KEK seminar
2020 Dec. 10
online

Atomistic approach to the Nd-Fe-B magnet
-- Coercivity at finite temperatures --

Seiji Miyashita

Physical society of Japan (JPSJ)

ISSP, The University of Tokyo

Elements Strategy Initiative Center for Magnetic Materials(ESICMM)

Collaborators

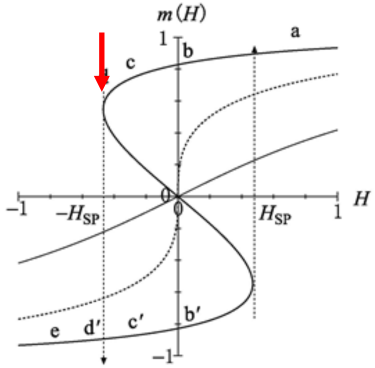
Yuta Toga, Masamichi Nishino, Taichi Hinokihara, Ismail. E. Uysal,

Akimasa Sakuma, Takashi Miyake ,Hisazumi. Akai, Satoshi Hirosawa

Magnets

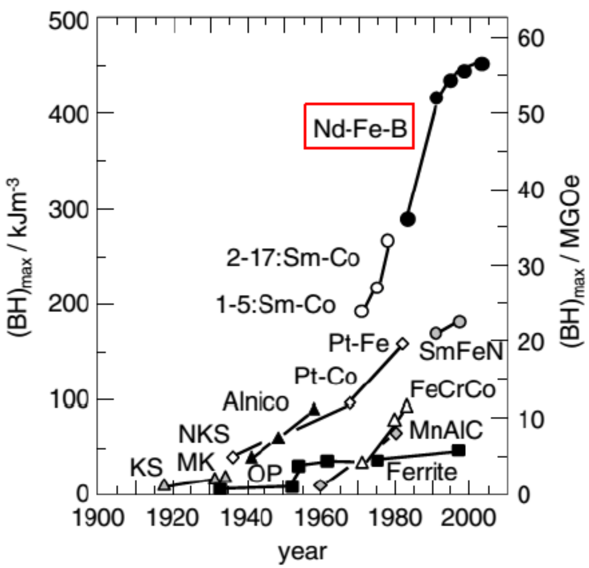
Ferromagnets

Coercivity

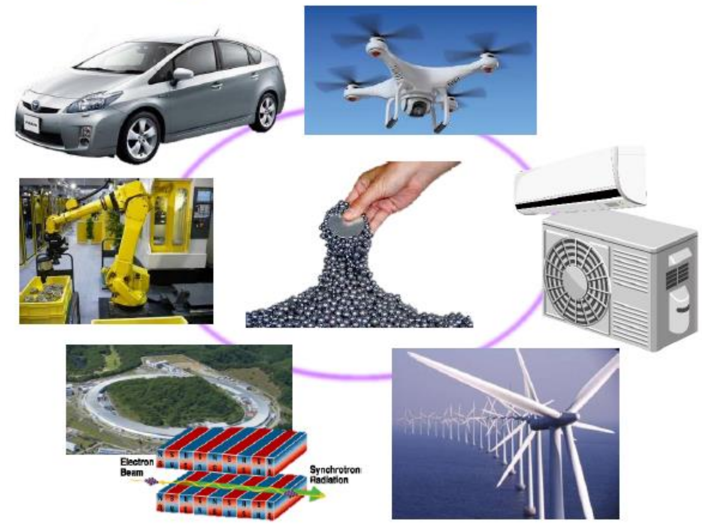


High performance magnet

$Nd_2Fe_{14}B$ (Masato Sagawa)



High performance permanent magnets ⇨ High efficiency motor, generator



Motor (cars)

Properties at high temperatures T (T<Tc)

Rare-earth elements (Dy)

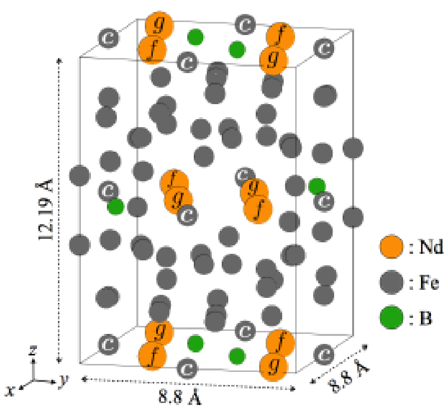
Mechanism of coercivity at finite temperatures

$Nd_2Fe_{14}B$ magnet at finite temperatures

Thermodynamic properties $$\langle A(T)\rangle = \frac{\mathrm{Tr}\, A e^{-\beta\mathcal{H}}}{\mathrm{Tr}\, e^{-\beta\mathcal{H}}}$$

All properties can be obtained by **an atomistic Hamiltonian $\mathcal{H}$**.

$$\hat{H} = \hat{H}_{\mathrm{ex}} + \hat{H}_{\mathrm{a}} + \hat{H}_{\mathrm{cry}} + \hat{H}_{\mathrm{z}},$$

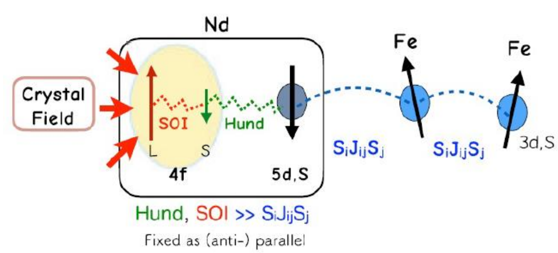


$$\hat{H}_{\mathrm{ex}} = -\sum_{\mathrm{Fe}} 2J_{ij}\boldsymbol{s}_i \cdot \boldsymbol{s}_j$$ First-principles calculation(KKR method by Akai)

$$\hat{H}_{\mathrm{a}} = -\sum_i D_i (s_i^z)^2$$ First-principles calculation (Y. Miura et al., J. Appl. Phys. 115, 17A765 (2014)).

