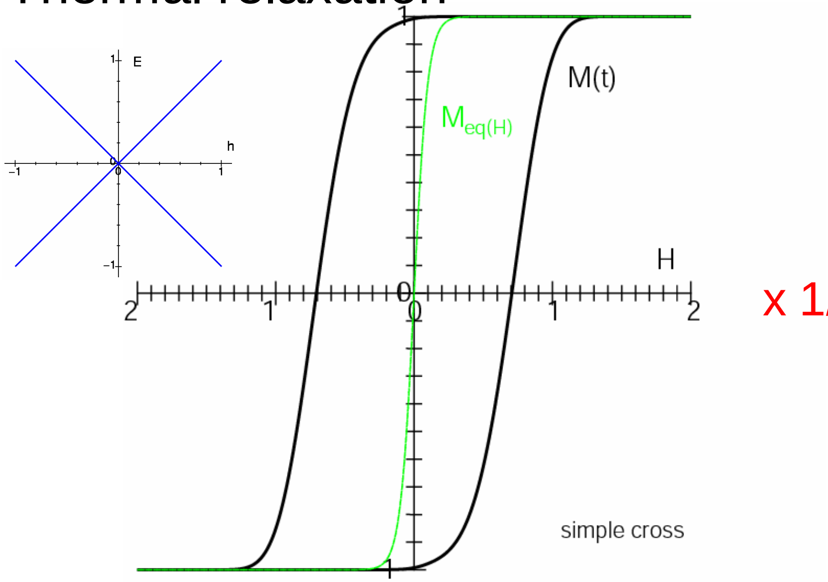
LZ transition + Thermal relaxation

Adiabatic change

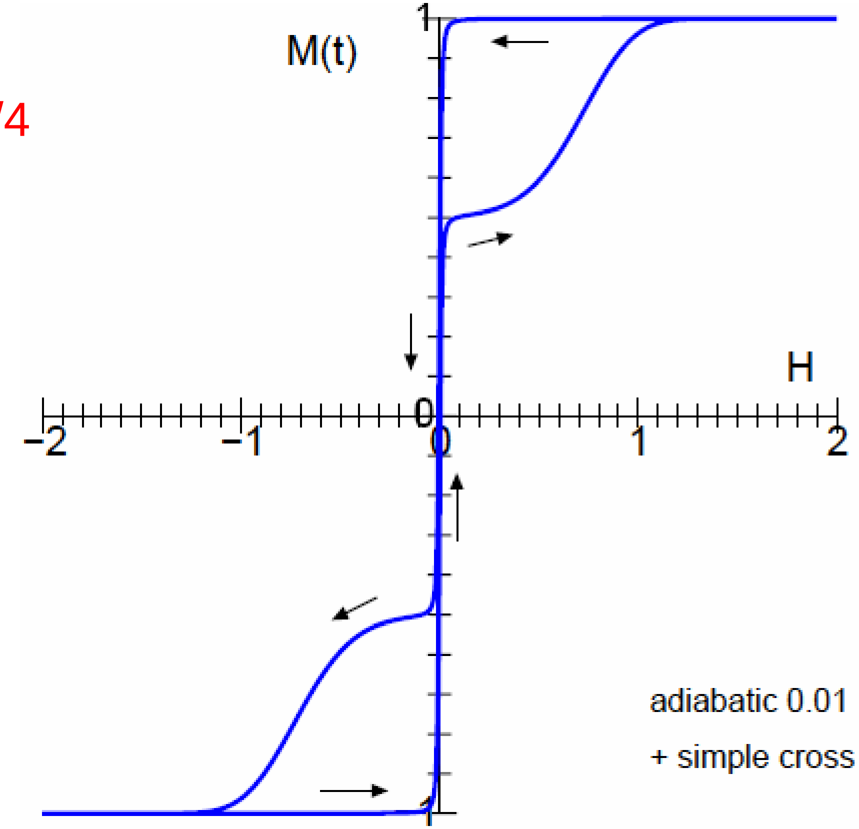


x 3/4

Thermal relaxation



x 1/4



cf. Tetra-nicle(II) D. H. Hendrickson

```
                                        g,vh,zi0,temp=   0.02000   0.00030   0.00000   0.30000
itime0, iout, maxtime=  366300     666  g,vh,zi0,temp=   0.02000   0.00050   0.00000   0.30000
itime0, iout, maxtime=  220000     400  g,vh,zi0,temp=   0.02000   0.00070   0.00000   0.30000
itime0, iout, maxtime=  157035     285  g,vh,zi0,temp=   0.02000   0.00100   0.00000   0.30000
itime0, iout, maxtime=  110000     200  g,vh,zi0,temp=   0.02000   0.00300   0.00000   0.30000
itime0, iout, maxtime=   36630      66  g,vh,zi0,temp=   0.02000   0.00500   0.00000   0.30000
itime0, iout, maxtime=   22000      40  g,vh,zi0,temp=   0.02000   0.01000   0.00000   0.30000
itime0, iout, maxtime=   11000      20  g,vh,zi0,temp=   0.02000   0.02000   0.00000   0.30000
itime0, iout, maxtime=    5500      10  g,vh,zi0,temp=   0.02000   0.03000   0.00000   0.30000
itime0, iout, maxtime=    3666       6      6666
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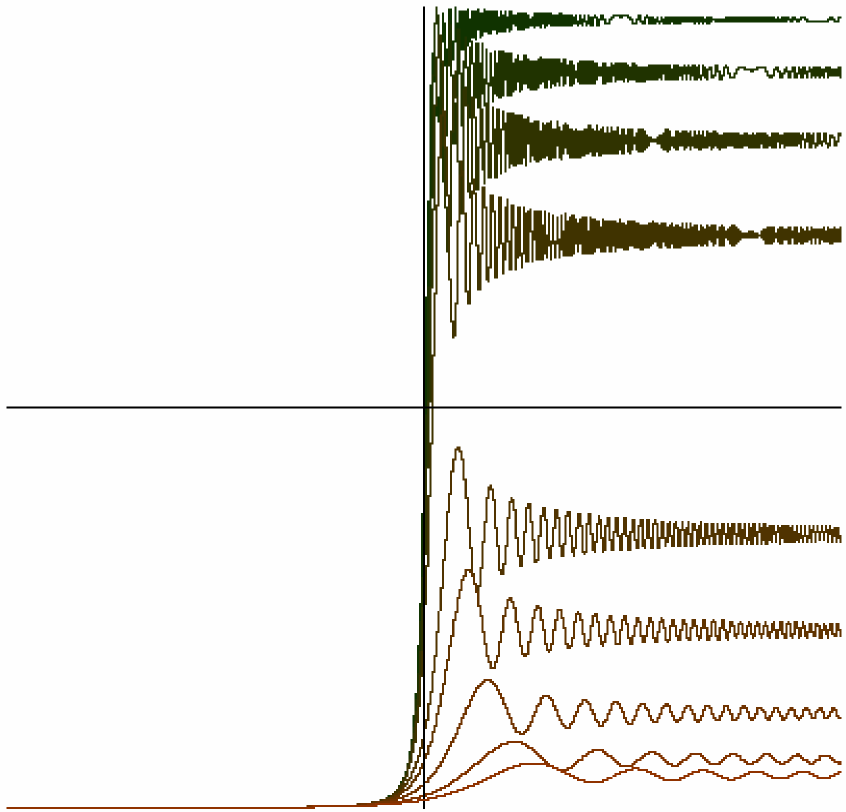

```
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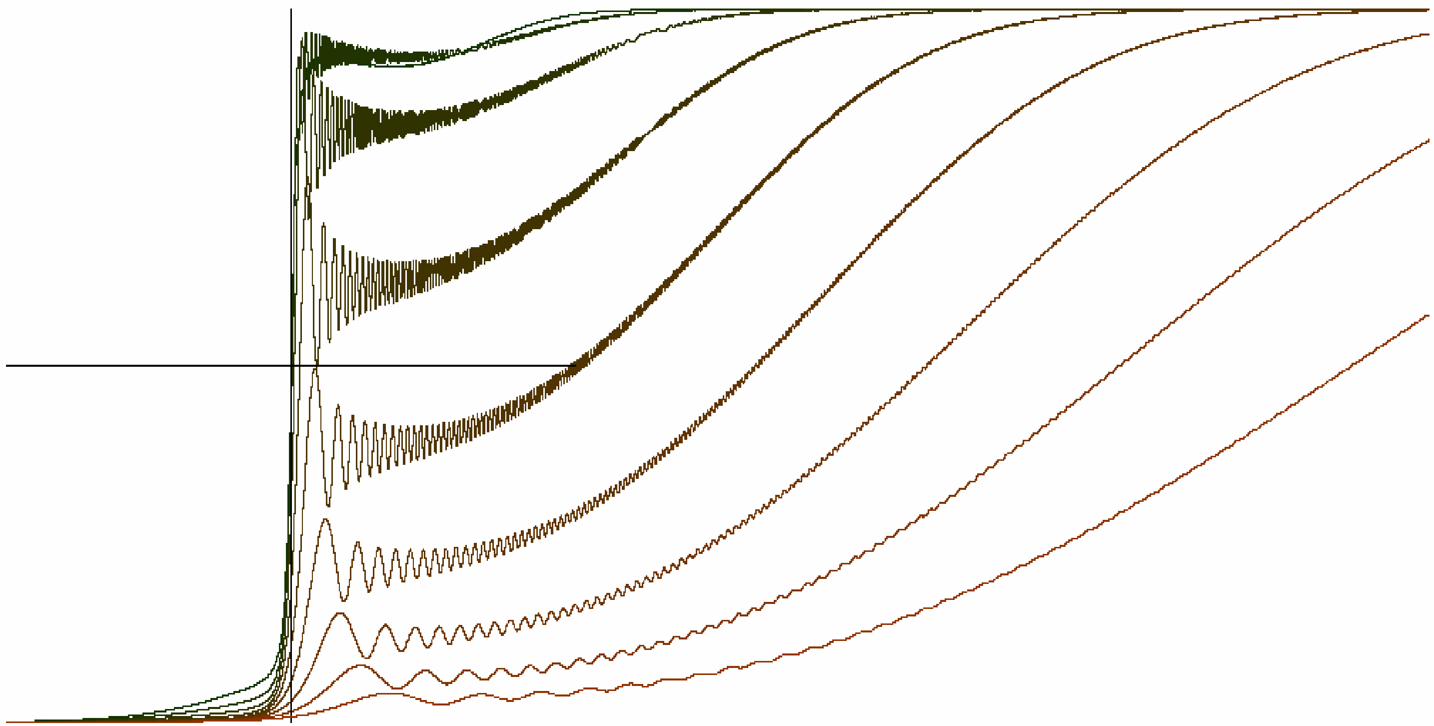
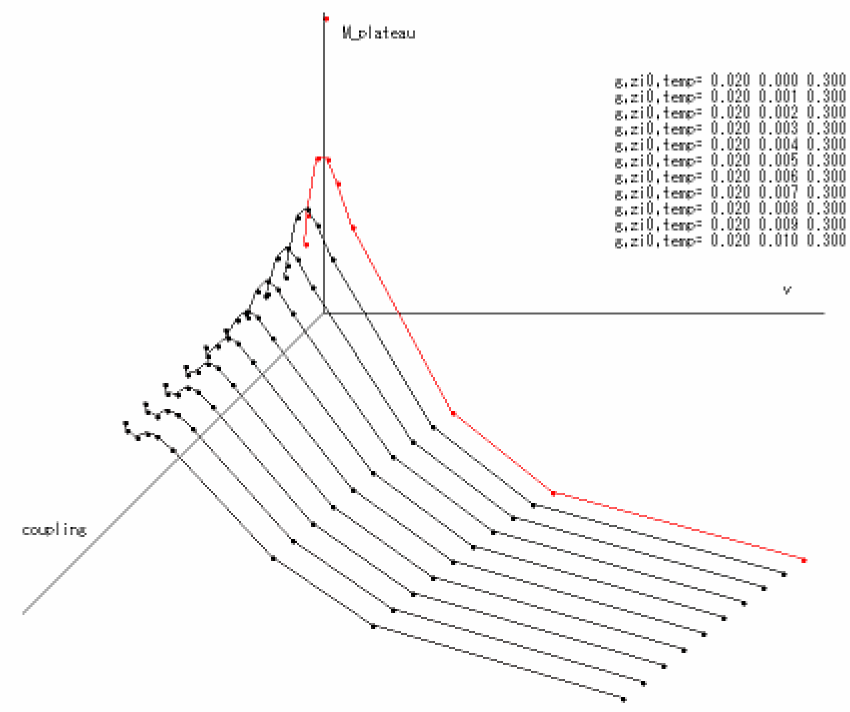
itime0, iout, maxtime=     275000        500   g,vh,zi0,temp=   0.02200   0.00080   0.00010   0.30000
itime0, iout, maxtime=     137500        250   g,vh,zi0,temp=   0.02200   0.00160   0.00010   0.30000
itime0, iout, maxtime=      68750        125   g,vh,zi0,temp=   0.02200   0.00320   0.00010   0.30000
itime0, iout, maxtime=      34348         62   g,vh,zi0,temp=   0.02200   0.00640   0.00010   0.30000
itime0, iout, maxtime=      17174         31   g,vh,zi0,temp=   0.02200   0.01280   0.00010   0.30000
itime0, iout, maxtime=       8580         15   g,vh,zi0,temp=   0.02200   0.02560   0.00010   0.30000
itime0, iout, maxtime=       4291          7   g,vh,zi0,temp=   0.02200   0.05120   0.00010   0.30000
itime0, iout, maxtime=       2148          3         9765
```




M_plateau
g,zi0,temp= 0.020 0.000 0.300
g,zi0,temp= 0.020 0.001 0.300
g,zi0,temp= 0.020 0.002 0.300
g,zi0,temp= 0.020 0.003 0.300
g,zi0,temp= 0.020 0.004 0.300
g,zi0,temp= 0.020 0.005 0.300
g,zi0,temp= 0.020 0.006 0.300
g,zi0,temp= 0.020 0.007 0.300
g,zi0,temp= 0.020 0.008 0.300
g,zi0,temp= 0.020 0.009 0.300
g,zi0,temp= 0.020 0.010 0.300
v
coupling

Diagonalization of L

$$\frac{\partial}{\partial t}\rho = \frac{1}{i\hbar}[H,\rho] - \gamma\left([X, R(t), \rho] + [X, R(t), \rho]^{+}\right)$$

matrix $\rho(i, j)$, $(i, j = 1, \cdots N)$

$$\Downarrow$$

vector $\vec{\rho}$, $(\rho(k), k = 1, \cdots N^2)$

$$\frac{\partial}{\partial t}\vec{\rho}(t) = L\,\vec{\rho}(t) \qquad \vec{\rho}(t) = \mathrm{e}^{iLt}\,\vec{\rho}(0)$$

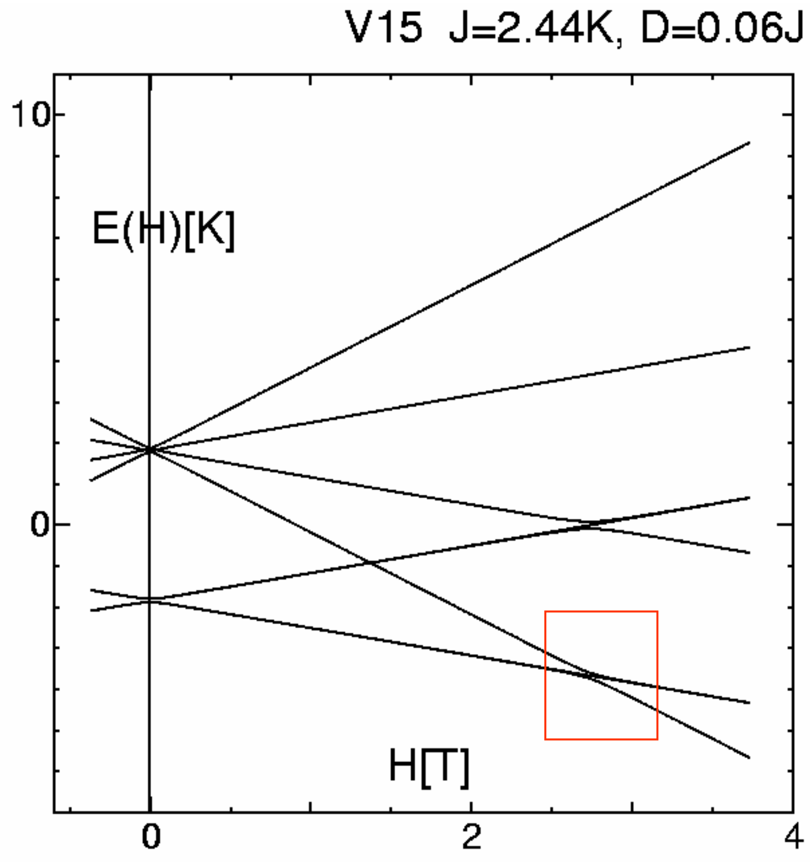
$$\vec{\rho}(t) = \prod_{k} \mathrm{e}^{iL_k t}\,\vec{\rho}(0)$$

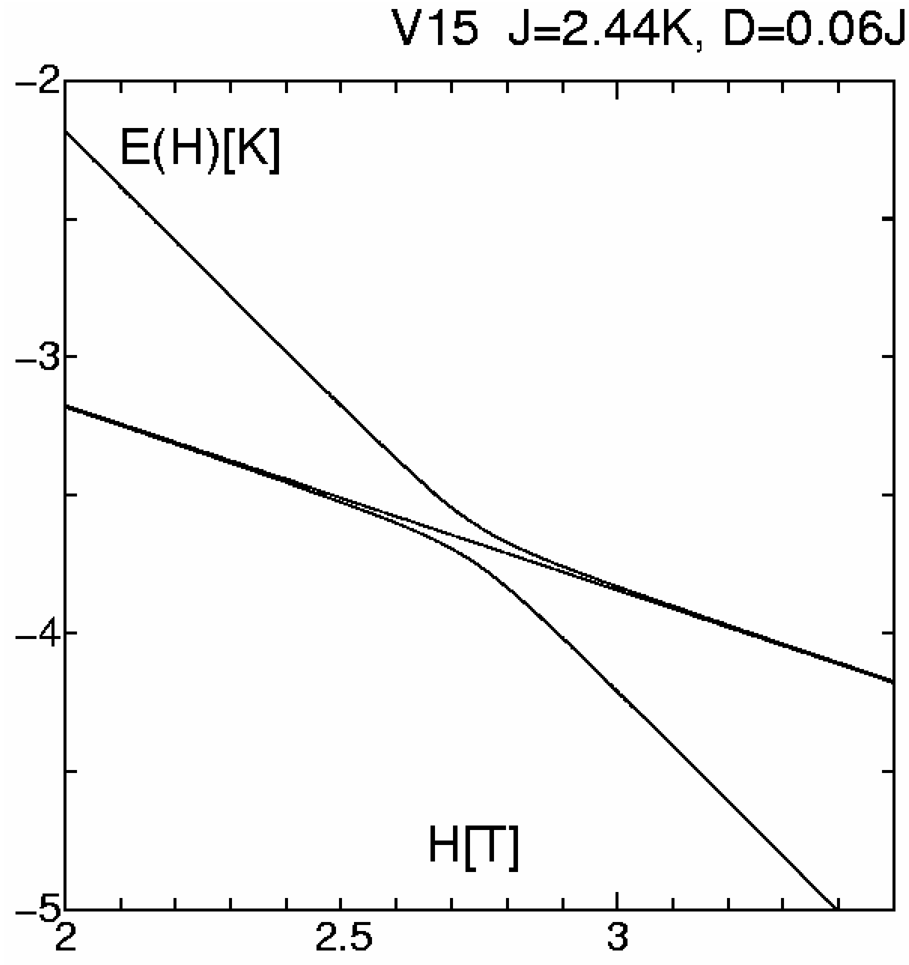
Field changes little in an interval. $H(t) \cong H(t_0)$

The spin evolves a lot in the interval (many precession).