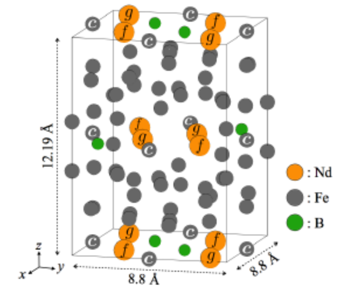
$$\hat{H}_{\mathrm{cry}} = \sum_i \sum_{l,m} A_l^m \langle r^i \rangle O_l^m,$$ M. Yamada et al., Phys. Rev. B 38, 620 (1988).

$$\hat{H}_{z} = -\sum_i h_i S_i^z$$



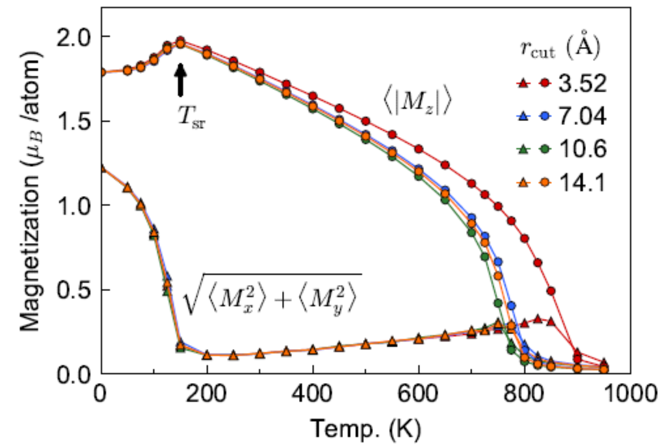
Y. Toga, M. Matsumoto, S. Miyashita, H. Akai, S. Doi, T. Miyake, and A. Sakuma, Phys. Rev. B **94**, 174433 (2016).

Atomic model of $Ne_2Fe_{14}B$ magnet



Magnetization $M(T)$

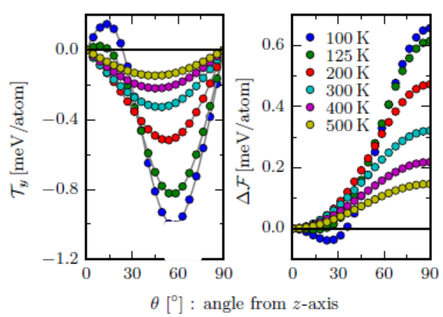
Reorientation phase transition
Ferromagnetic phase transition

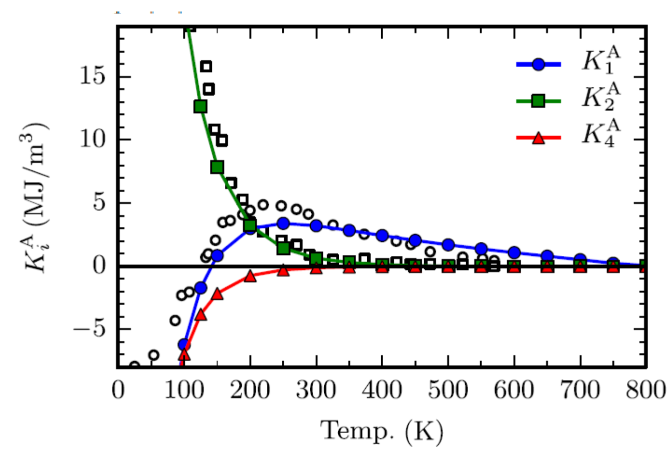


Anisotropy energies

$$\mathcal{T}_y(\theta, T) = -\frac{\partial}{\partial\theta}\Delta\mathcal{F}(\theta, T),$$

$$\Delta\mathcal{F}(\theta, T) = K_1^{\rm A}(T)\sin^2\theta + K_2^{\rm A}(T)\sin^4\theta + K_4^{\rm A}(T)\sin^6\theta.$$



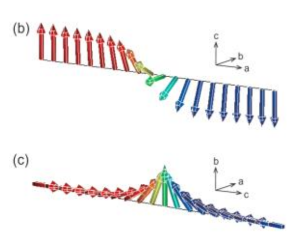


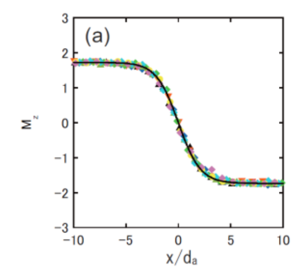
Y. Toga, M. Matsumoto, S. Miyashita, H. Akai, S. Doi, T. Miyake, and A. Sakuma: P.RB 94, 174433 (2016).

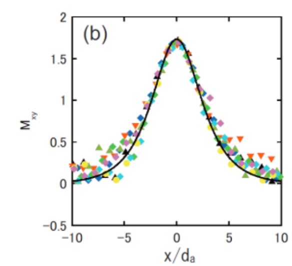
Domain wall profile

Type I: Bloch type wall

Type II: Neel type wall





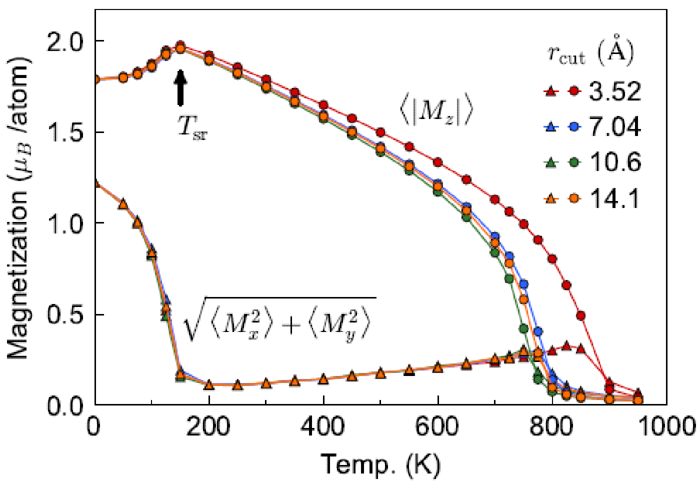


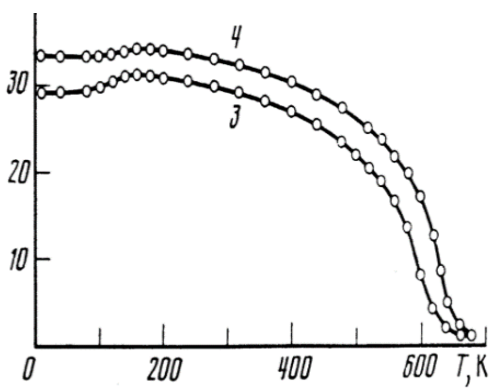
M. Nishino, Y. Toga, S. Miyashita, H. Akai, A. Sakuma, and S. Hirosawa, Phys. Rev. B 95, 094429 (2017).