Effects of doubly degenerate structure

Transition from 1/2 ⟺ 3/2





Smooth magnetization process in the ground state

cf. Heisenberg spin models:

Limit of N infinity

Continuous energy levels vs. Gap

Finite system (steps like magnetization in the Heisenberg model, where [H,Mz]=0)

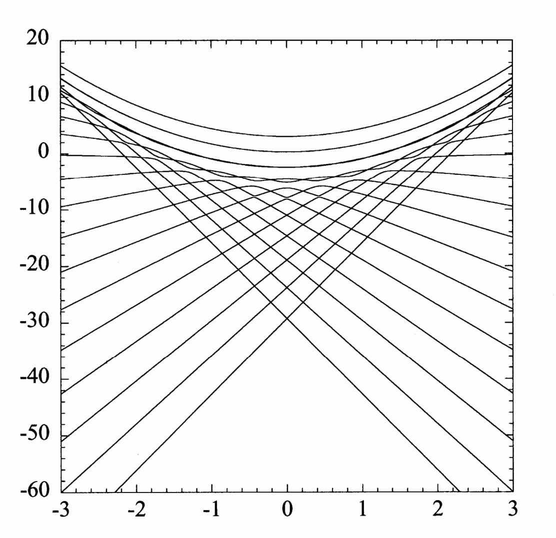
Adiabatic change: smooth magnetization process at T=0

Some mixing term (quantum fluctuation)

Origin of the adiabatic change

$$[H, M_z] \neq 0$$

S: even Large S (S=10) Mn12, Fe8



$$H = -D(S^z)^2 - hS^z$$
$$+ E((S^x)^2 - (S^y)^2)$$
$$+ C((S^+)^4 - (S^-)^4) + \text{etc.}$$

S: odd (S=1/2) V15 No anisotropy & Kramers doublet

Extra-degeneracy + Dzyloshinskii-Moriya interaction

SM, &. N. Nagaosa, Prog. Theor. Phys. 106 (2001) 533

Dzyaloshinskii-Moriya interaction

- V15
- Fe-rings Fe10,etc.
- Cu-Benzoate (1DH)
- SrCu2(BO3)2 (SS)

$$H_{ij} = \sum_{\alpha,\beta} S_i^{\alpha} A_{ij}^{\alpha\beta} S_j^{\beta} = \sum_{\alpha} J_{ij}^{\alpha} S_i^{\alpha} S_j^{\alpha} + \vec{D}_{ij} \left(\vec{S}_i \times \vec{S}_j \right)$$

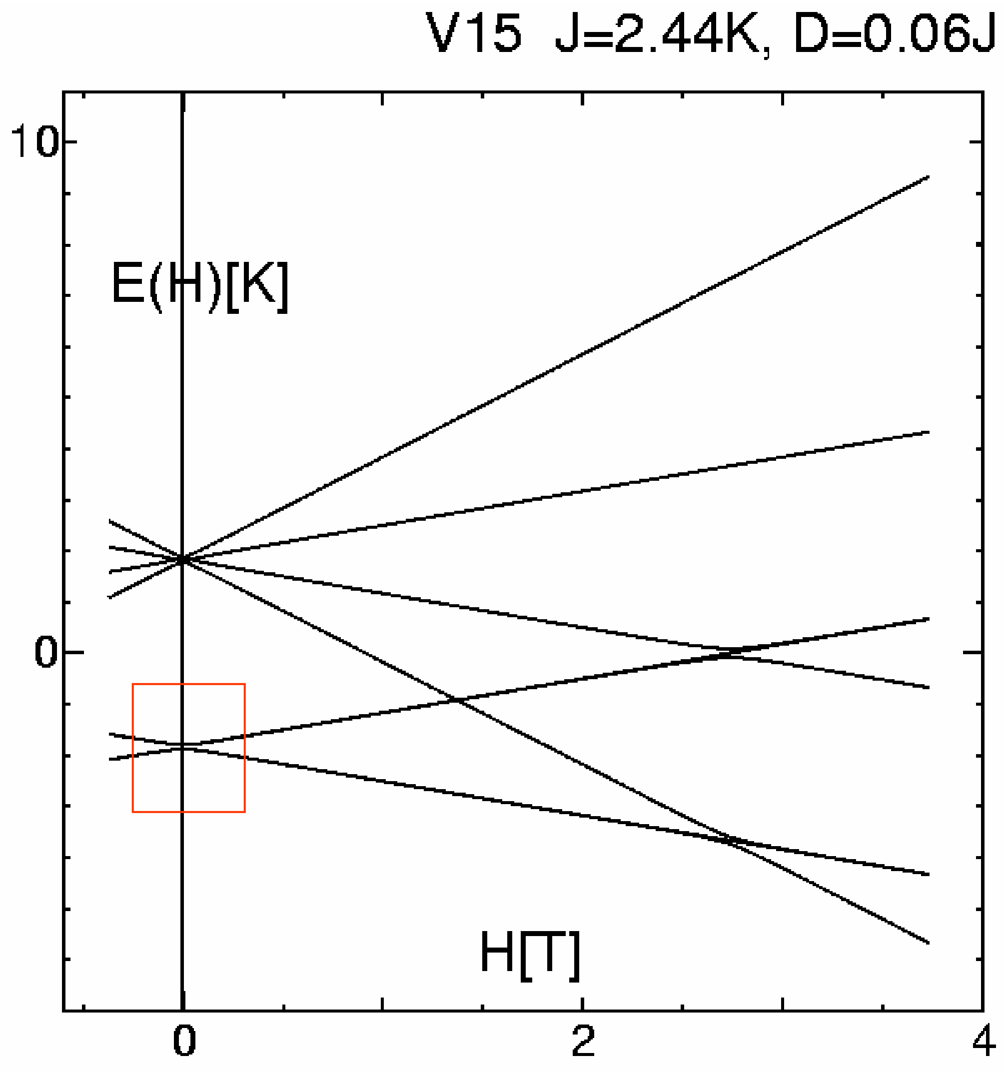
symmetric part $A_{ij}^{\alpha\beta} + A_{ij}^{\beta\alpha}$

asymmetric part $A_{ij}^{\alpha\beta} - A_{ij}^{\beta\alpha}$

cf.

transverse field $H_x S_x$

DM interaction



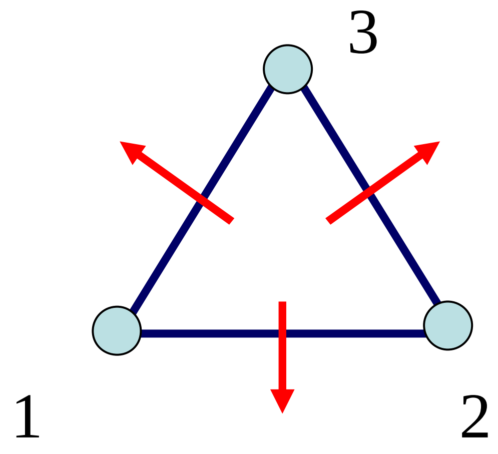
S: odd (S=1/2) V15
Kramers doublet
No tunneling?

Extra-degeneracy +
Dzyloshinskii-Moriya
interaction

SM, &. N. Nagaosa,
. Theor. Phys. 106 (2001) 533

Anisotropy of DM interaction

DM interaction on a triangle lattice



C_3symmetry (axis //z)

1. $$\left(\sum_{ij} \vec{D}_{ij}\right) \times \vec{z} = 0,$$

2. $\vec{D}_{ij} \cdot \vec{z}$ is the same for all ij

No adiabatic change if $\vec{h} // \vec{z}$

Decoupling of states

$$H = H_0 + D_{12}\left(S_1^z S_2^x - S_1^x S_2^z\right) + D_{23}\left(S_2^z S_3^x - S_2^x S_3^z\right) + D_{31}\left(S_3^z S_1^x - S_3^x S_1^z\right)$$
$$+ d_{12}\left(S_1^y S_2^z - S_1^z S_2^y\right) + d_{23}\left(S_2^y S_3^z - S_2^z S_3^y\right) + d_{31}\left(S_3^y S_1^z - S_3^z S_1^y\right)$$

$$H_{\mathrm{DM}}\left|+++\right\rangle = \left(x_{31} - x_{12}\right)\left|-++\right\rangle + \left(x_{12} - x_{23}\right)\left|+-+\right\rangle + \left(x_{23} - x_{31}\right)\left|++-\right\rangle$$

$$x_{ij} = D_{ij} - \mathrm{i}d_{ij}$$

$$H_{\mathrm{DM}}\left|++-\right\rangle = \left(x_{12} + x_{23}\right)\left|+--\right\rangle - \left(x_{12} + x_{31}\right)\left|-+-\right\rangle + \left(\bar{x}_{23} - \bar{x}_{31}\right)\left|+++\right\rangle$$

$$H_{\mathrm{DM}}\left|+-+\right\rangle = \left(x_{12} + x_{31}\right)\left|--+\right\rangle - \left(x_{23} + x_{31}\right)\left|+--\right\rangle + \left(\bar{x}_{12} - \bar{x}_{23}\right)\left|+++\right\rangle$$

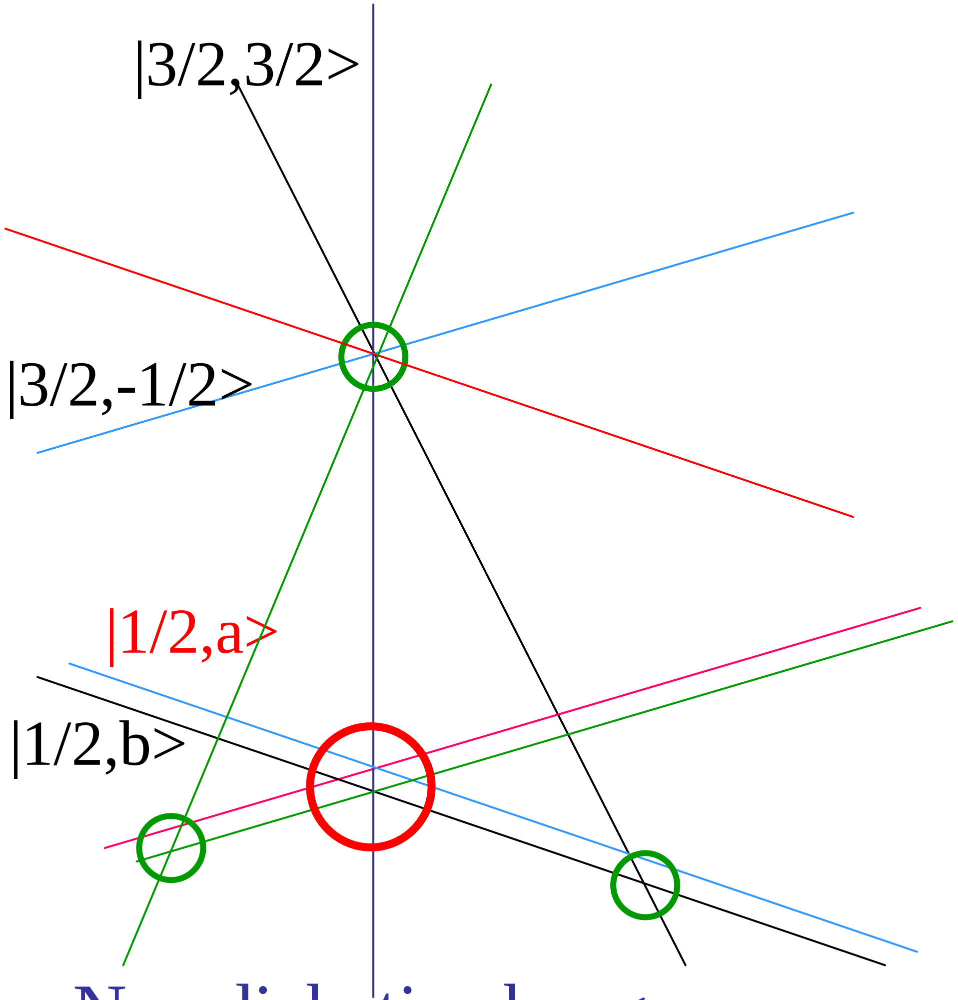
$$H_{\mathrm{DM}}\left|-++\right\rangle = \left(x_{23} + x_{31}\right)\left|-+-\right\rangle - \left(x_{12} + x_{23}\right)\left|--+\right\rangle + \left(\bar{x}_{31} - \bar{x}_{12}\right)\left|+++\right\rangle$$

$$H_{\mathrm{DM}}^2\left|+++\right\rangle = \left(2x_{23}^2 - 2x_{12}x_{31}\right)\left|+--\right\rangle + \left(2x_{31}^2 - 2x_{12}x_{23}\right)\left|-+-\right\rangle + \left(2x_{12}^2 - 2x_{23}x_{31}\right)\left|--+\right\rangle$$
$$+ \left(\left|x_{31} - x_{12}\right|^2 + \left|x_{12} - x_{23}\right|^2 + \left|x_{23} - x_{31}\right|^2\right)\left|+++\right\rangle$$

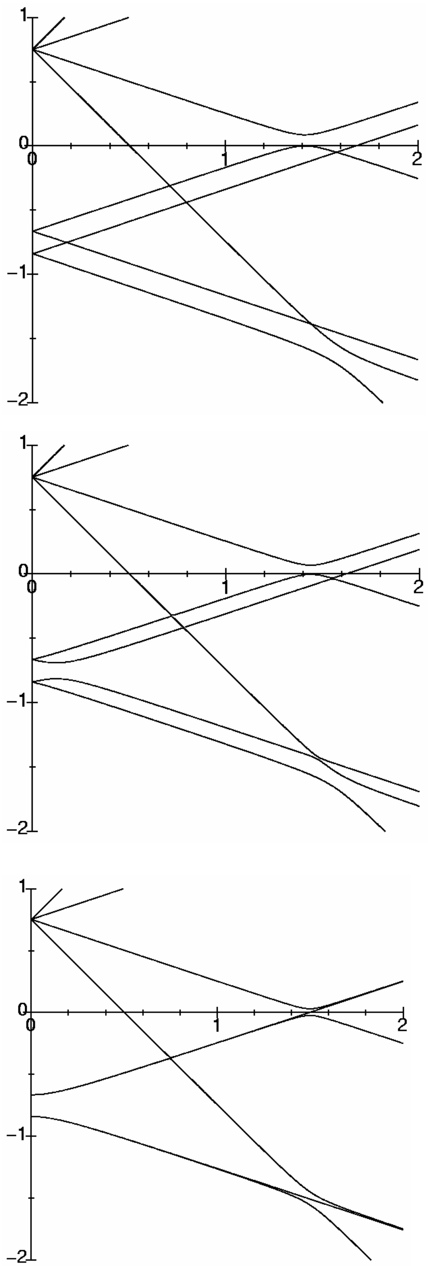
$$\text{if} \quad x_{12} + x_{23} + x_{31} = x_{12} + e^{i2\pi/3}x_{12} + e^{i4\pi/3}x_{12} = 0$$
$$\left(2x_{23}^2 - 2x_{12}x_{31}\right) = \left(2x_{31}^2 - 2x_{12}x_{23}\right) = \left(2x_{12}^2 - 2x_{23}x_{31}\right) = 0$$

$$\left|+++\right\rangle \Rightarrow \left|S = 1/2, M = 1/2\right\rangle \equiv \left|a\right\rangle \Rightarrow \gamma\left|+++\right\rangle$$
$$\left|---\right\rangle \Rightarrow \left|S = 1/2, M = -1/2\right\rangle \equiv \left|b\right\rangle \Rightarrow \gamma\left|---\right\rangle$$
$$\left|S = 3/2, M = -1/2\right\rangle \Rightarrow \left|S = 1/2, M = 1/2\right\rangle \equiv \left|b\right\rangle \qquad \left\langle a\middle|b\right\rangle = 0$$
$$\left|S = 3/2, M = 1/2\right\rangle \Rightarrow \left|S = 1/2, M = -1/2\right\rangle \equiv \left|b'\right\rangle \qquad \left\langle a'\middle|b'\right\rangle = 0$$

Energy structure with DM



No adiabatic change at H=0



$\theta = 0^\circ$

$\theta = 45^\circ$

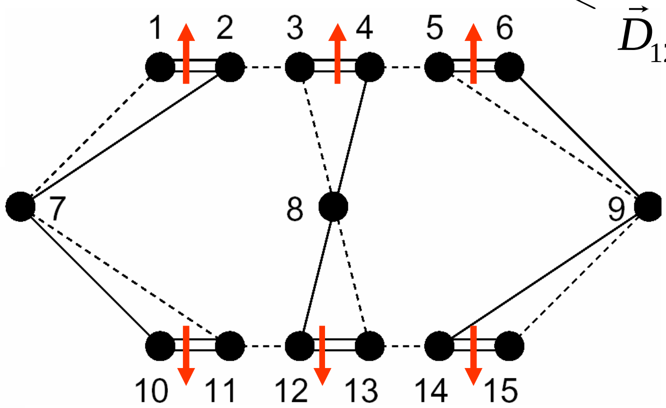
$\theta = 90^\circ$

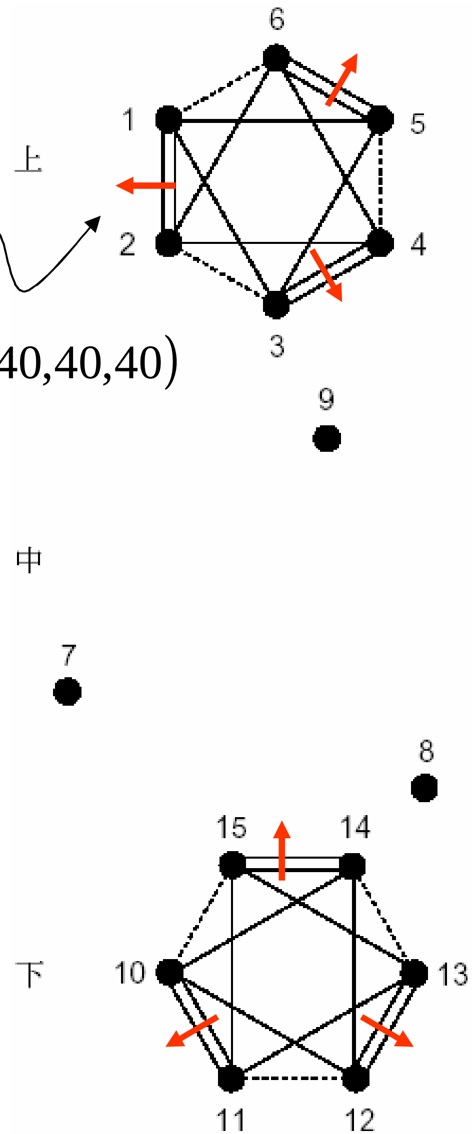
V15 structure

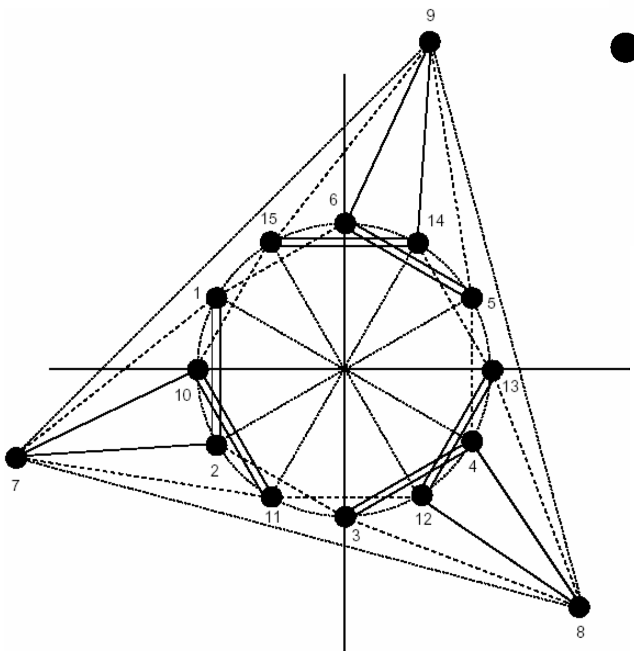
$$\vec{H}_s \perp c$$

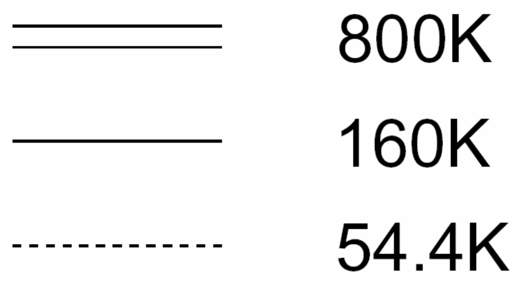
$$H = \sum_{\langle ij \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j + \sum_{\langle ij \rangle} \vec{D}_{ij} \cdot \left(\vec{S}_i \times \vec{S}_j \right) - H_s \sum_i S_i^x$$

$$\vec{D}_{12} = (40,40,40)$$

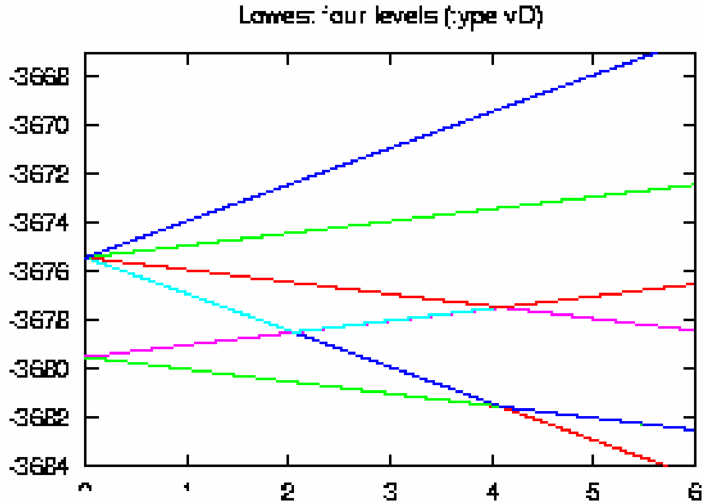


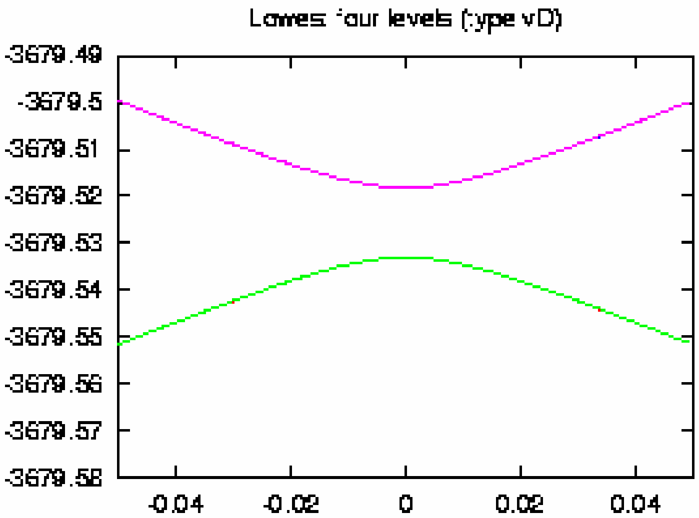


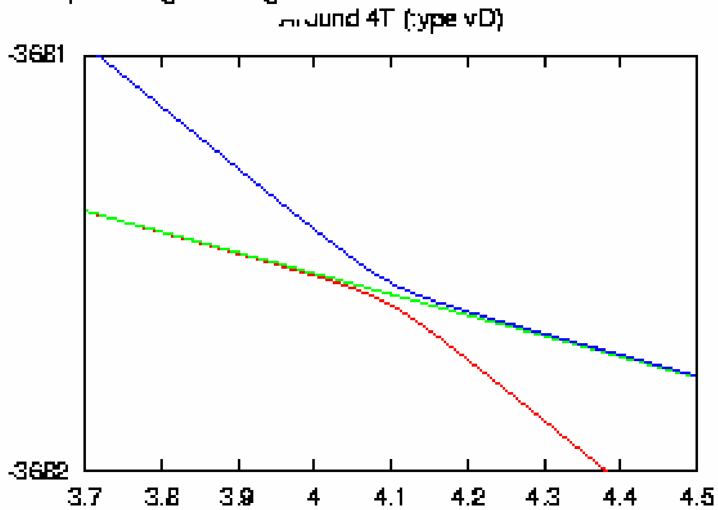




Anisotropy of the Gap: Hard axis





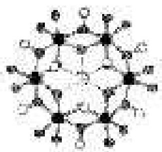


I.Chiorescu, W. Wernsdorfer, A. Mueller, SM, and B. Barbara:
PRB 67 (2003) 020402

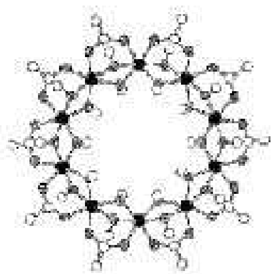
Fe ring

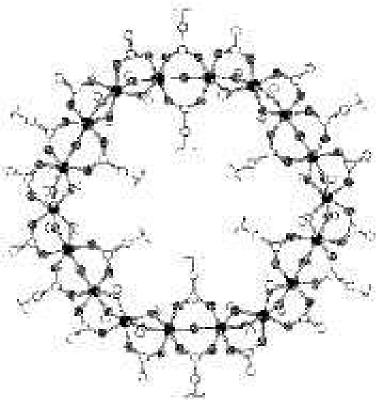
Fe^{3+} plays a role of a localized spin with S=5/2 and L=0.

n=6

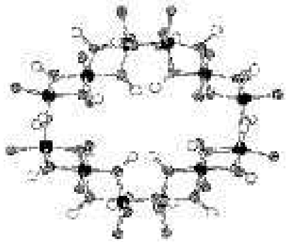


n=10

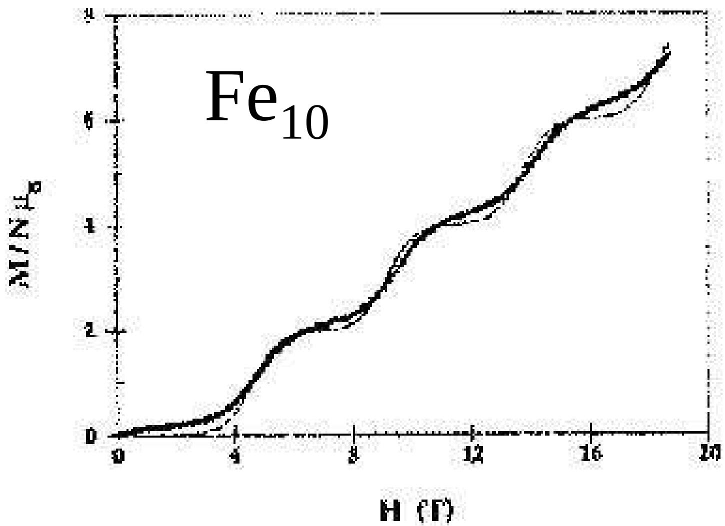


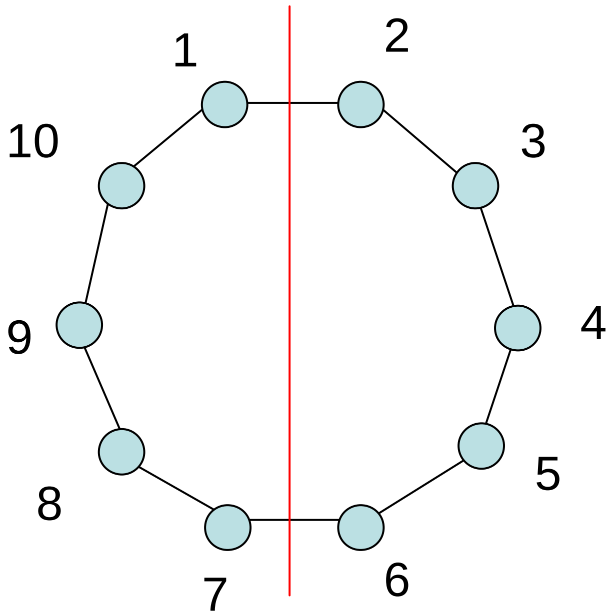


n=18



n=12





Fe10

$$\sum_{\alpha ij} D_\alpha \left(S_i^\beta S_j^\gamma - S_i^\gamma S_j^\beta \right)$$

C_{10} +Reflection ➡ S_{10}

Mirror symmetry

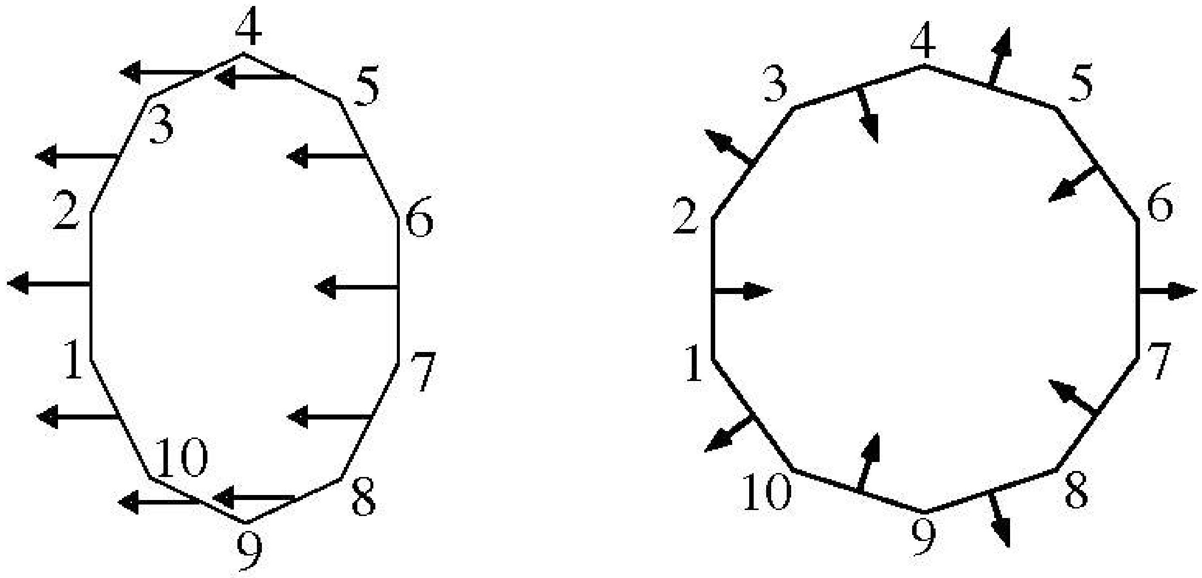
$$\begin{pmatrix} S_5^x \\ S_5^y \\ S_5^z \end{pmatrix} \to \begin{pmatrix} -S_8^x \\ S_8^y \\ -S_8^z \end{pmatrix}, \quad \begin{pmatrix} S_6^x \\ S_6^y \\ S_6^z \end{pmatrix} \to \begin{pmatrix} -S_7^x \\ S_7^y \\ -S_7^z \end{pmatrix}. \qquad (13)$$

These transformations give

$$\begin{aligned} & \boldsymbol{d}_{56} \cdot (\boldsymbol{S}_5 \times \boldsymbol{S}_6) \\ = \quad & d_{56}^x (S_5^y S_6^z - S_5^z S_6^y) + d_{56}^y (S_5^z S_6^x - S_5^x S_6^z) + d_{56}^z (S_5^x S_6^y - S_5^y S_6^x) \\ \to \quad & d_{56}^x (-S_8^y S_7^z + S_8^z S_7^y) + d_{56}^y (S_8^z S_7^x - S_8^x S_7^z) + d_{56}^z (-S_8^x S_7^y + S_8^y S_7^x) \\ = \quad & d_{56}^x (S_7^y S_8^z - S_7^z S_8^y) - d_{56}^y (S_7^z S_8^x - S_7^x S_8^z) + d_{56}^z (S_7^x S_8^y - S_7^y S_8^x). \qquad (14) \end{aligned}$$

Thus, we obtain $d_{56}^x = d_{78}^x$, $d_{56}^y = -d_{78}^y$ and $d_{56}^z = d_{78}^z$. When one divide a $\boldsymbol{d}$ vector into a component parallel to the mirror plane and a component

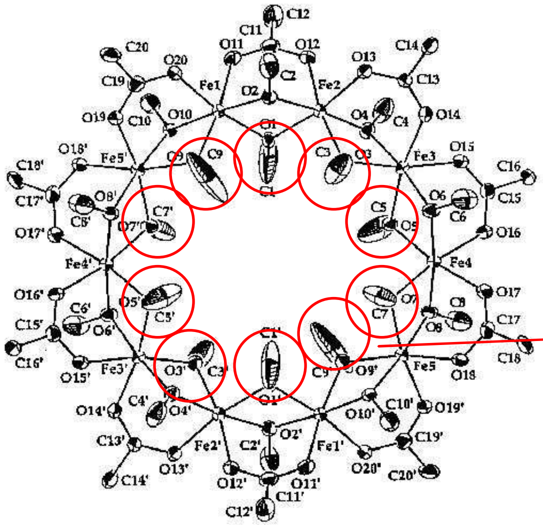
A set of *D* vectors from static regular structure



$$<\psi_M|\mathcal{H}_{\mathrm{DM}}|\psi_{M+1}>=0$$

The DM interaction of the above ***D*** vectors is not the origin of the peaks in dM/dH.

Oscillation of methyl groups



Structure is measured at T_{st}=226 K.

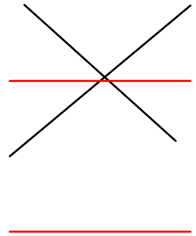
Each ellipsoid shows 50% possibility.

Oblong thermal ellipsoids
with the longer radius a

Elastic constant of an elastic energy of a methyl group is briefly estimated as $K \sim 0.67^2 k_B T_{st}/a^2$.