Thermodynamic properties : magnetization

Reorientation phase transition T~150K

Magnetization $M(T)$

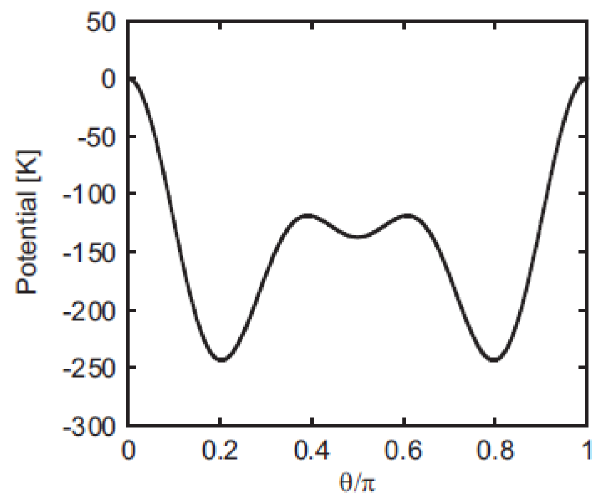


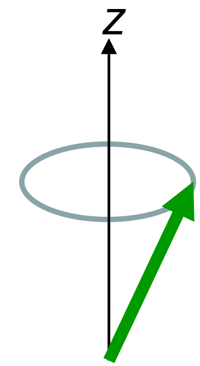


3: Nd2Fe14B,　4:Nd2Fe14BH3.8

Tilted anisotropy

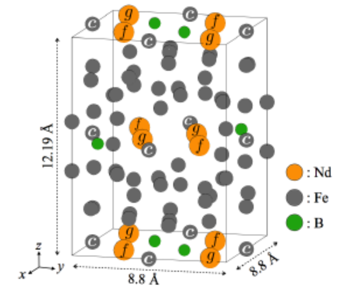
Ndの異方性エネルギー $\hat{H}_{\mathrm{cry}} = \sum_i \sum_{l,m} A_l^m \langle r^i \rangle O_l^m,$





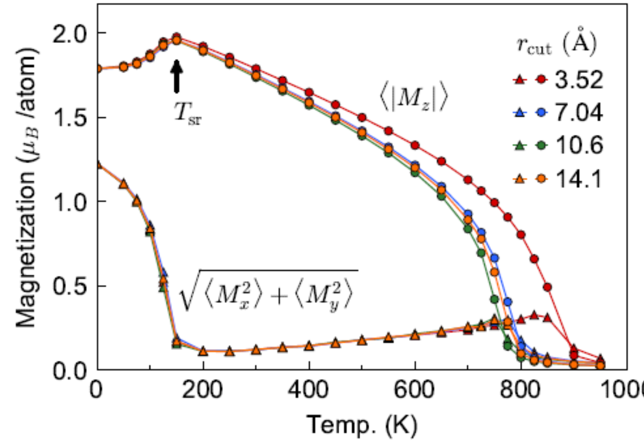
A. V. Andreev, et al. Sov. Phys. JETP 63(3) 608 (1986).
S. Hirosawa, et al. J. Appl. Phys 59 (1986)873.

Atomic model of Ne$_2$Fe$_{14}$B magnet



Magnetization $M(T)$

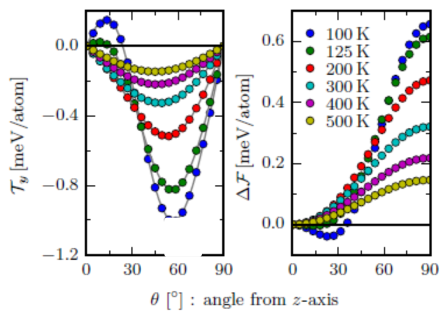
Reorientation phase transition
Ferromagnetic phase transition

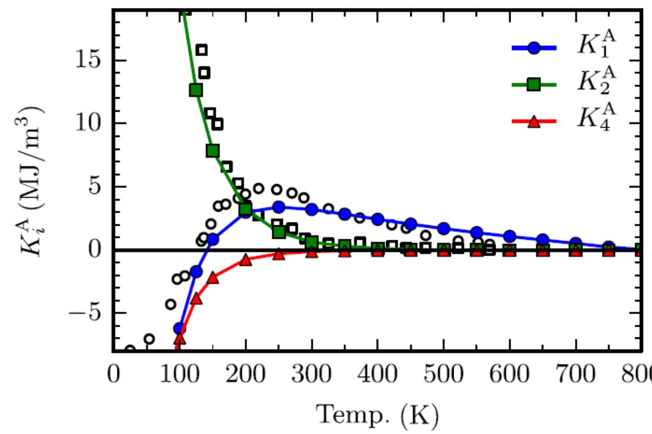


Anisotropy energies

$$\mathcal{T}_y(\theta,T) = -\frac{\partial}{\partial\theta}\Delta\mathcal{F}(\theta,T),$$

$$\Delta\mathcal{F}(\theta,T) = K_1^A(T)\sin^2\theta + K_2^A(T)\sin^4\theta + K_4^A(T)\sin^6\theta.$$



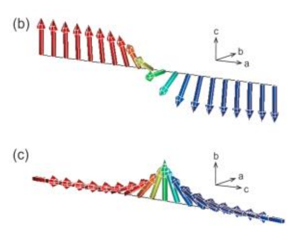


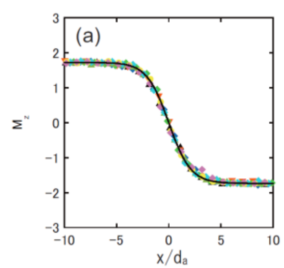
Y. Toga, M. Matsumoto, S. Miyashita, H. Akai, S. Doi, T. Miyake, and A. Sakuma: P.RB 94, 174433 (2016).

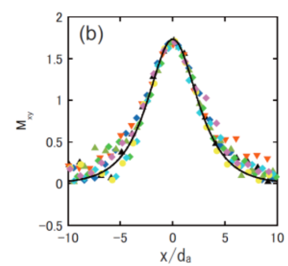
Domain wall profile

Type I: Bloch type wall

Type II: Neel type wall



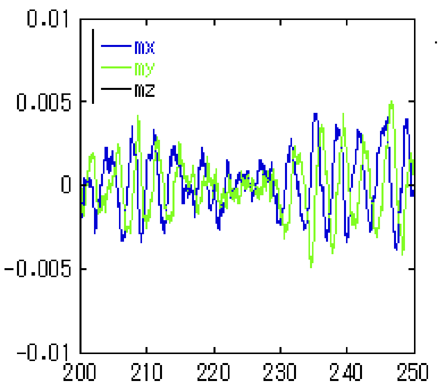




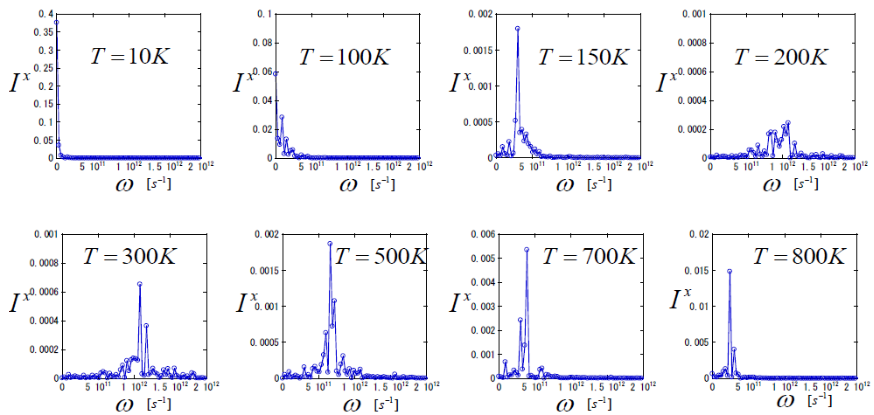
M. Nishino, Y. Toga, S. Miyashita, H. Akai, A. Sakuma, and S. Hirosawa, Phys. Rev. B 95, 094429 (2017).

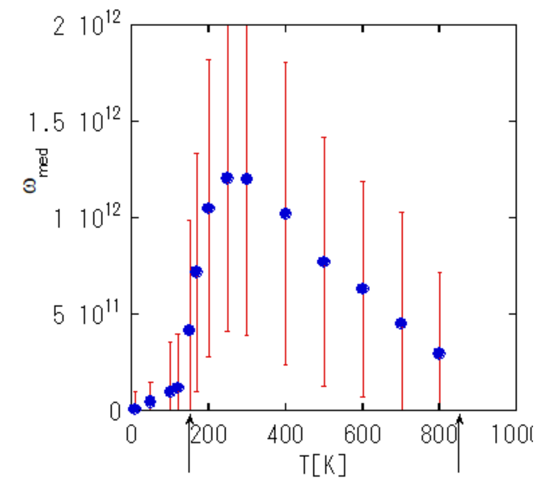
Dynamical response (FMR)

Power Spectrum: $$I^{\alpha}(f) \equiv \frac{1}{T}\int_{t_0}^{t_0+T} d\tau\, I^{\alpha}(\tau) e^{i2\pi f\tau}, \quad I^{\alpha}(\tau) = \frac{1}{T}\int_{t0}^{t0+T} dt \left\langle \bar{S}^{\alpha}(t)\bar{S}^{\alpha}(t+\tau)\right\rangle$$



$$\bar{S}^{\alpha} = \frac{1}{N_{site}}\sum_i S_i^{\alpha},\ \alpha = x, y, z$$





M. Nishino, S. Miyashita: PRB 100 (2019) 020403(R).

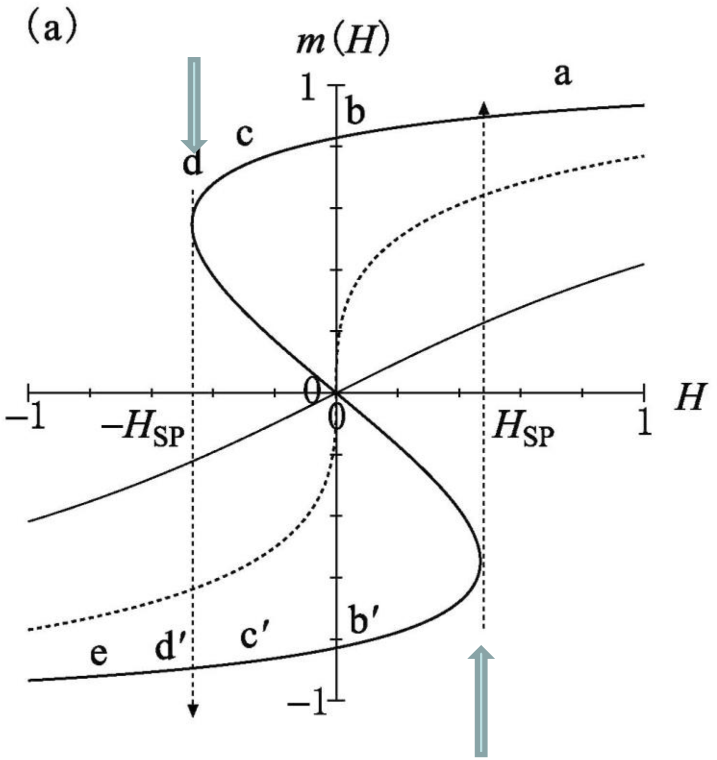
Stochastic LLG(Landau-Lifshitz-Gilbert) equation

$$\frac{d\mathbf{M}_i}{dt} = -\frac{\gamma}{1+\alpha_i^{\,2}}\mathbf{M}_i\times\mathbf{H}_{\mathrm{eff},i} - \frac{\alpha_i\gamma}{(1+\alpha_i^{\,2})M_{\mathrm{i}}}\mathbf{M}_i\times(\mathbf{M}_i\times\mathbf{H}_{\mathrm{eff},i}), \qquad i = 1, 2, \cdots, \mathrm{N}$$

$$\mathbf{H}_{\mathrm{eff,i}} \to \mathbf{H}_{\mathrm{eff,i}} + \boldsymbol{\xi}_{\mathrm{fl,i}} \qquad \left\langle \xi_i^{\alpha}(t)\right\rangle = 0,\ \left\langle \xi_i^{\alpha}(t)\xi_j^{\beta}(s)\right\rangle = 2D_i\delta_{ij}\delta_{\alpha\beta}\delta(t-s). \qquad \frac{\alpha_i}{M_i} = \frac{\gamma D_i}{k_B T}$$

J. L. Garcia-Palacios and Francisco J. Lazaro: Phys. Rev. B58 (1998) 14937.
T. A. Ostler, et al.: Phys. Rev. B84, 024407 (2011).
R. F. L. Evans, et al: J. Phys. Condens. Matter 26 (2-14) 103202.
M. Nishino and S. Miyashita, Phys. Rev. B91, 134411(1-13) (2015).