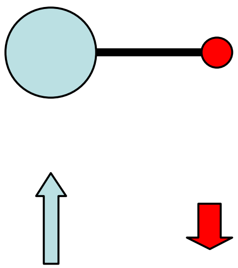
H. Nakano and SM: JPSJ 71 (2002) 2580

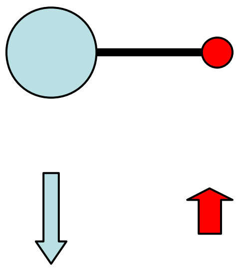
Change of state

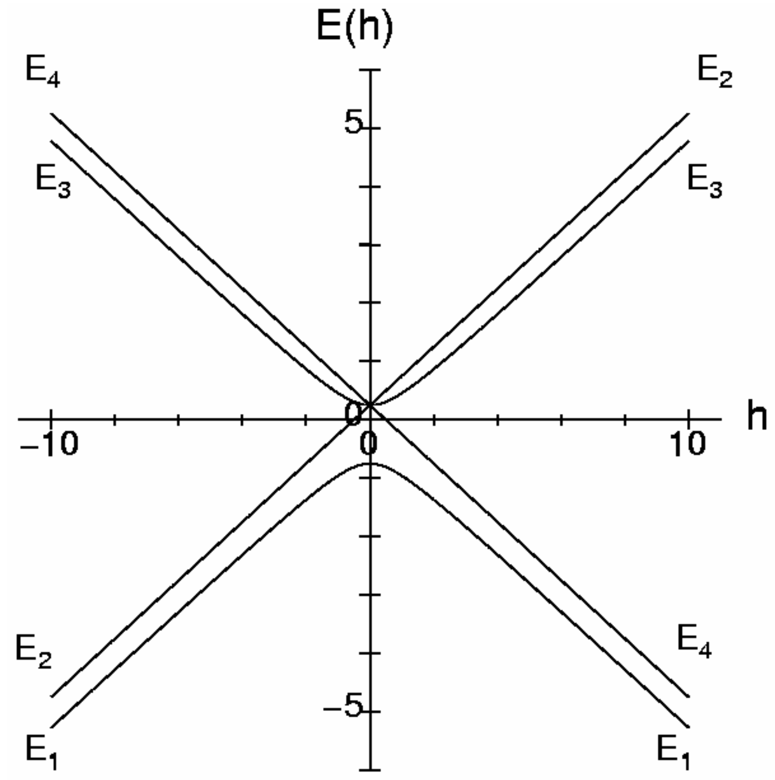


$$\begin{cases} |\pm\rangle = \dfrac{1}{\sqrt{8A^2 + 2\Delta^2 \pm 2\Delta\sqrt{\Delta^2 + 4A^2}}}\left(-2A|+-\rangle + (-\Delta \mp \sqrt{\Delta^2 + 4A^2}|-+\rangle\right) \\ |++\rangle \\ |--\rangle \\ \Delta = h - h' \end{cases}$$

$$\lim_{\Delta\to\infty}|\pm\rangle = |+-\rangle, \qquad \lim_{\Delta\to-\infty}|\pm\rangle = |-+\rangle$$

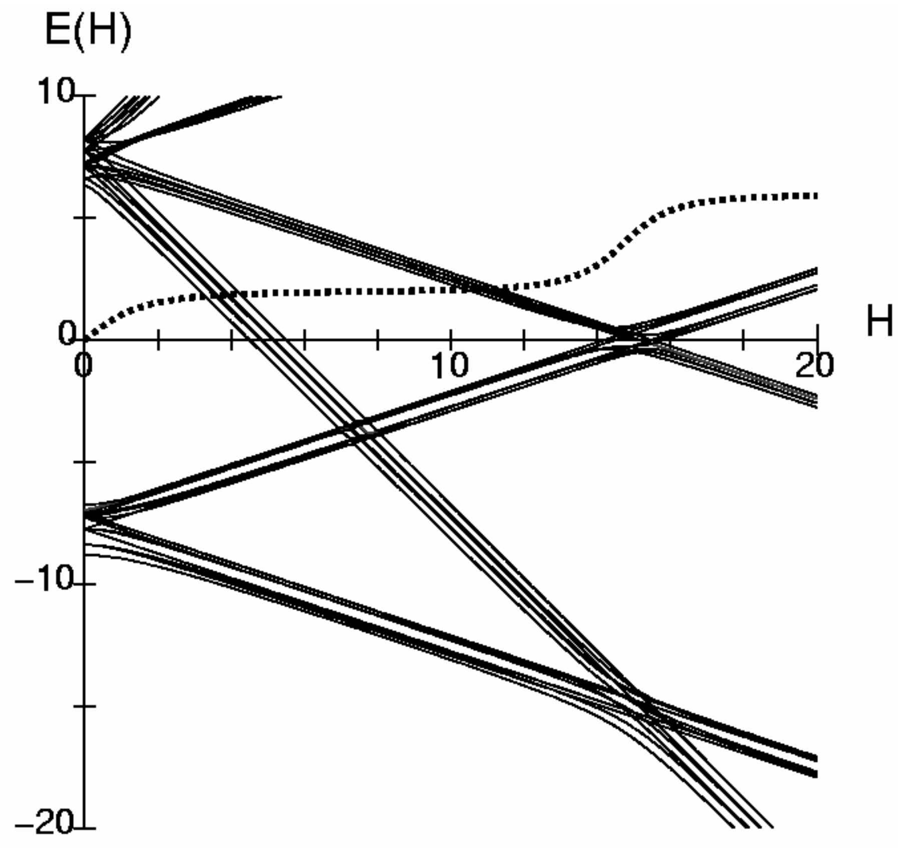


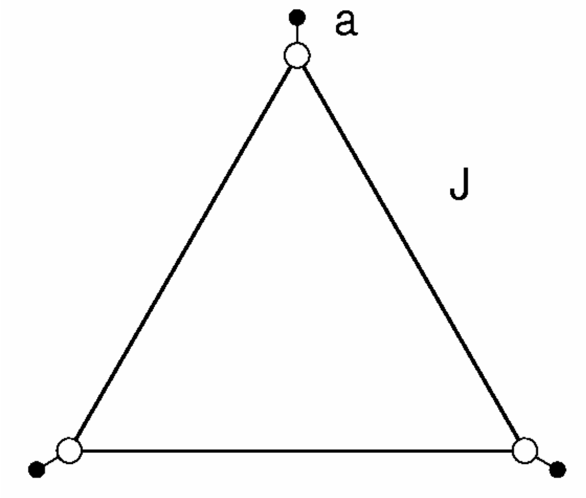


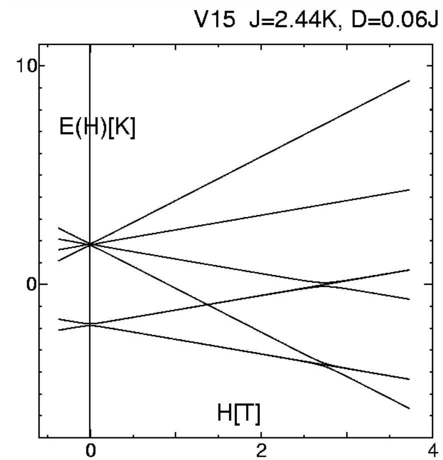


Triangle system

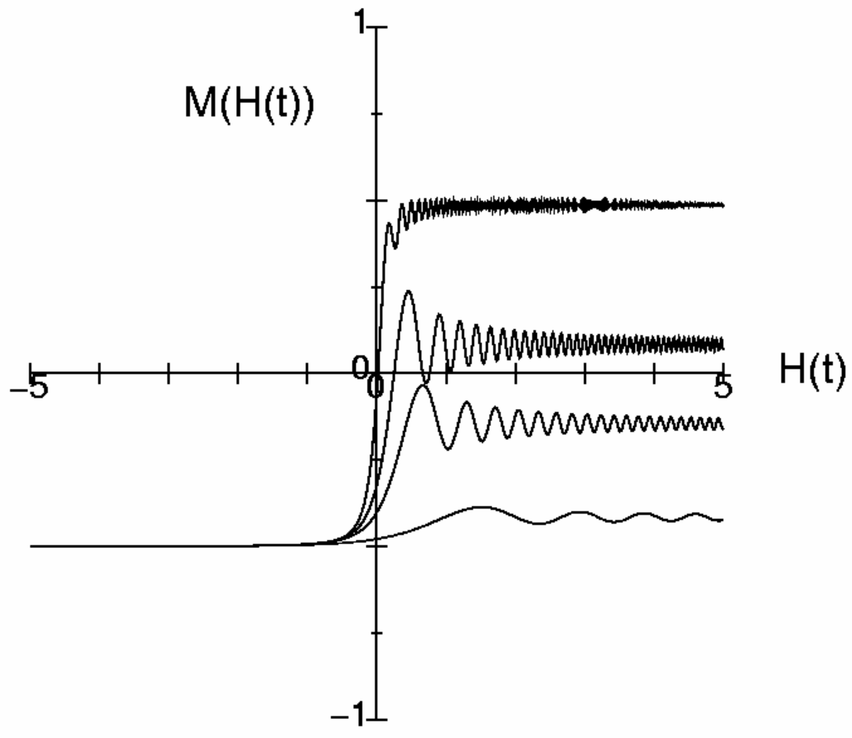
$$H = J\sum_{<ij>} \vec{S}_i \cdot \vec{S}_j + \sum_i A\vec{S}_i \cdot \vec{\sigma}_i$$
$$-\sum_i \left(g\mu_B \vec{S}_i \cdot \vec{h} + g_N \mu_N \vec{S}_i \cdot \vec{h} \right)$$







M(t) from the ground state



$$P = \left|\left\langle G(H_0)\middle|\Psi(t_f)\right\rangle\right|^2$$

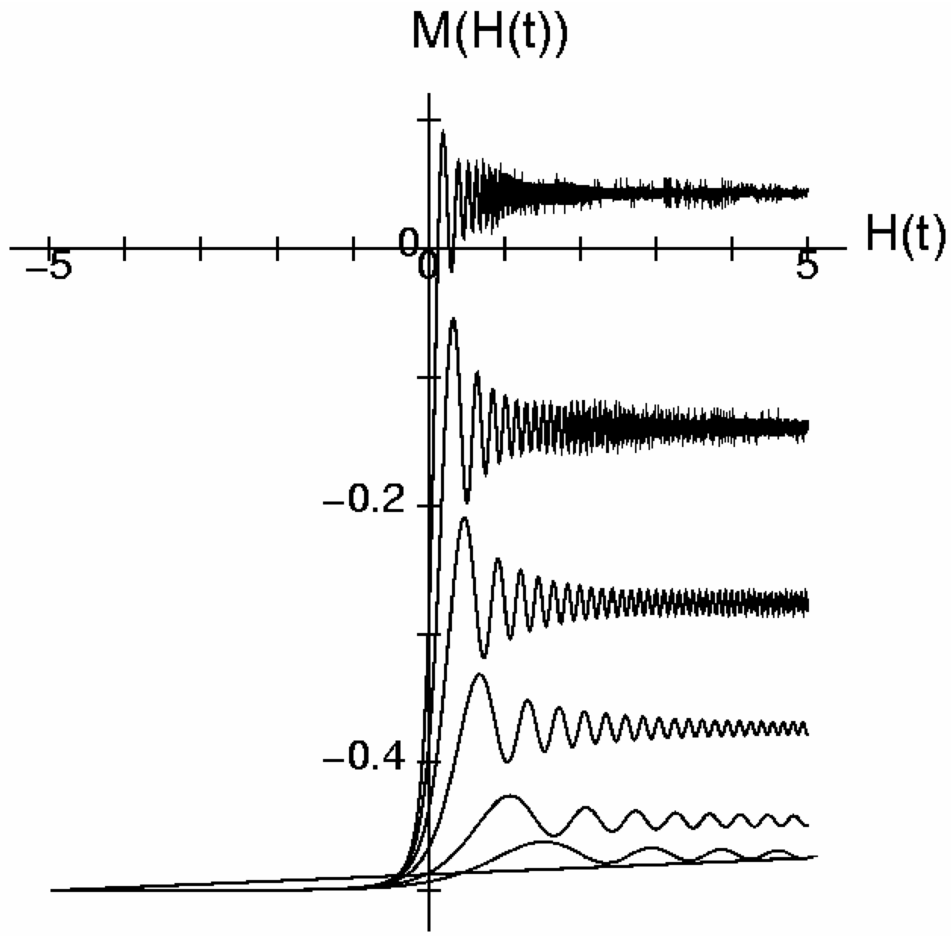
$$\Delta E = \sqrt{-2v\log(1-P)/\pi}$$

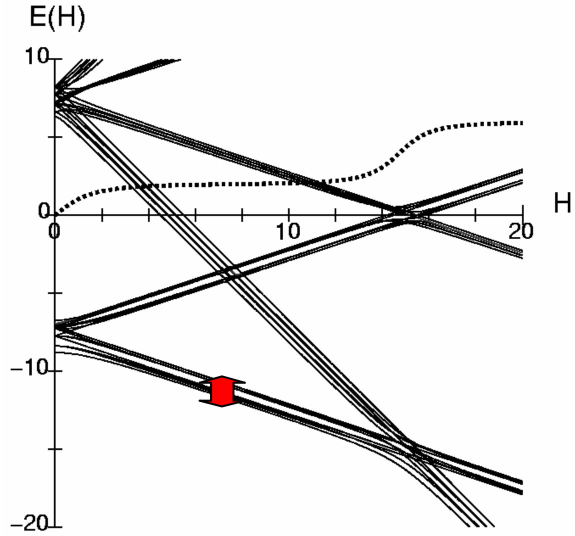
v	P	ΔE
0.500	0.08371	0.16681
0.250	0.16067	0.16697
0.100	0.35481	0.16702
0.050	0.58378	0.16704
0.025	0.82678	0.16704
0.010	0.98751	0.16704

$$P = 1-\exp\left(-\frac{\pi(\Delta E)^2}{2\hbar v}\right)$$

Apparent LZS relation

Finite temperature





$$m(t) = \frac{\sum m_i(t) e^{-\beta E_i}}{\sum e^{-\beta E_i}}$$

$$P_{\text{eff}} = \sum_{i=1}^{16} \left| \left\langle i(H_0) \middle| \Psi(t_f) \right\rangle \right|^2$$

Reaction to the nuclear spin

$$\left[\sum_i S_i^z + \sigma_i^z, H\right] = 0,$$

$$\text{c.f}\ \left[\sum_i g\mu_B S_i^z + g\mu_N \sigma_i^z, H\right] \neq 0,$$

$$(S_1, S_2, S_3, \sigma_1, \sigma_1, \sigma_1,)$$

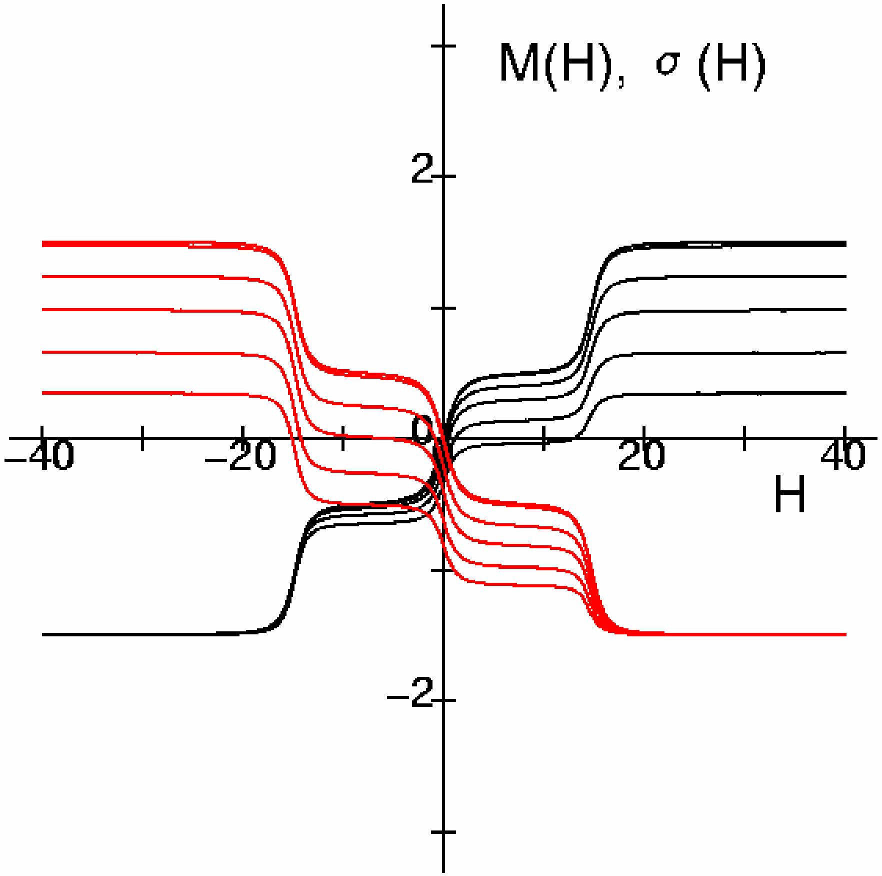
$$(------) \Rightarrow (------)$$

$$(---+--) \Rightarrow (+-----),$$

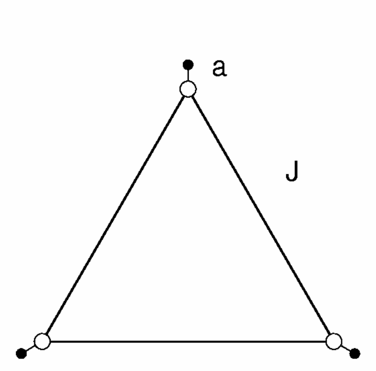
$$(---++-) \Rightarrow (++----),$$

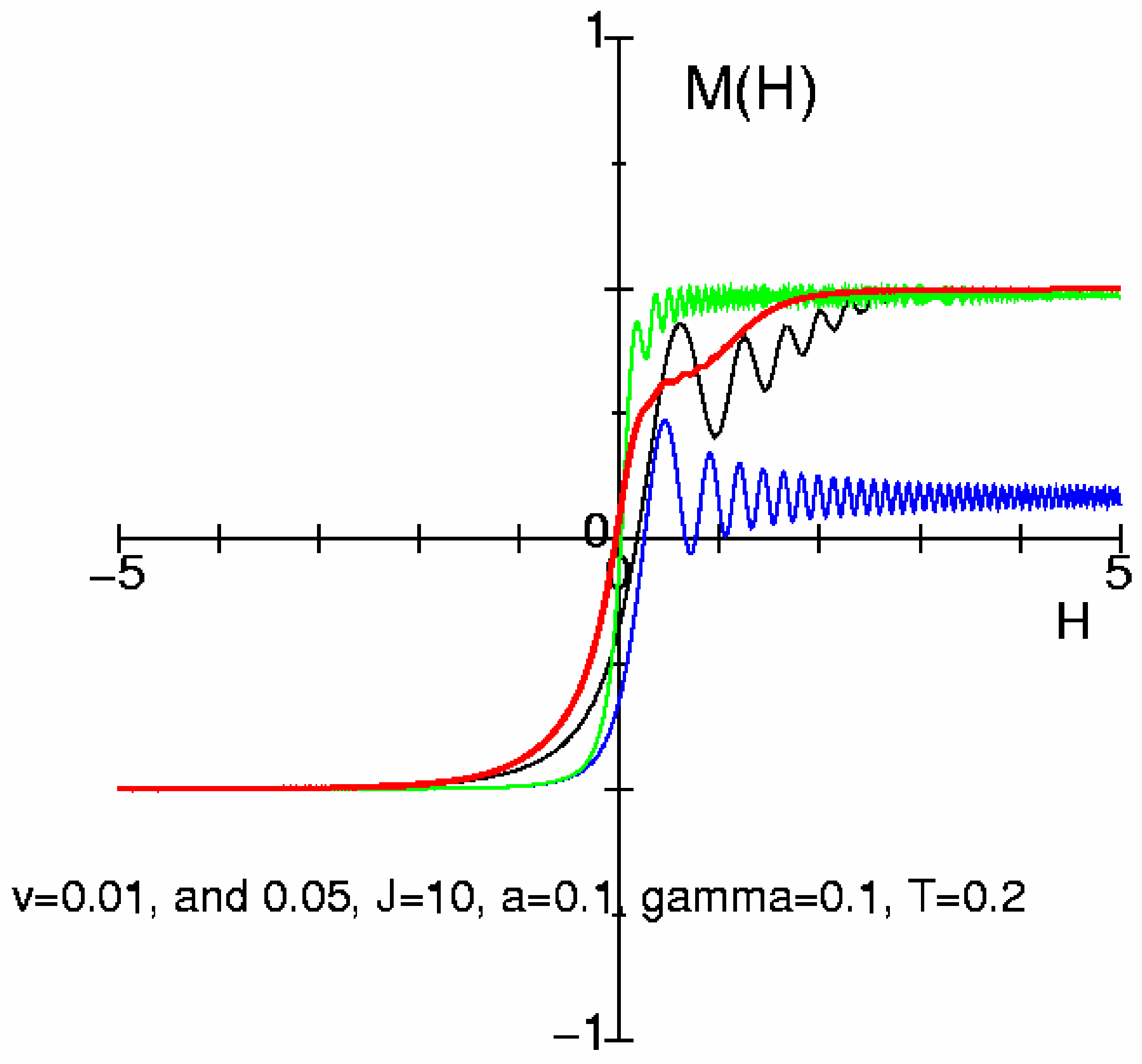
$$(---+++) \Rightarrow (+++---), \text{etc.}$$

$$\langle\sigma\rangle \approx 0 \Rightarrow \langle\sigma\rangle < 0$$



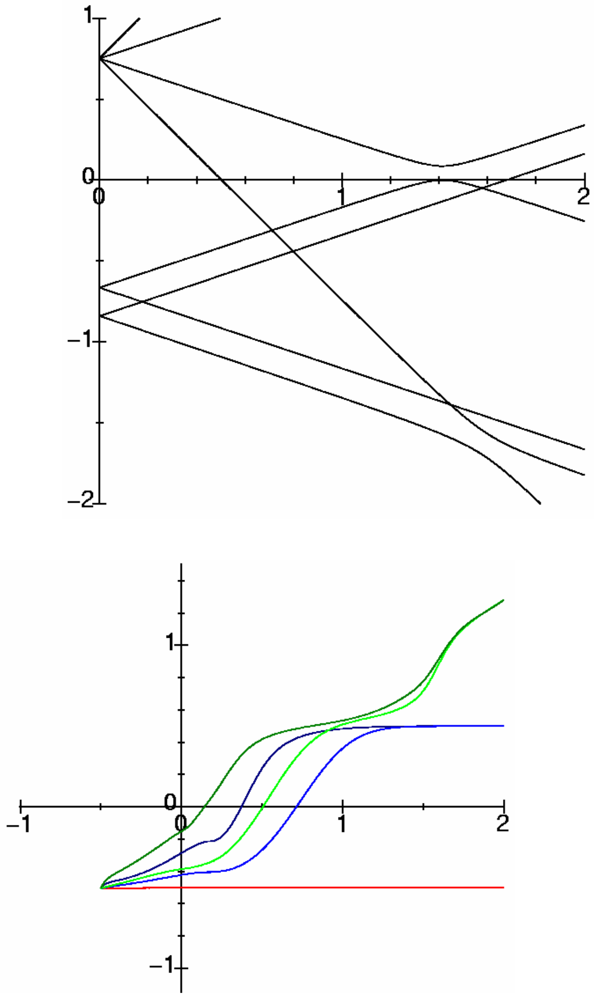
Effects of environments



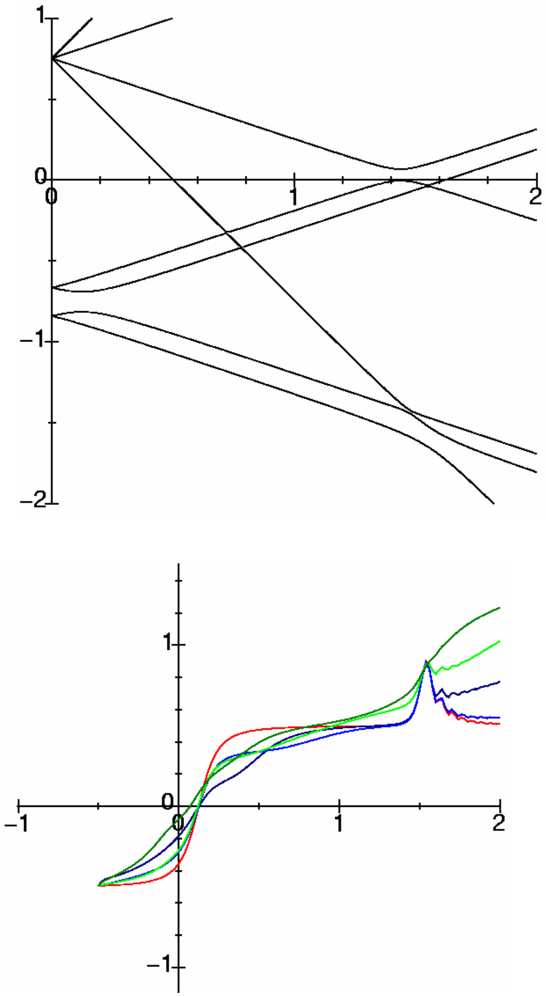


Angle dependence of the energy levels

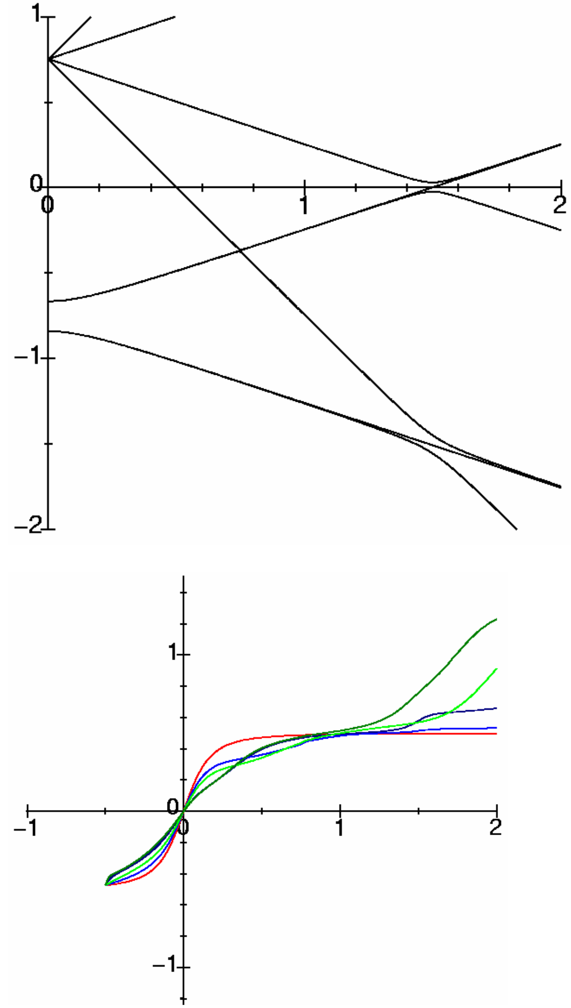
$\theta = 0^\circ$



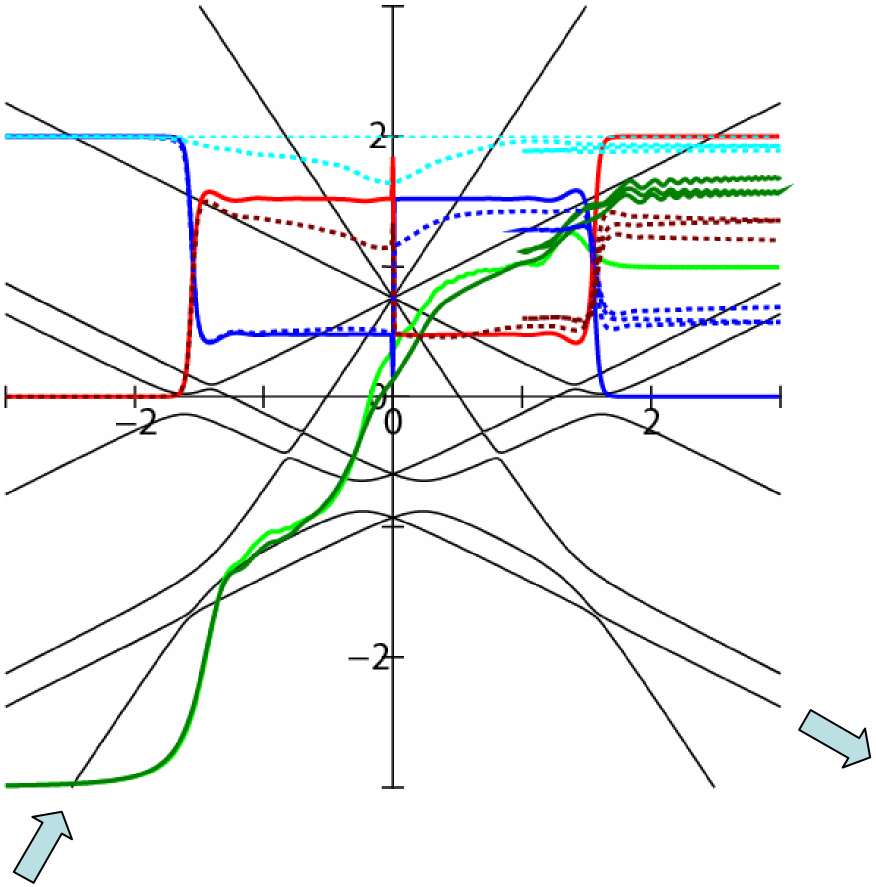
$\theta = 45^\circ$



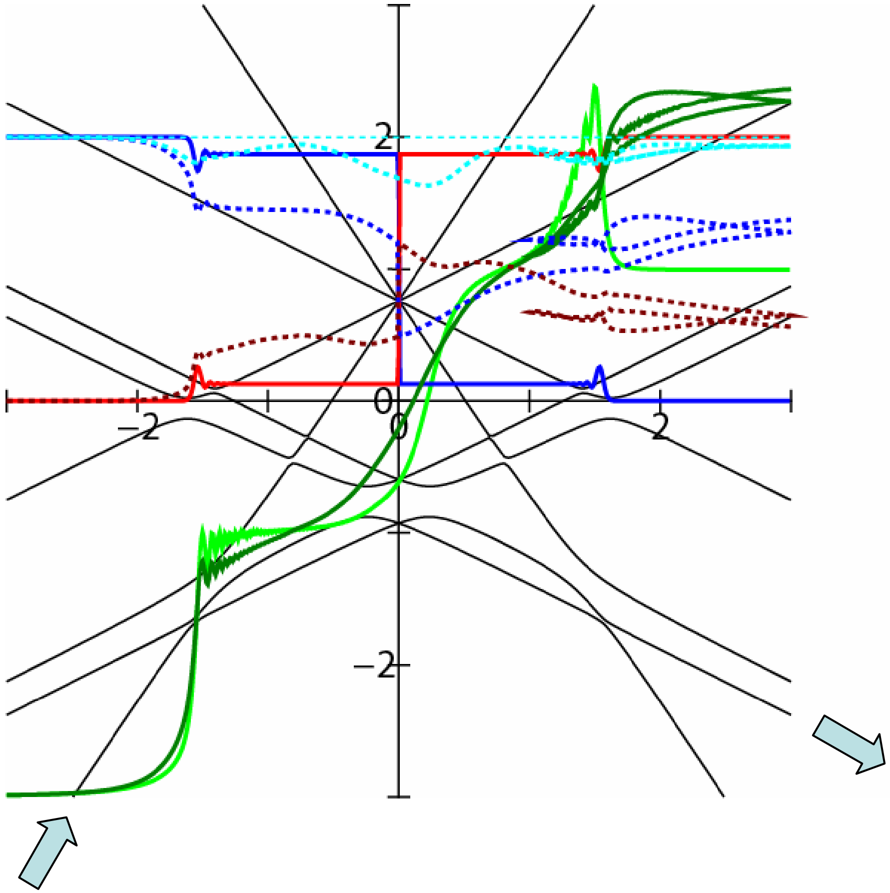
$\theta = 90^\circ$



Nontrivial coherence



V=0.01



V=0.001

Fluctuation-induced adiabatic transition

Temporal symmetry-breaking induced DM interaction

NaV_2O_5 : charge fluctuation reduces the symmetry
=> virtual DM ESR

Nojiri, et al.: JPSJ 69 (2000) 2291

Fe12 : configuration fluctuation reduces the symmetry
=> virtual DM M(H)

H. Nakano and SM: JPSJ 71 (2002) 2580

$SrCu_2(BO_3)_2$: configuration fluctuation reduces the symmetry => Raman,ESR

Cepas and Zimann cond-mat 0401240
SM & Ogasahara: JPSJ 72 (2003) 2350

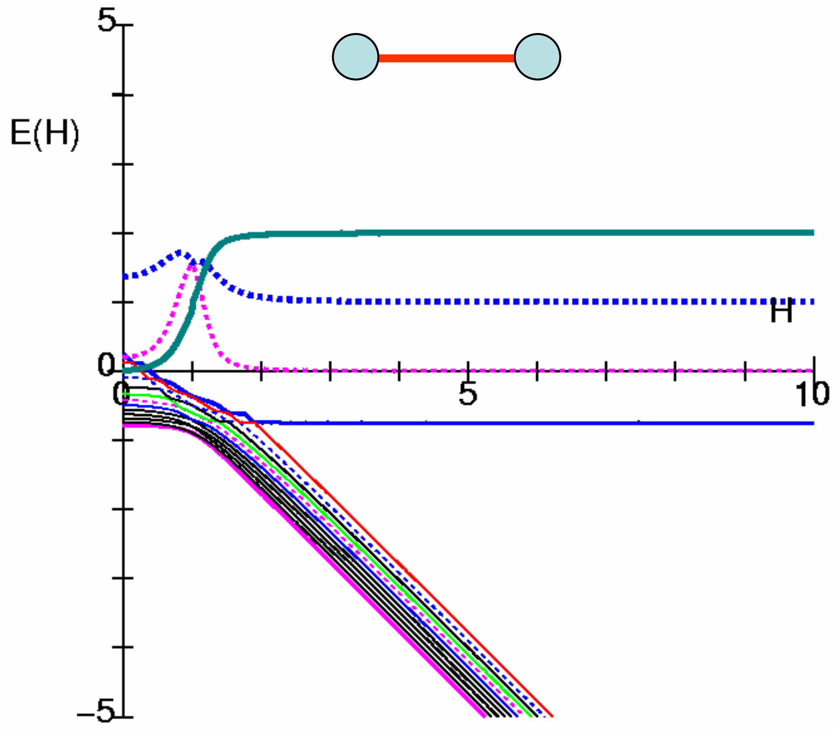
Charge transfer, Phonon,
Orbital degree of freedom, etc.

Fluctuation induced DM for a dimer

$$H = J\vec{S}_1\cdot\vec{S}_2 + \vec{d}\cdot\left(\vec{S}_1\times\vec{S}_2\right) + H\left(S_1^z + S_1^z\right) + \frac{k}{2}x^2 + \frac{1}{2m}p^2$$

$$\vec{d} = \vec{d}_0 x \qquad [x,p] = i\hbar \quad \langle x\rangle = 0$$

m=10,omega=0.1,D_x=0.1

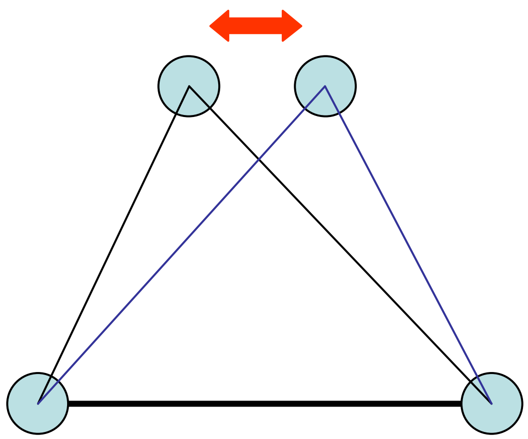


$$H = H_{\mathrm{Spin}} + H_{\mathrm{SP}} + H_{\mathrm{Phonon}}$$

$$\begin{cases} H_{\mathrm{Spin}} = J\vec{S}_1\cdot\vec{S}_2 \\ H_{\mathrm{SP}} = \sum_k \left(\alpha_k a_k^+ + \alpha_k^+ a_k\right)\vec{D}\cdot(\vec{S}_1\times\vec{S}_2) \\ H_{\mathrm{Phonon}} = \sum_k \omega_k a_k^+ a_k \end{cases}$$

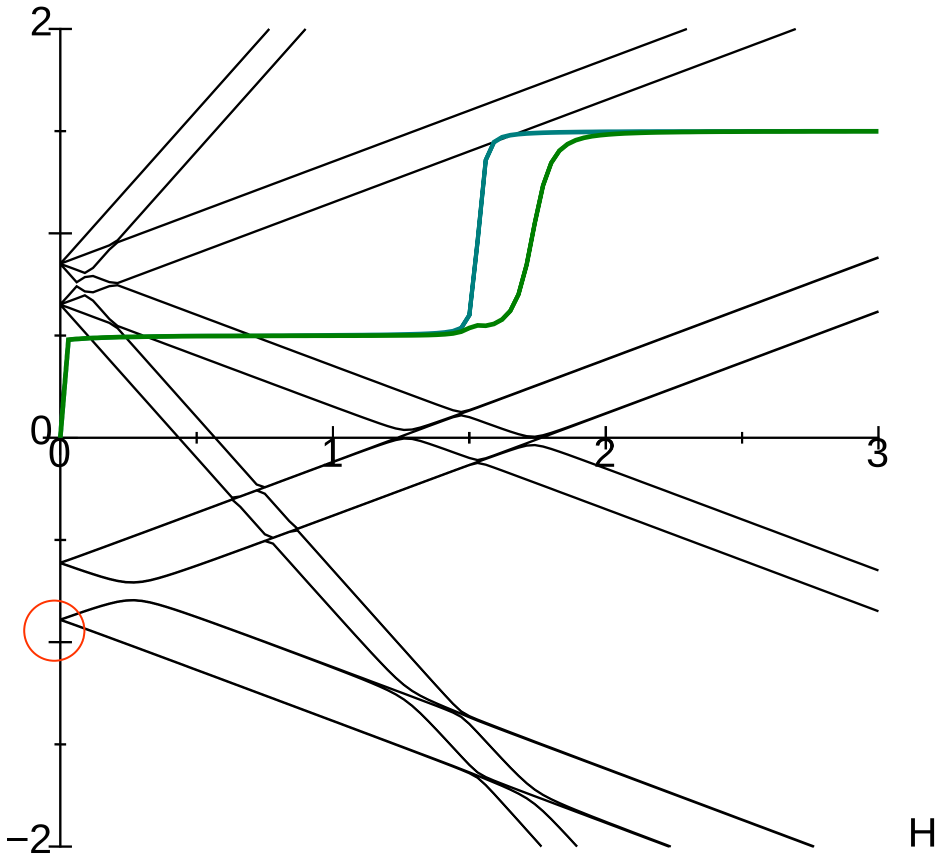
Effect of bond fluctuation
A minimal model

$$H = \sum_{<ij>} \left(J_{ij} + \sigma^z \Delta J_{ij} \right) \vec{S}_i \cdot \vec{S}_j + \sigma^z \sum_{<ij>} \vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j + a\sigma^x$$