Cocercivity(保磁力)



End of the metastable state (spinodal point)

Metastable state (Mean-field picture)

Ising model

$$\mathcal{H} = -J \sum_{<ij>} \sigma_i \sigma_j - H \sum_i \sigma_i$$

σ_i=±1, <ij>格子上の最近接対、その個数 z

$$m = \tanh(\beta J z m + \beta H)$$

Free energy F(m:T,H)

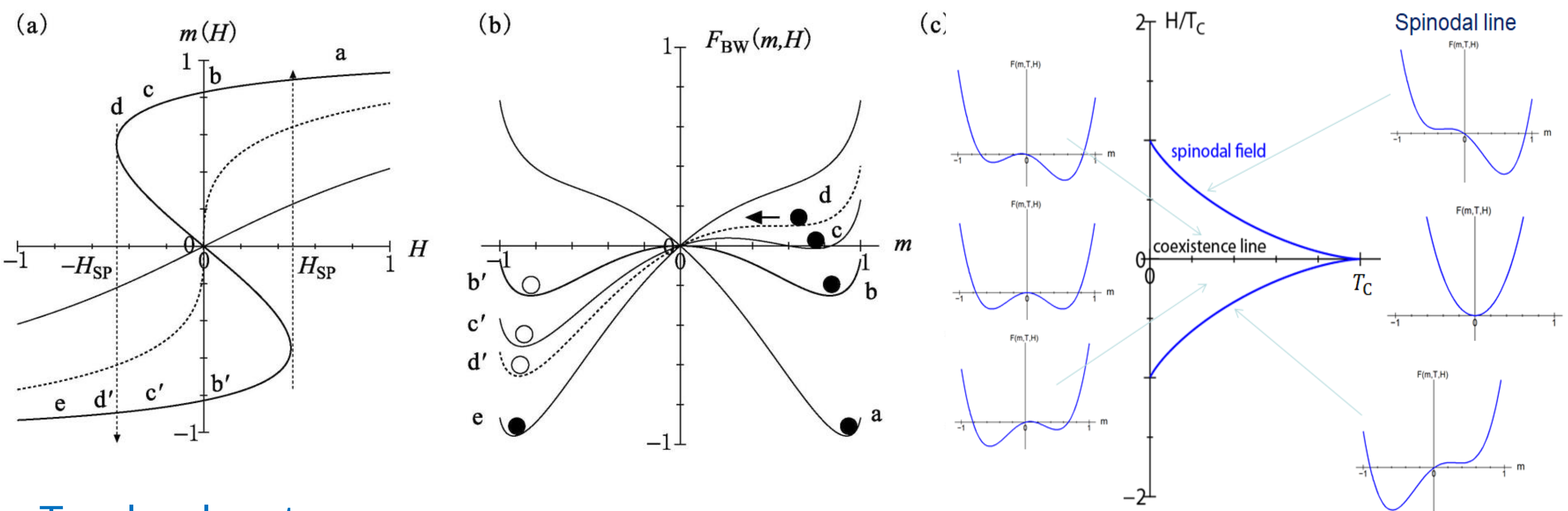
Bragg-Williams-近似

$$\frac{1}{N} F(m{:}T,H) = -\frac{Jz}{2} m^2 - Hm + k_{\mathrm{B}} T \left(\frac{1+m}{2} \ln \frac{1+m}{2} + \frac{1-m}{2} \ln \frac{1-m}{2} \right)$$

ゆらぎ無視の近似

$$\frac{1}{N} F(m{:}T,H) = \frac{Jz}{2} m^2 - k_{\mathrm{B}} T \ln(2\cosh(\beta(zJm + H)))$$

「ゆらぎと相転移」,丸善出版（2018）



Two-level systems:

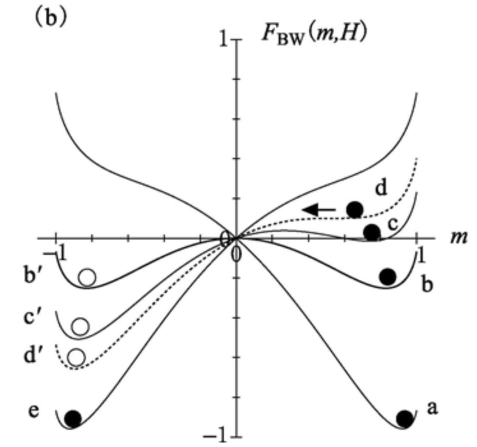
Ising model (spin), binary alloy (A,B atoms), spin-crossover systems (HS,LS), adsorption(on, off), lattice-gas model(occupation, vacancy) , vote (yea, nay), etc.

Hysteresis loop

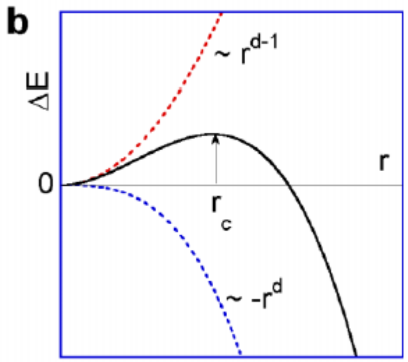
Mean-field theory (Spinodal point)

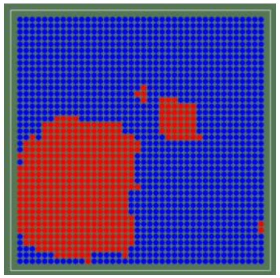
$$m = \tanh(\beta J z m + \beta H)$$

$$\frac{1}{N}F(m:T,H) = -\frac{Jz}{2}m^2 - Hm$$
$$+k_B T\left(\frac{1+m}{2}\ln\frac{1+m}{2} + \frac{1-m}{2}\ln\frac{1-m}{2}\right)$$