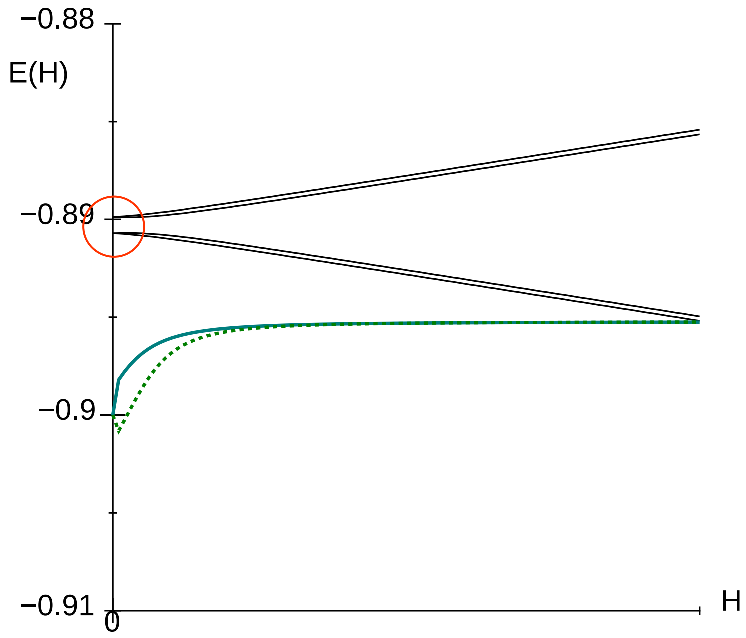


Small energy split at H=0

0.10 0.10 0.05 90



0.05 0.05 0.05 90



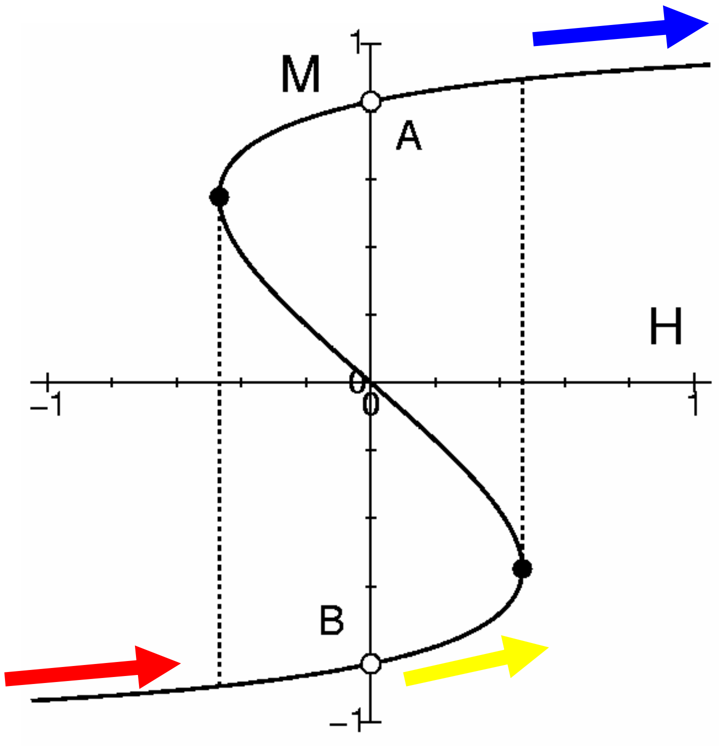
$\theta = 90°$

Quantum Switching

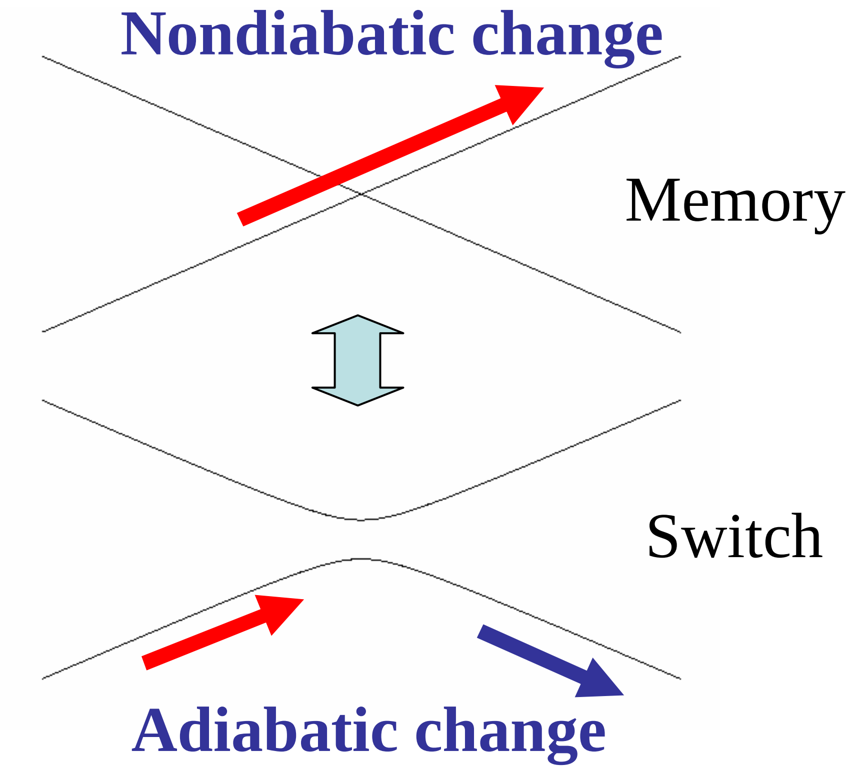
Memory：　2-values＋Metastability

Classical：Hysteresis

Dissipation



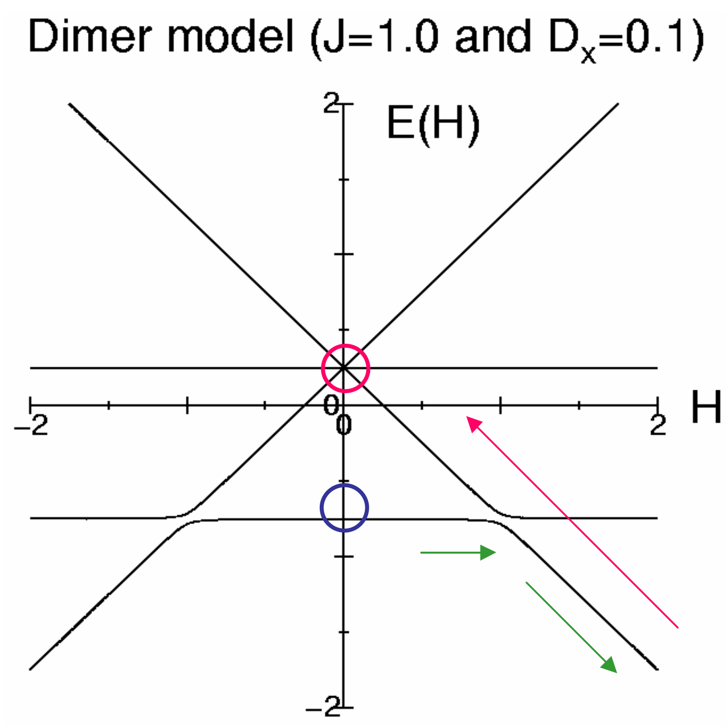
Quantum： Dynamics

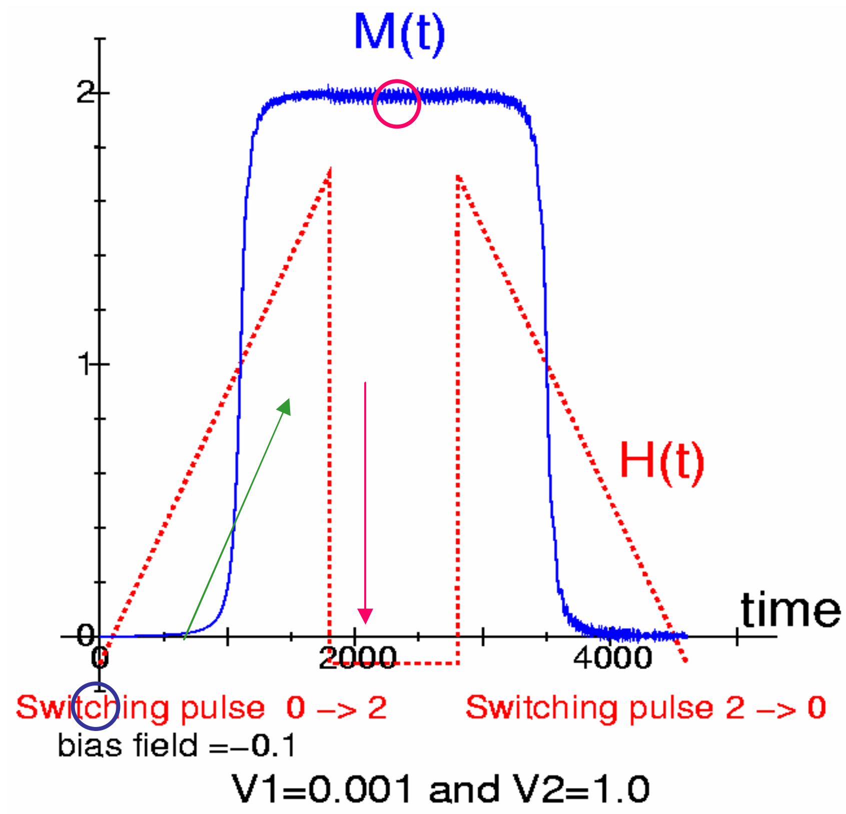


No Hysteresis loss

2. Sweeping velocity control

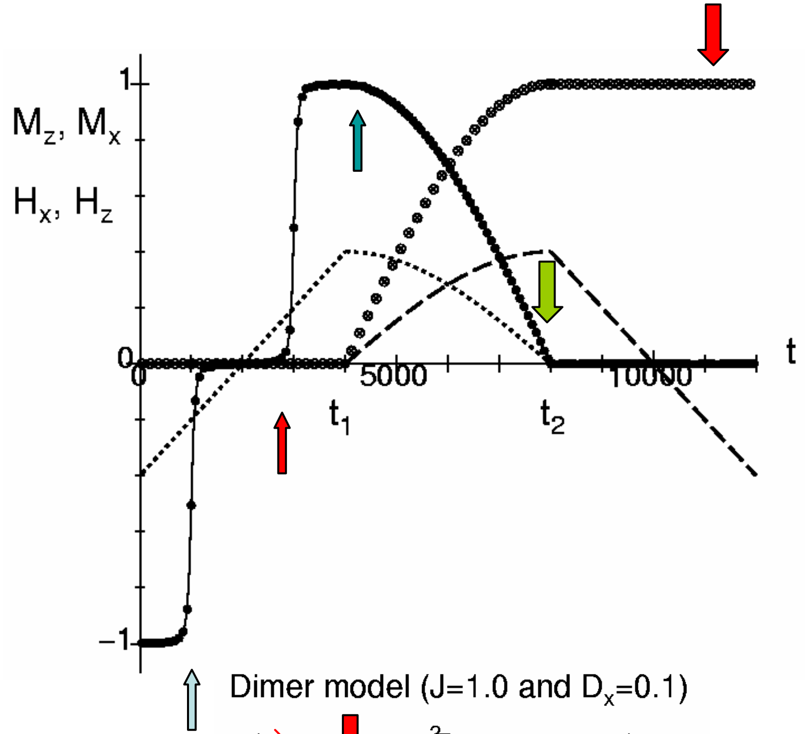
$$H = J\vec{S}_1 \cdot \vec{S}_2 - \vec{D} \cdot \vec{S}_1 \times \vec{S}_2 \qquad \vec{D} = (0.1,0,0)$$

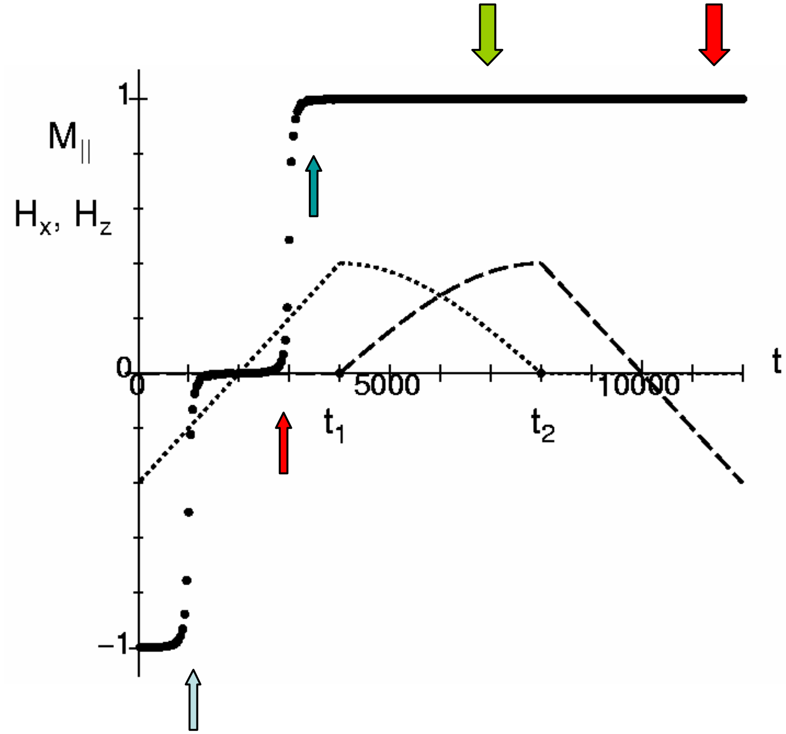




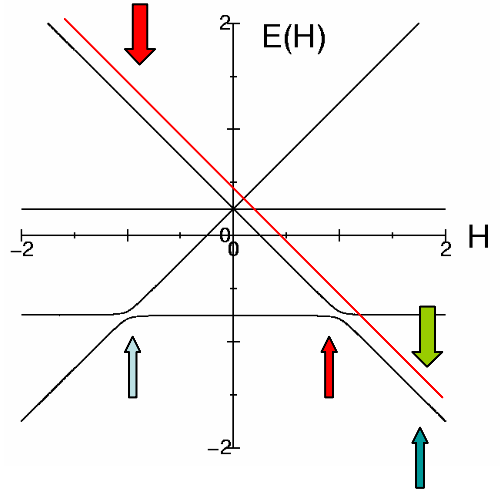
Switching between different S values

3.Rotation of the field (or sample)

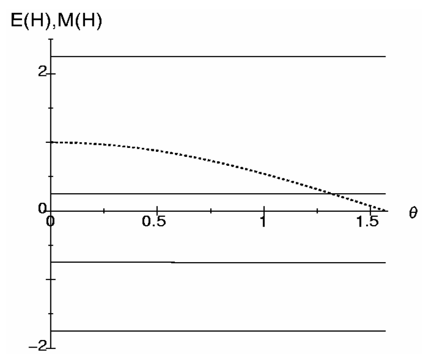




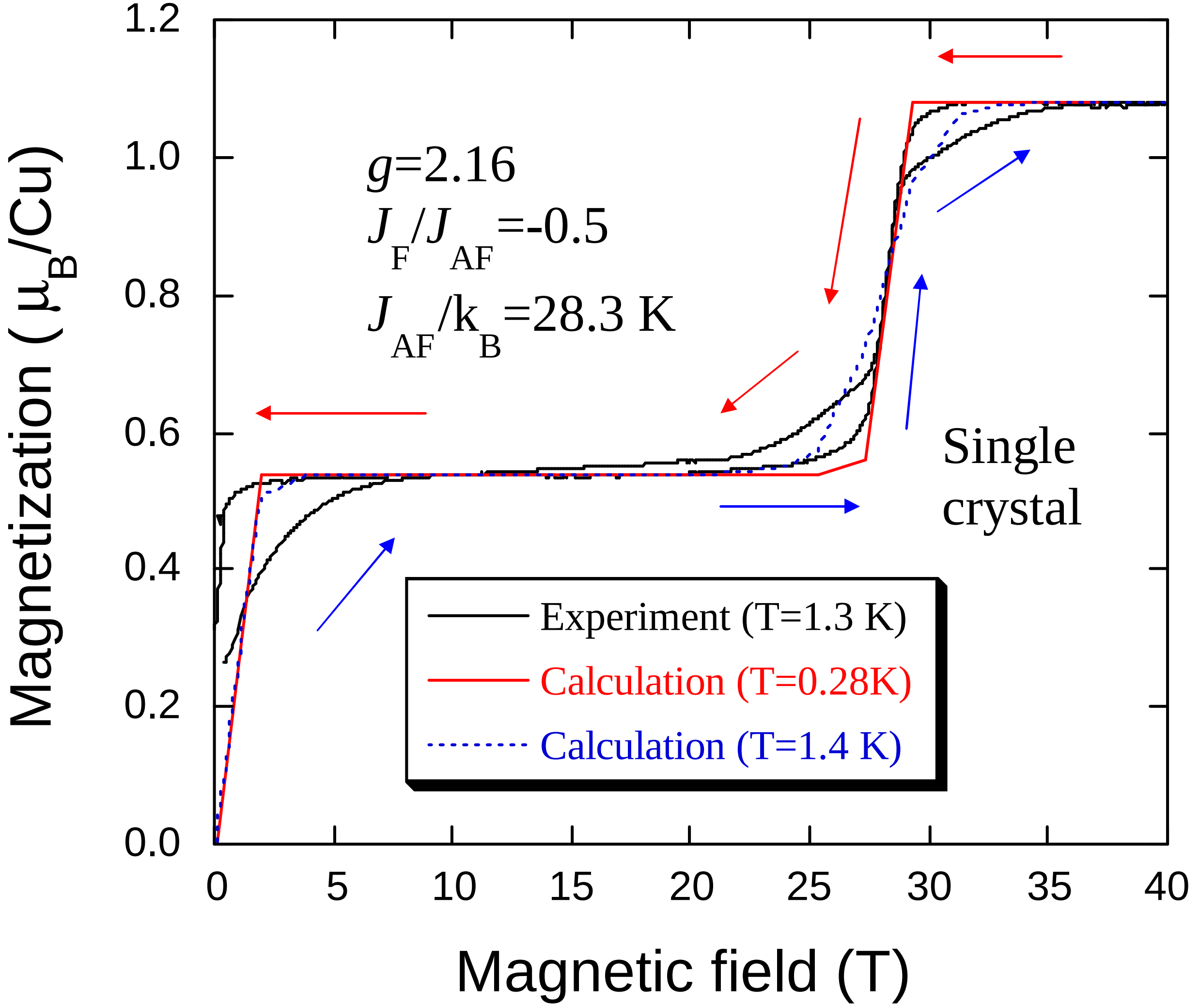
Dimer model (J=1.0 and D_x=0.1)



Rosen-Zener process



Magnetization as a function of magnetic field



Adiabatic motion in Heisenberg model

$$H = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j + Q - h(t) \sum_i S_i^z$$

$$M_x = \sum_i S_i^x, M_y = \sum_i S_i^y, M_z = \sum_i S_i^z$$

$$M_z(t) = \mathrm{e}^{iHt/\hbar} M_z \mathrm{e}^{-iHt/\hbar}$$

$$i\hbar \frac{\partial M_z(t)}{\partial t} = [M_z, H] = [M_z, Q], \text{ etc.}$$

$$Q = \Gamma M_x \qquad Q - h(t) \sum_i S_i^z = -\vec{h} \cdot \vec{M}$$

$$i\hbar \frac{\partial \vec{M}}{\partial t} = \left[\vec{M}, -\vec{h} \cdot \vec{M}\right]$$

Torque equation

$$\frac{\partial \vec{M}}{\partial t} = \vec{M} \times \vec{h}$$

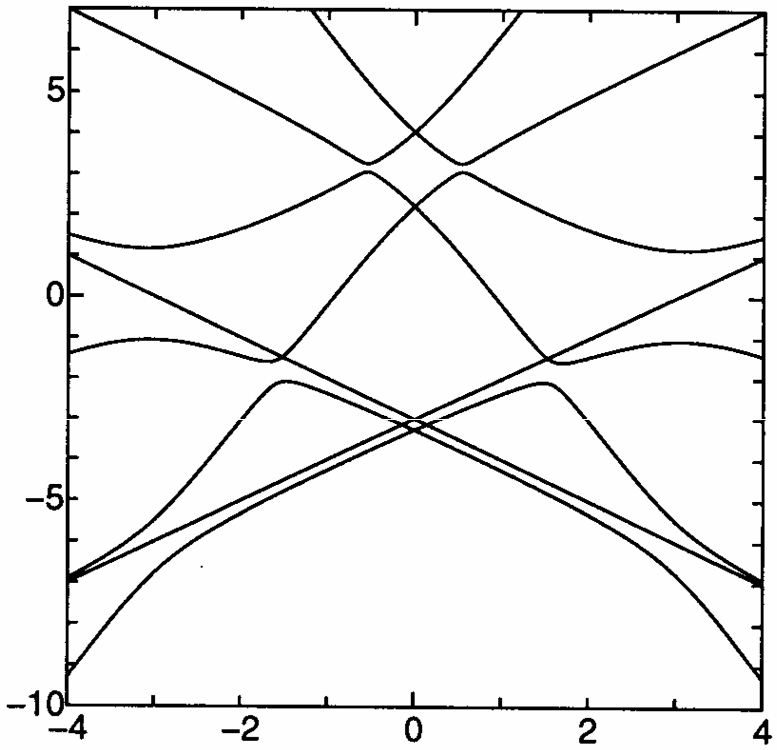
Response is the same as that of S=1/2 single spin

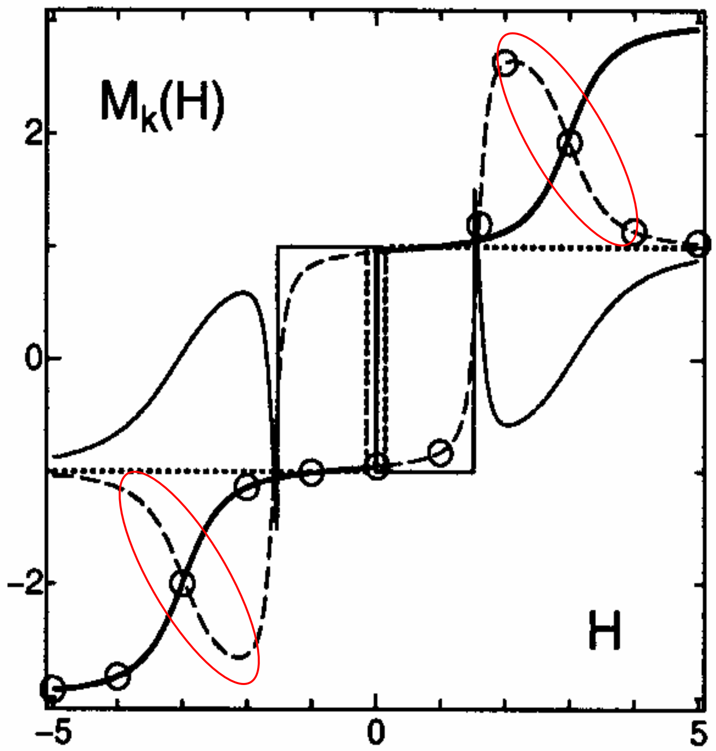
Weak coercive force

Quantum Control of state: Non AdiabaticTransitions

Non-monotonic magnetization process

SM, &. N. Nagaosa, Prog. Theor. Phys. 106 (2001) 533





Observation and Control of the Quantum Dynamics

- Magnetic field
- Pressure
- Bias voltage
- Temperature
- Photo-irradiation
- etc.

Quantum Mechanical Response

- Molecular magnets:

  isolation

  Magnetic field

$$g\mu_{\mathrm{B}}HS \quad 9.274\times10^{-24}[\mathrm{J}]$$

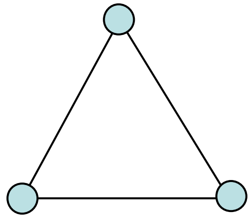
- Electric polarization :

  couple to distortion

  Electric filed

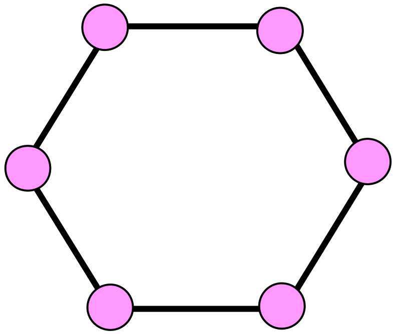
$$eE \quad 1.602\times10^{-19}[\mathrm{J}]$$

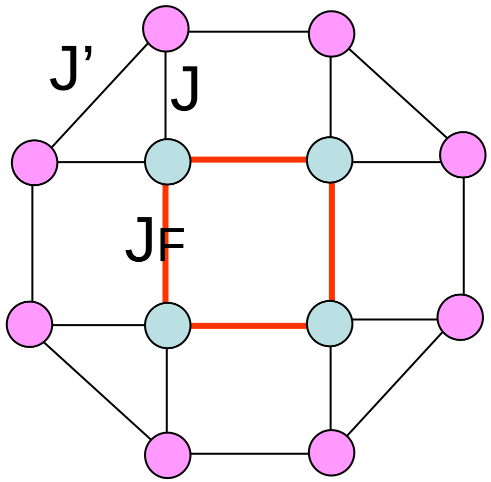
Types of microscopic spin states



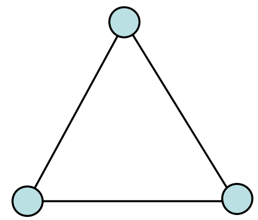
- Triangle lattice and odd rings
- Even rings
- New types of microscopic spin state

    Non-collinear ferrimagnetism





Local magnetization



Distorted case (without C3 symmetry)

$$\langle S_1 \rangle, \ \langle S_2 \rangle, \ \langle S_3 \rangle \qquad \langle S_1 + S_2 + S_3 \rangle = \frac{1}{2}$$

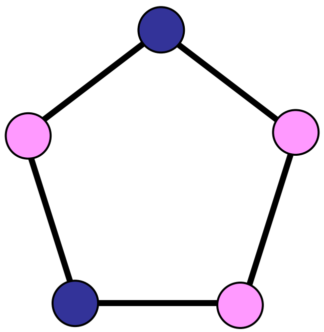
+ – +

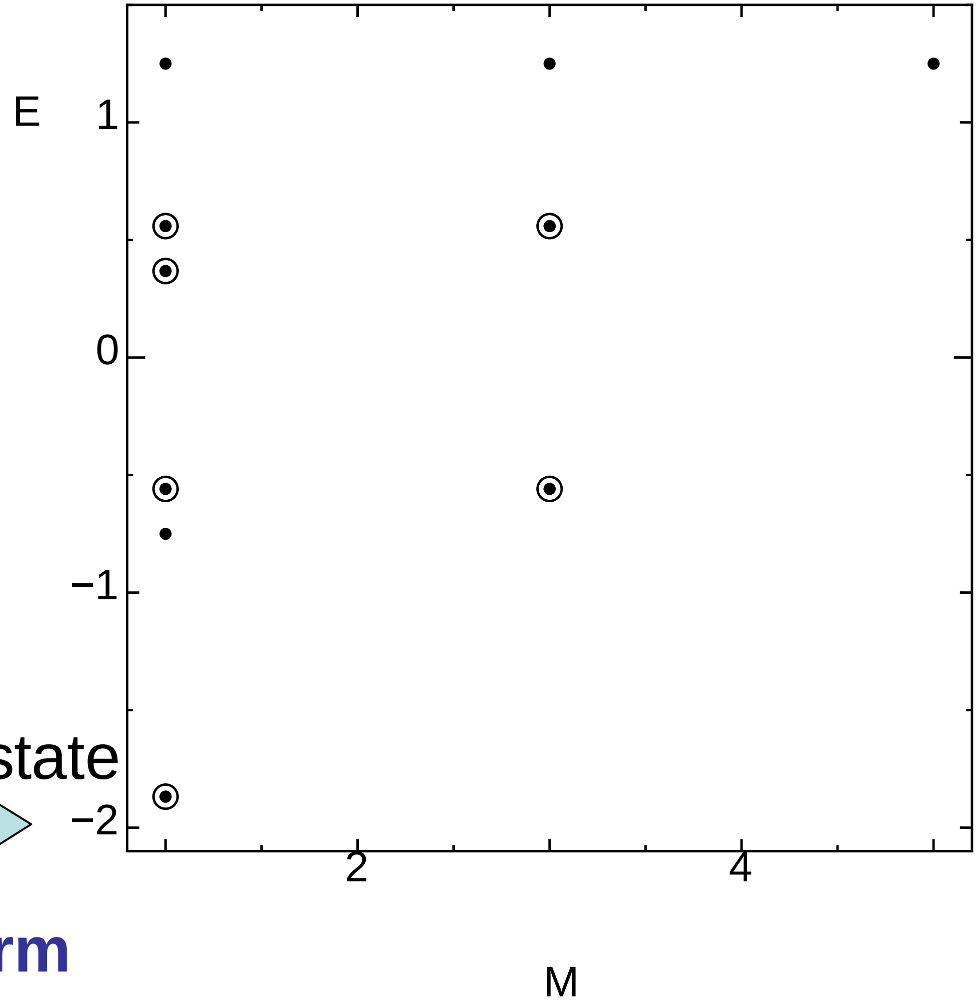
with C3 symmetry

$$\langle S_1 \rangle, \ \langle S_2 \rangle, \ \langle S_3 \rangle \qquad \langle S_1 + S_2 + S_3 \rangle = \frac{1}{2}$$

? ? ?

Odd ring N=5

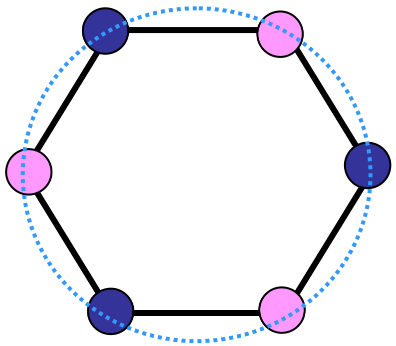


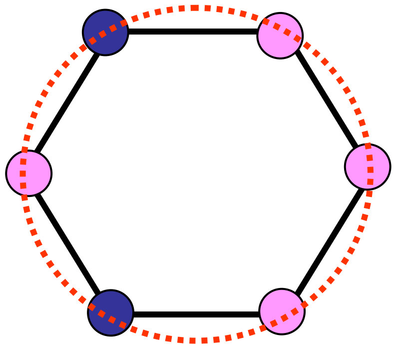


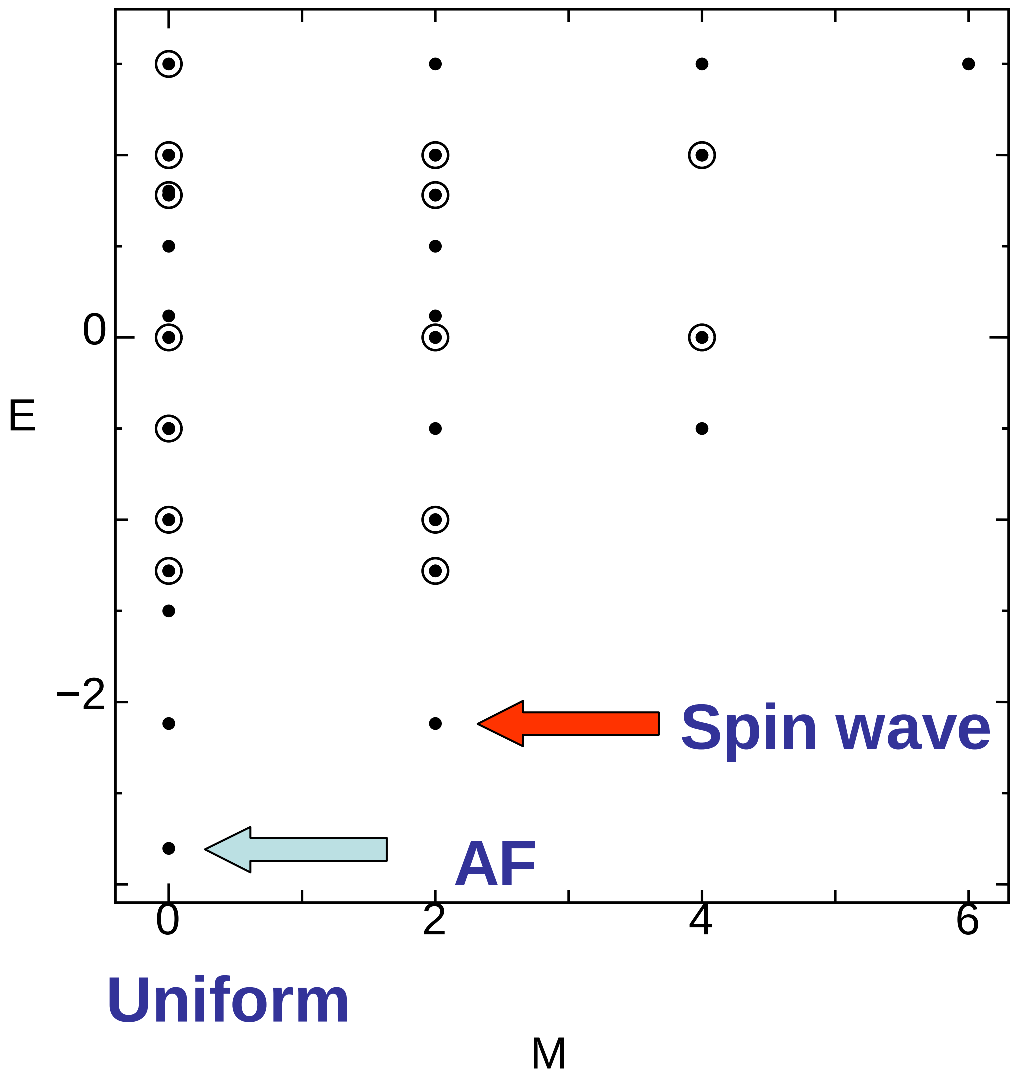
Degenerate ground state

Non-uniform

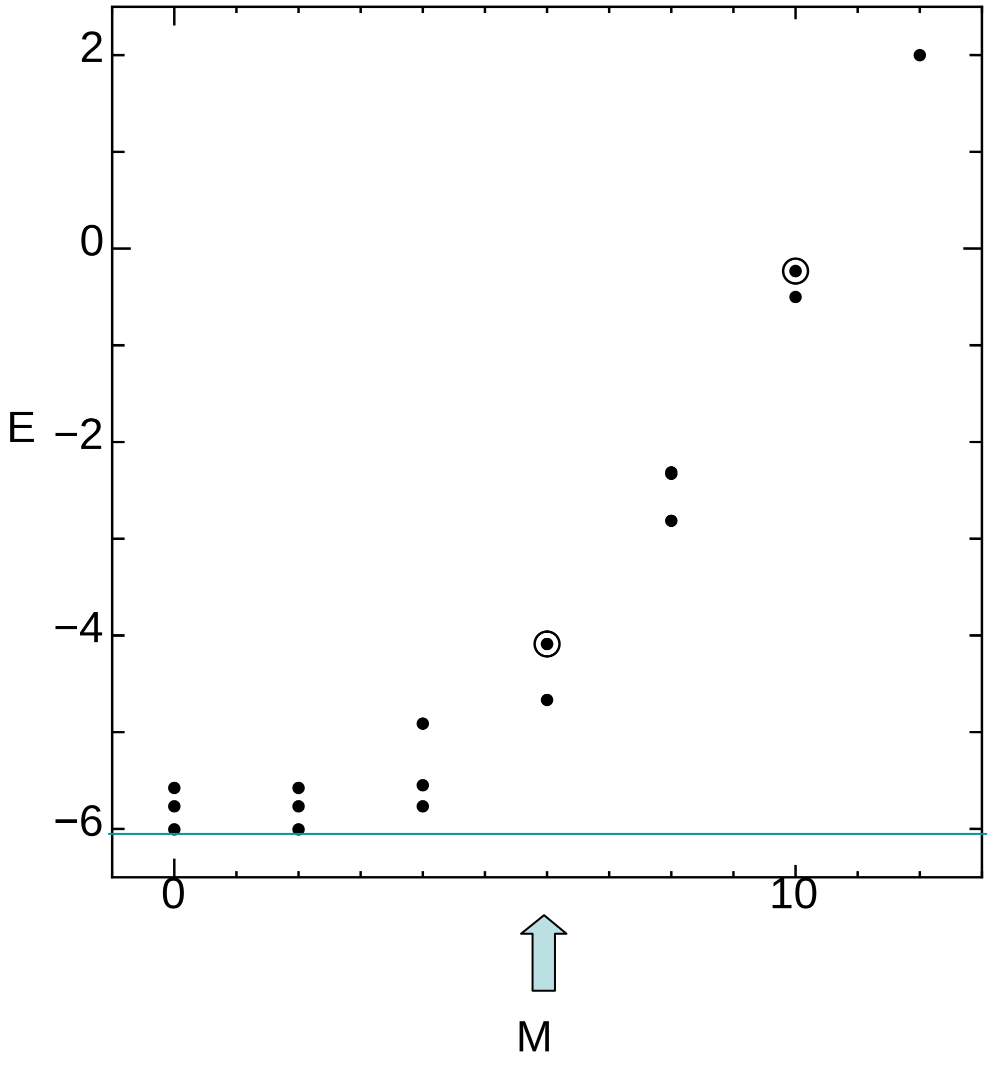
Even spin case   N=6

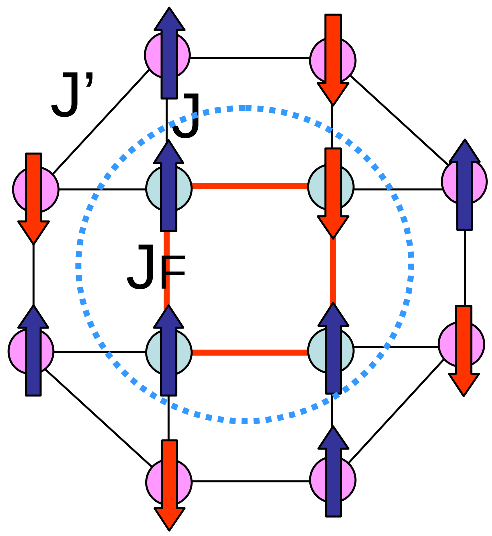






N=12 Non-collinear Ferrimagnetism



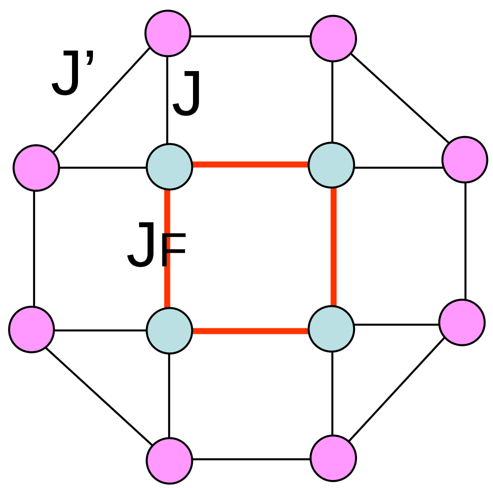


$J=J'=1,\ J_F=2$

Uniformly fluctuating magnetic state?