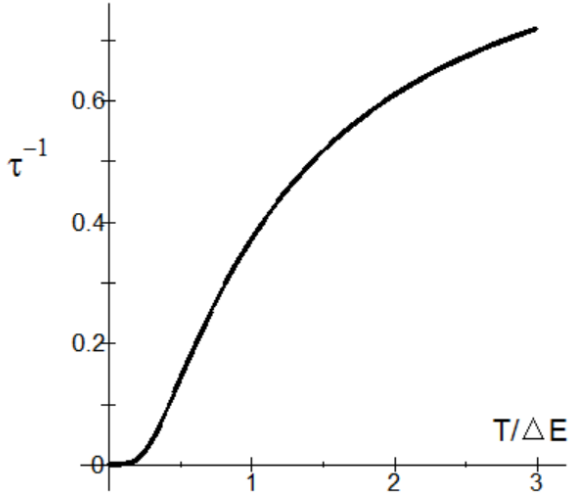
Nucleation   Finite energy barrier





$$p_{\text{nucleation}} \propto e^{-\beta \Delta E_c}, \quad \tau = p_{\text{nucleation}}^{-1}$$

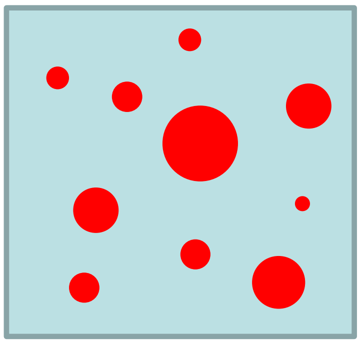
Metastable states must relax, if we wait long.



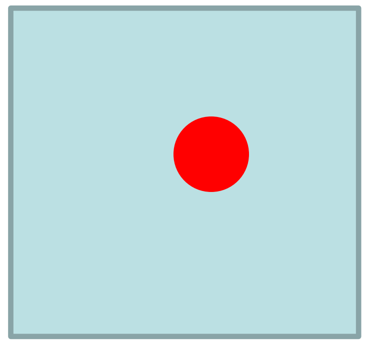
Dynamical spinodal point

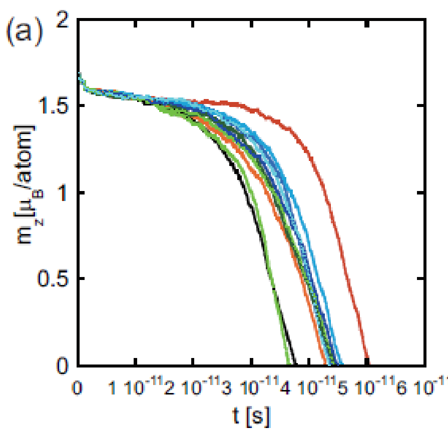
Multi-nucleation process(Avrami process, deterministic)
Single-nucleation process (stochastic)

Large *H*

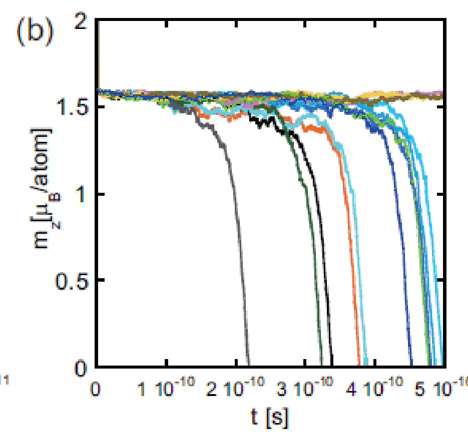


small *H*

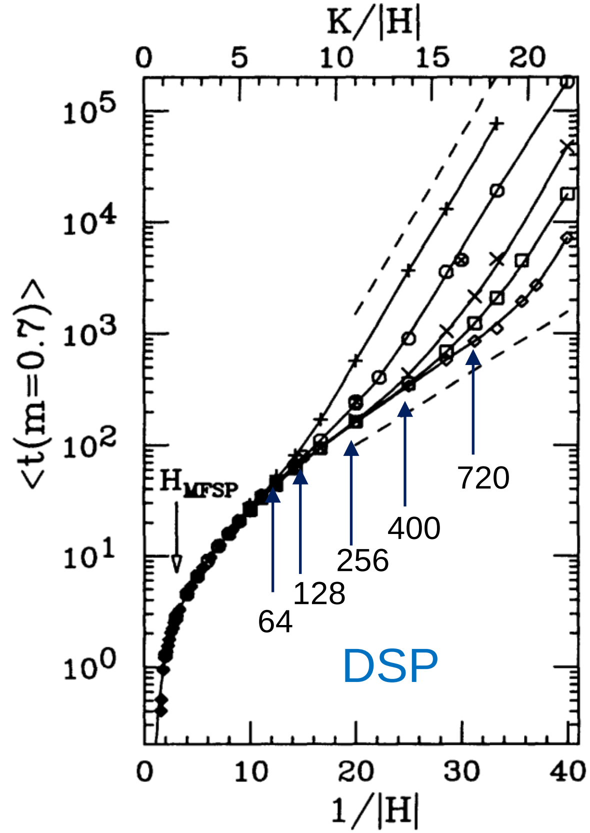




deterministic

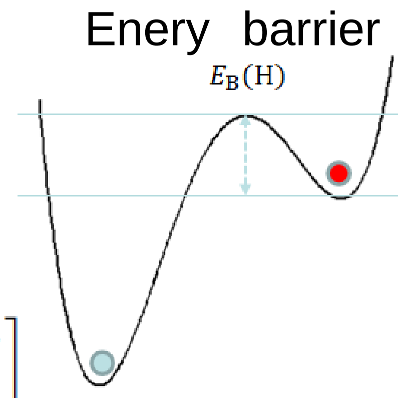


stochastic



P. A. Rikvold, H. Tomita, S. Miyashita and S. W. Sides: Phys. Rev. E 49, 5080-5090 (1994).

Néel-Arrhenius process



The probability to survive until time t:

$$P(t = n\Delta t) = \prod_n \left[1 - ae^{-\beta E_{\rm B}(H(n\Delta t)}\Delta t\right] = \exp\left[-a\int_{-\infty}^{t} e^{-\beta E_{\rm B}(H(t')dt'}\right]$$