Spin wave type $$|\Psi\rangle = \frac{1}{\sqrt{3}}\left(|++-\rangle + |+-+\rangle + |-++\rangle\right)$$

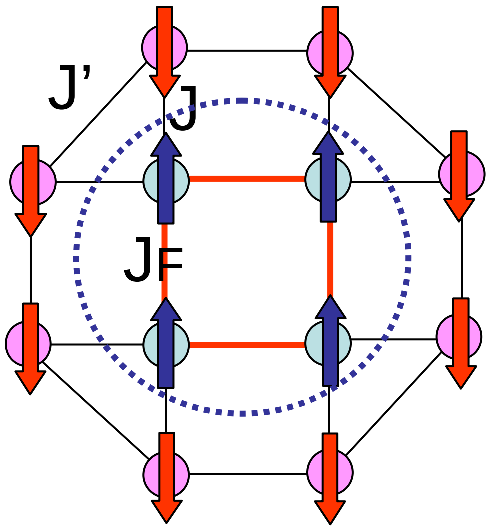
Non-collinear ferrimagnetic state: ground state

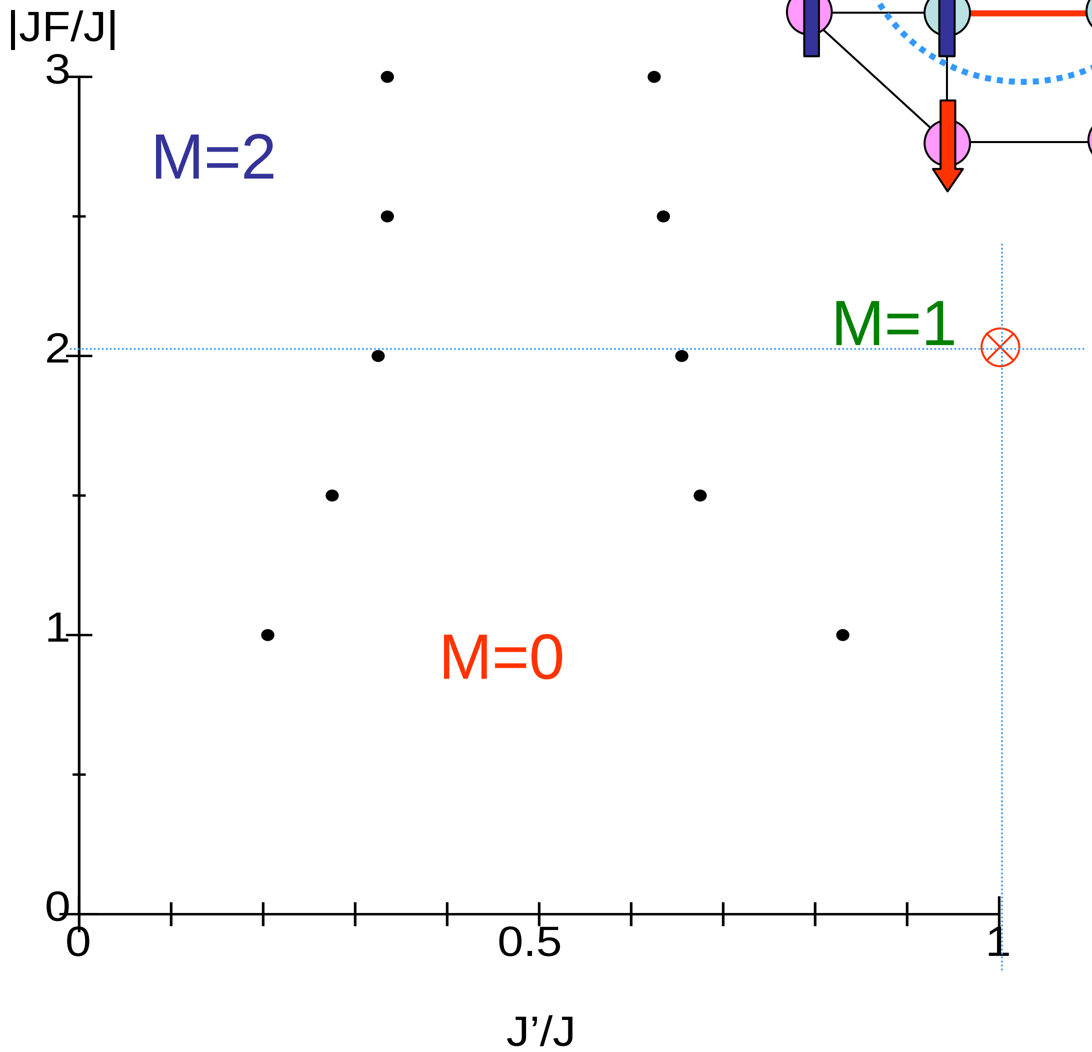


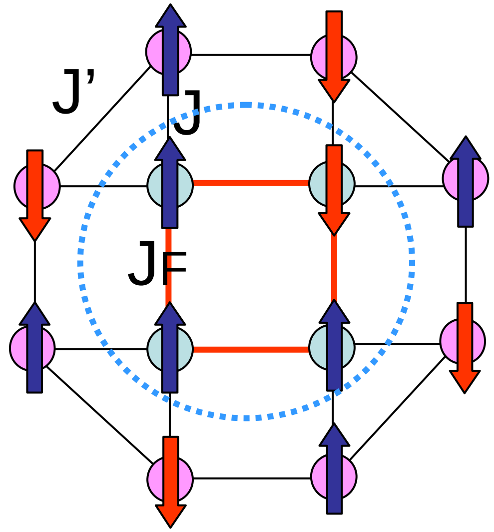
J >> J' : LMFR

J << J' : Non-LMFR

LM & Non-Collinear ferrimagnetism







Uniform nonzero magnetization in the ground state?

Ferri-magnetic state

- Lieb-Mattis type

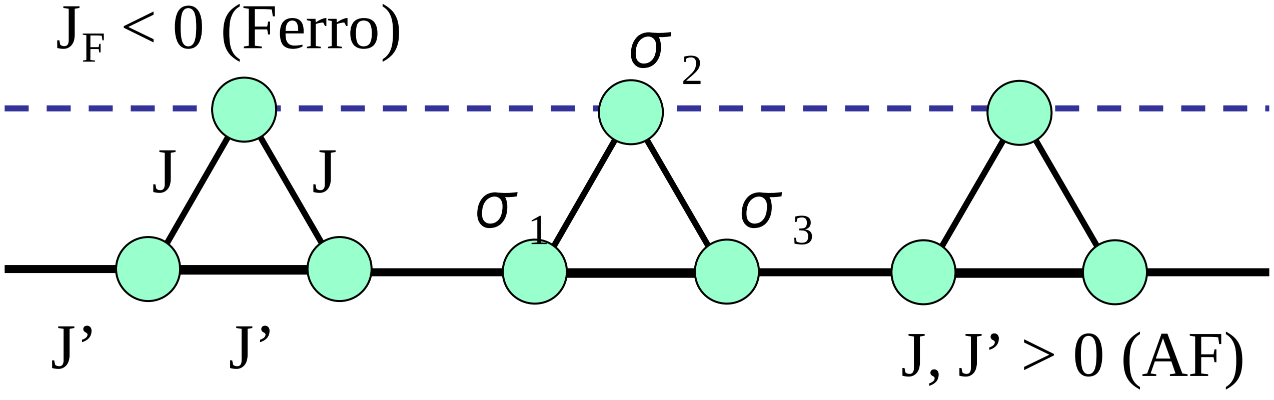
    Localized magnetic moment

- Non-collinear type

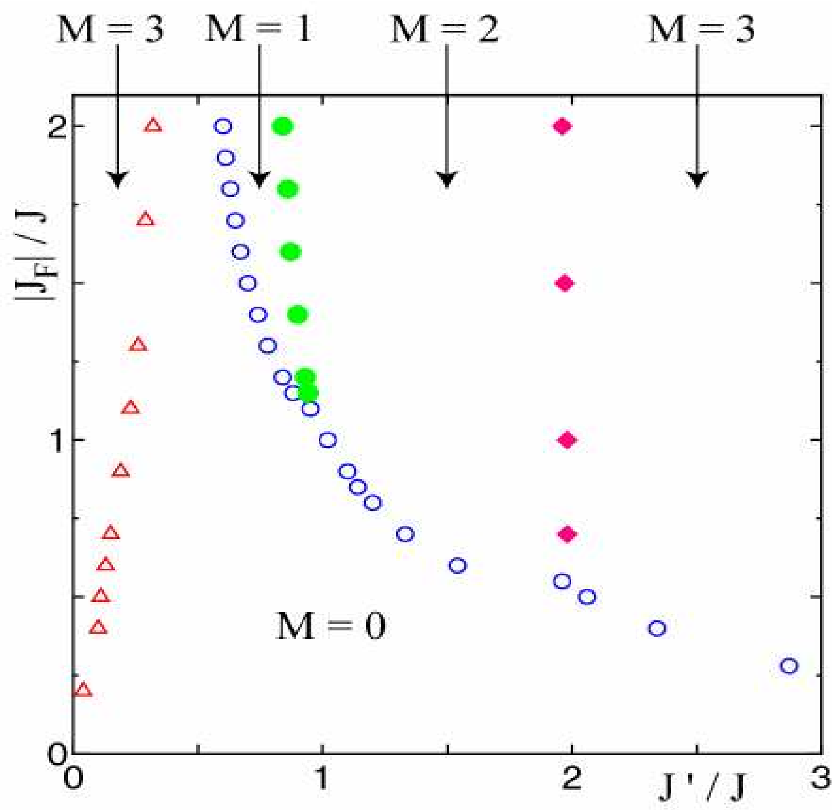
    Uniform magnetic moment

S. Yoshikawa and S. Miyashita: JPSJ Supple 2005

Non-saturated magnetized state



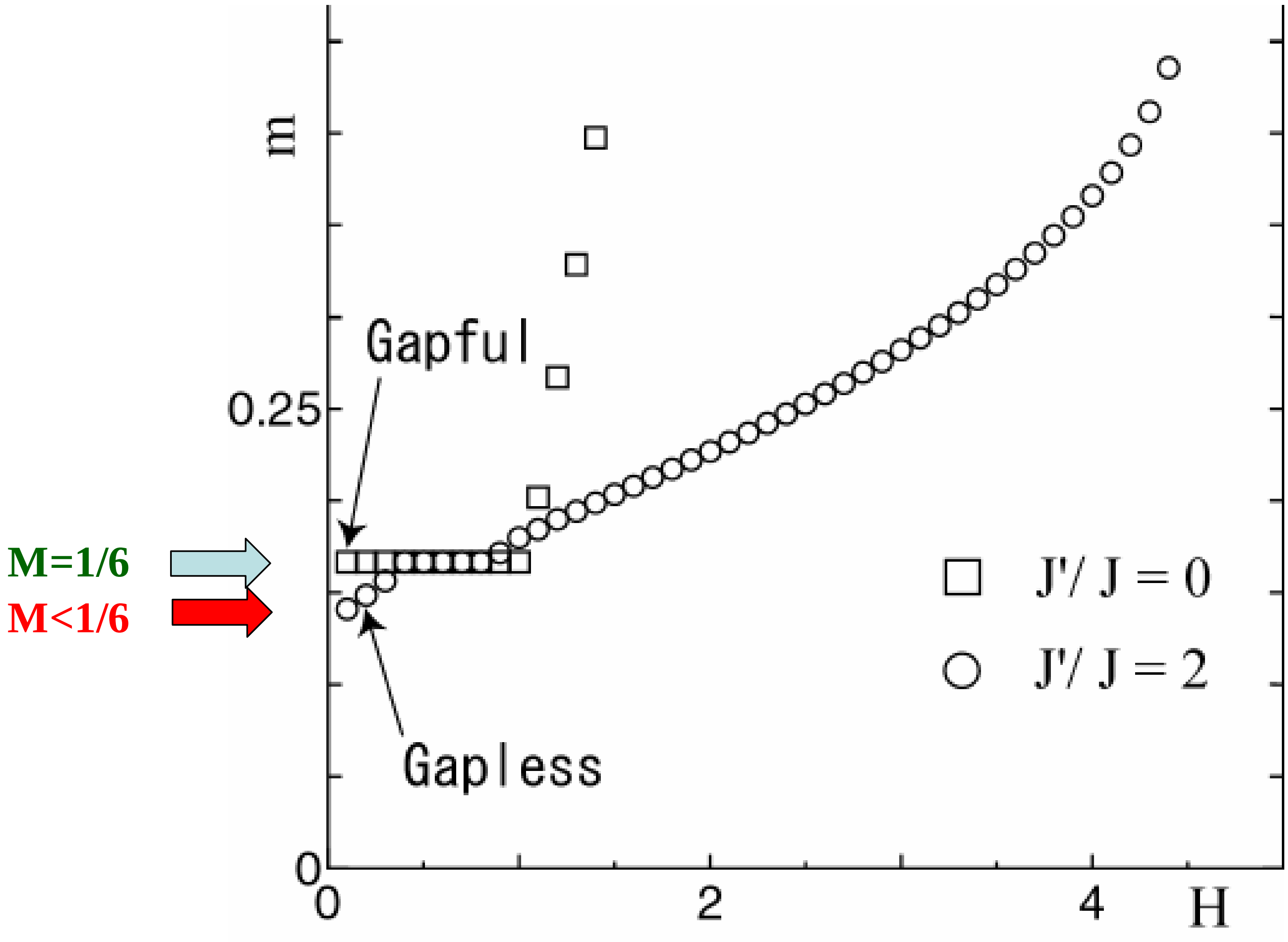
N=18



J >> J' : LMFR

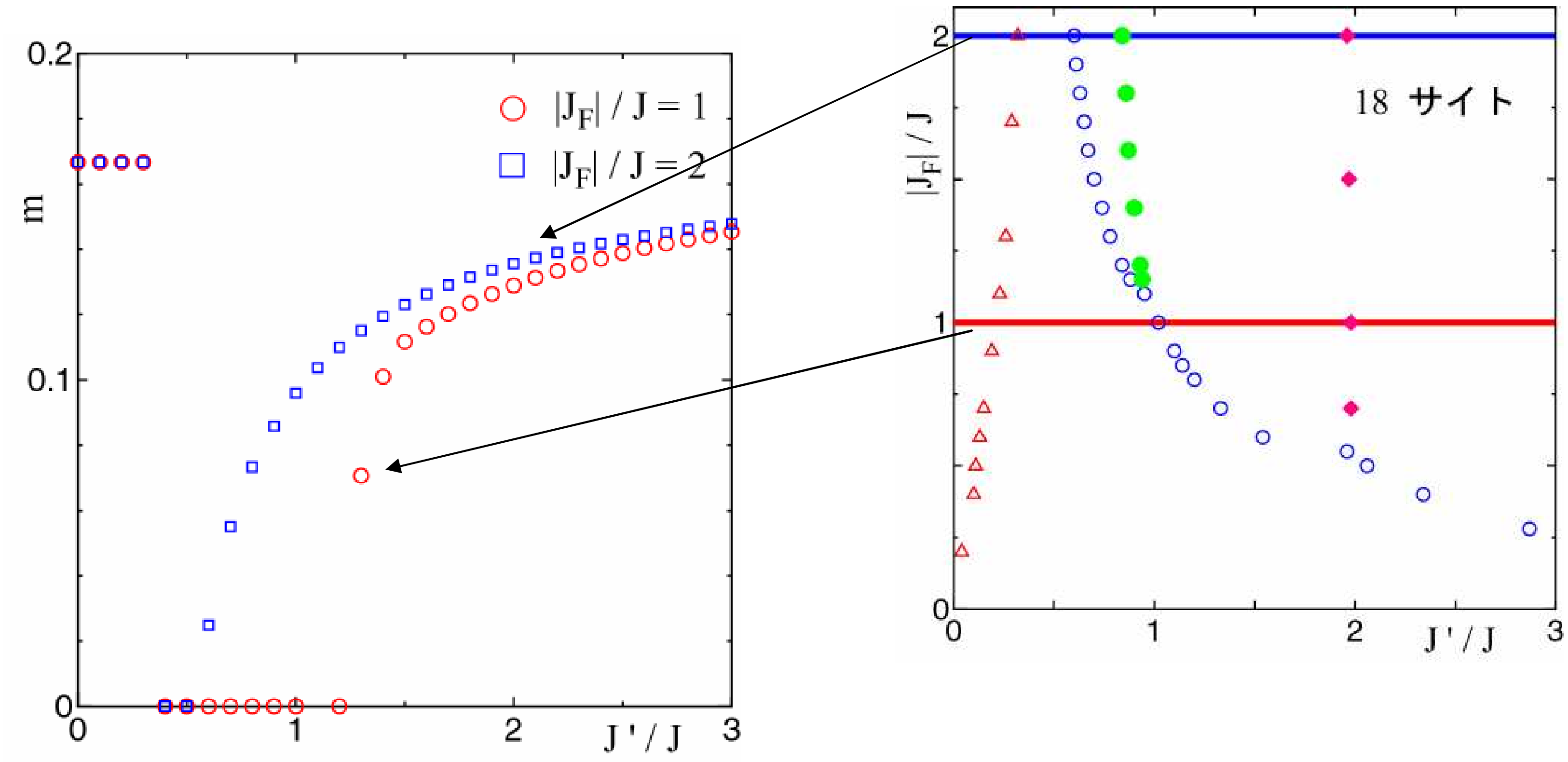
J << J' : Non-LMFR

Magnetization processes



Phase diagram

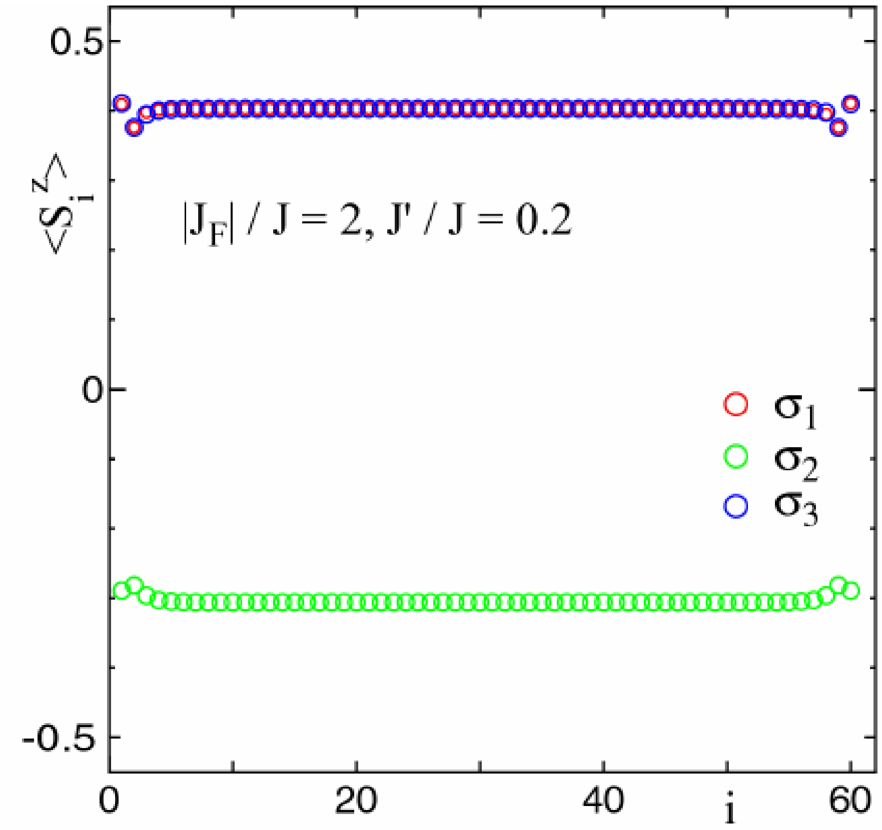
J_F fixed



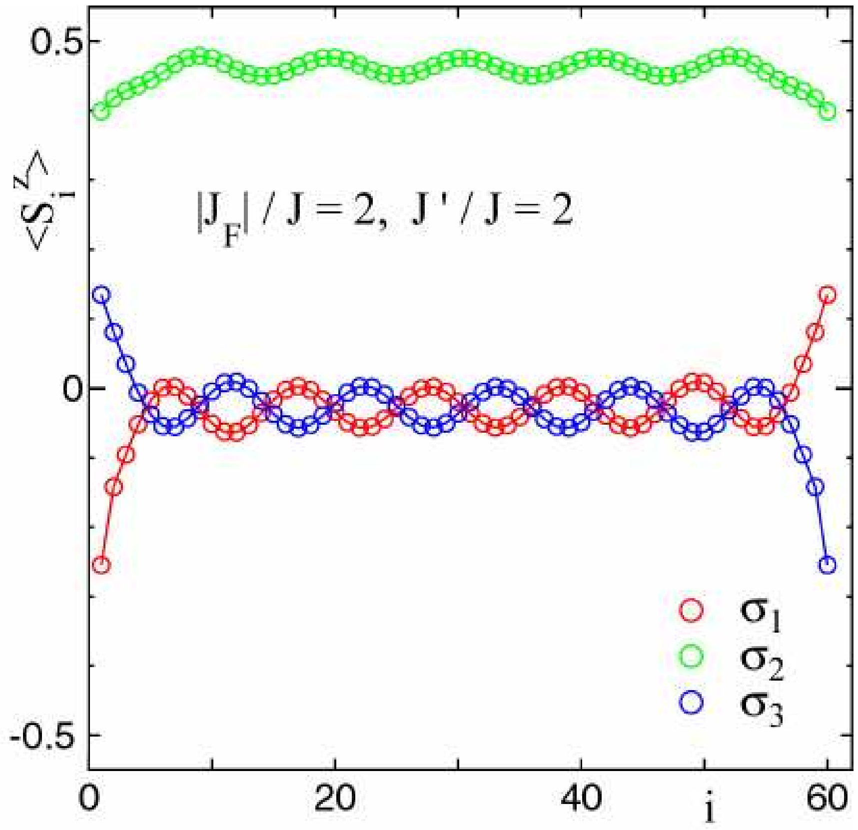
As J’ increases the jump of the magnetization becomes large.

Local magnetic structures

LM phase



Non-collinear phase



3×60