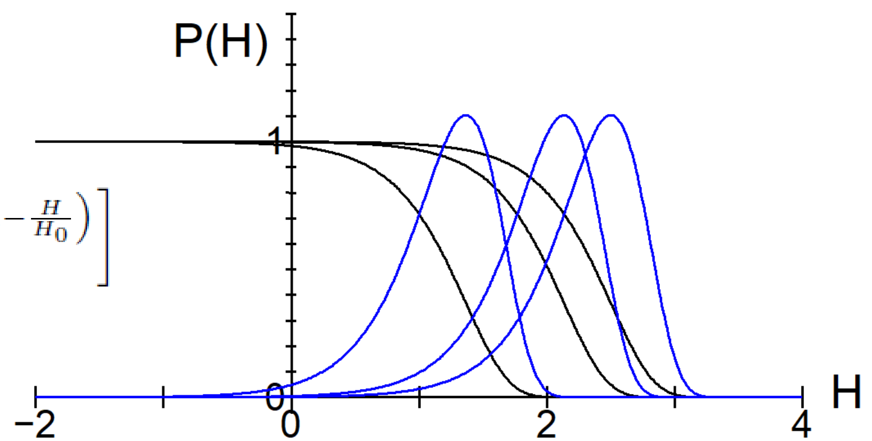
If the field is swept linearly $H(t) = vt$, $\frac{dH}{dt} = v$, then the probability as a function of H is

$$P(H) = \exp\left[-a\int_{H(-\infty)=-\infty}^{H(t)} e^{-\beta E_{\rm B}(h)}\frac{dt}{dh}dh\right] = \exp\left[-\frac{a}{v}\int_{-\infty}^{H} e^{-\beta E_{\rm B}(h)}dh\right]$$

Here we assume $E_{\rm B}(H) = E_0\left(1 - \frac{H}{H_0}\right)^n$, $n = 1$.Then, the propability is

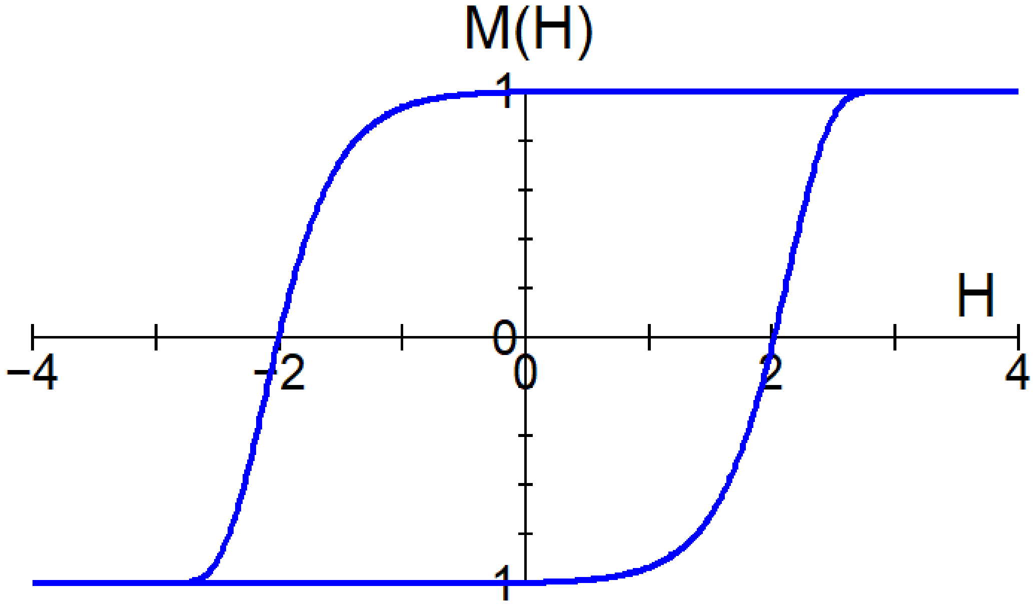
$$P(H) = \exp\left[-\frac{aH_0}{v\beta E_0}e^{-\beta E_0\left(1-\frac{H}{H_0}\right)}\right]$$

$$\frac{dP(H)}{dH} = -\frac{a}{v}e^{-\beta E_0\left(1-\frac{H}{H_0}\right)}\exp\left[-\frac{aH_0}{v\beta E_0}e^{-\beta E_0\left(1-\frac{H}{H_0}\right)}\right]$$



v=0.1, 1, 3, beta=3 E_0=H_0=1, a=0.1d0

Néel-Arrhenius hysteresis

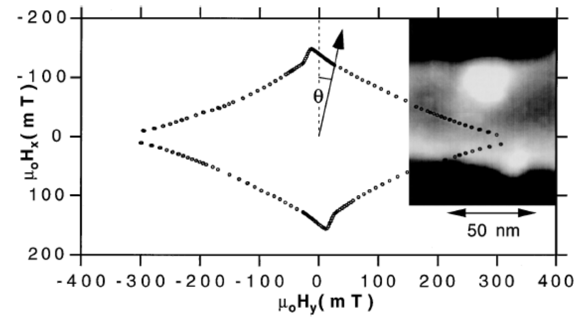


v=1, beta=3 E_0=H_0=1, a=0.1d0

Single magnetic model: Anisotropy

Single nano-size particle

Superparamagentism
Uniform rotation (Stoner-Wohlfarth)

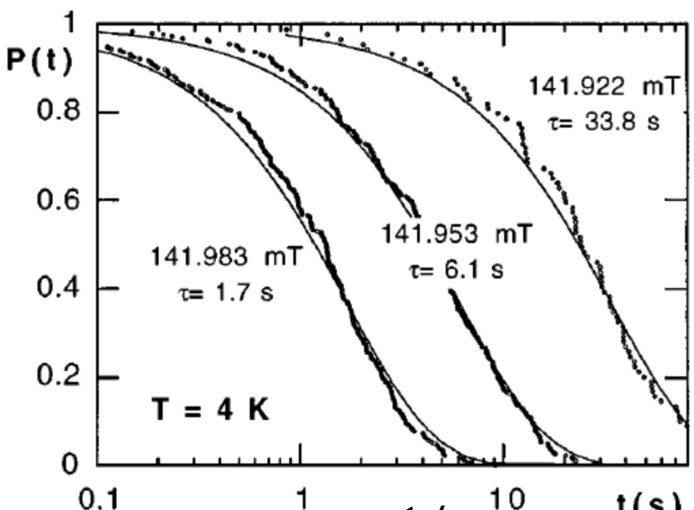


Co nanoparticle (25 ± 5 nm in diameter) on the microbridge

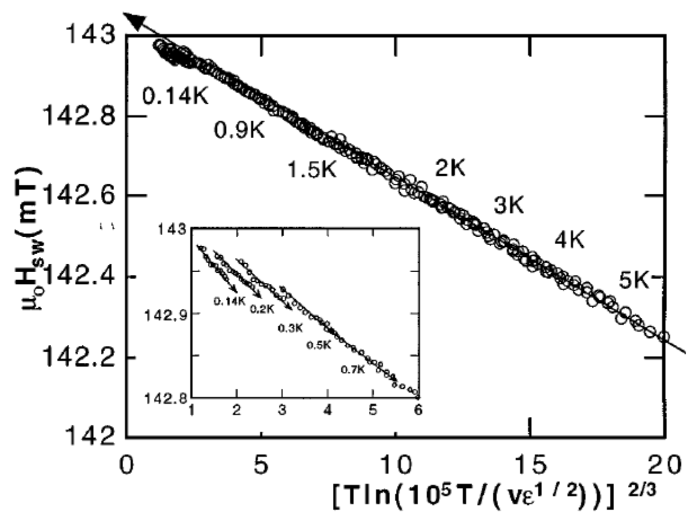
Ne'el-Brown model

W. Wernsdorfer, et at. PRL 78 (1997) 1791,
PRL 79 (1997) 4014.

Ni, Co, Dy
BaFeCoTiO

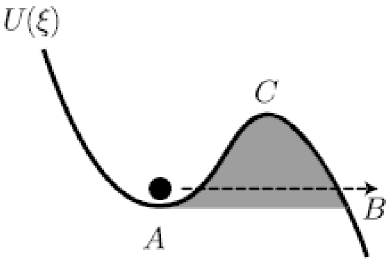


$\tau = 1/v$



$$\frac{\partial P(x)}{\partial t} = \frac{\partial}{\partial x}\left[-h(x)P + D\frac{\partial}{\partial x}P\right], \quad \frac{\partial F(x)}{\partial x} = -h(x)$$

Kramers' method



$$\frac{\partial P(M)}{\partial t} = \frac{\partial}{\partial m}\left[g_1(m)P + \varepsilon\frac{\partial}{\partial m}\left(g_2(m)P\right)\right]$$

$$g_1(m) = \left(\frac{\partial f(m)/\varepsilon D}{\partial m}\right), \quad g_2(m) = 1 - m\tanh[\beta(Jm+H)] \to D$$

current

$$-w := g_1(m)P + \varepsilon\frac{\partial}{\partial m}(DP) = e^{-f(m)/\varepsilon D}\frac{\partial}{\partial m}\left(e^{f(m)/\varepsilon D}(\varepsilon DP)\right)$$

$$\frac{w}{De^{-f(m)/\varepsilon D}} = -\frac{\partial}{\partial m}\left(e^{f(m)/\varepsilon D}\varepsilon DP\right)$$

$$w = \varepsilon D\frac{e^{f(m)/\varepsilon D}P\Big|_{\mathrm{B}}^{\mathrm{A}}}{\int_{\mathrm{A}}^{\mathrm{B}} e^{f(m)/\varepsilon D}dm}$$

$$e^{f(m_{\mathrm{A}})/\varepsilon D}P_{\mathrm{eq}}(m_{\mathrm{A}}) = e^{f(m_{\mathrm{B}})/\varepsilon D}P_{\mathrm{eq}}(m_{\mathrm{B}}), \quad e^{f(m_{\mathrm{A}})/\varepsilon D}P_0(m_{\mathrm{A}}) \simeq 1 >> e^{f(m_{\mathrm{B}})/\varepsilon D}P_{\mathrm{eq}}(m_{\mathrm{B}}) \simeq 0$$

$$f(m) = \Delta f - C(m - m_{\mathrm{C}})^2$$

$$\boxed{w \propto \varepsilon De^{-\Delta f/\varepsilon D} \propto e^{-\Delta F(m_{\mathrm{C}})/D}, \quad F(m) = Nf(m)}$$

H. A. Kramers, Physica 7, 284 (1940).

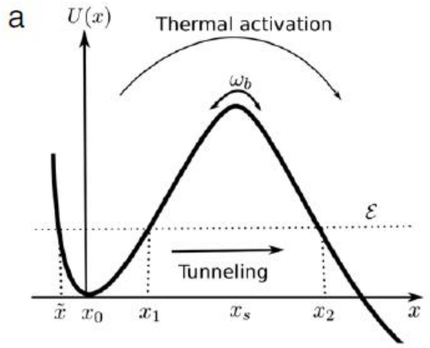
H. Tomita, A. Ito and H. Kidachi, Prog. Theor. Phys. 56, 786 (1976).
T. Mori, S. Miyashita and P. A. Rikvold: Phys. Rev. E 81, 011135 (1-10) (2010).

Quantum tunneling at each crossing

Macroscopic quantum tunneling and quantum–classical phase transitions of the escape rate in large spin systems.

Quantum tunneling

Wentzel–Kramers–Brillouin (WKB) method (Landau and Lifshitz, 1977)
"instanton" method (Coleman, 1977, 1985)

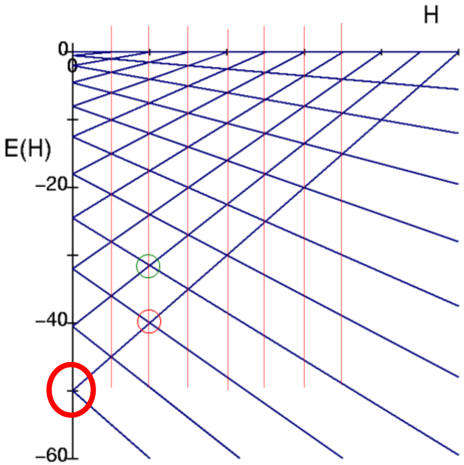


$$S[x(t)] = \int_0^{t'} dtL, \quad L = \frac{1}{2}m\left(\frac{dx}{dt}\right)^2 - U(x)$$

Large magnetic spin systems

Only for Single crossing

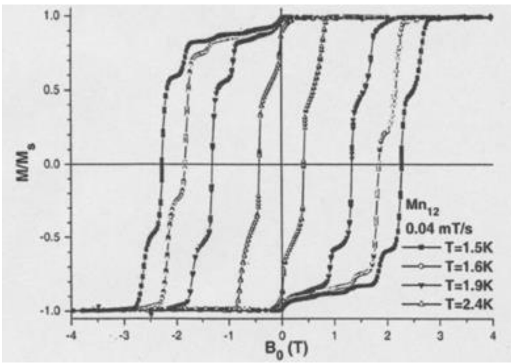
$$\Delta = \frac{4Ds^{3/2}}{\pi^{1/2}}\left(\frac{eh_x}{2}\right)^{2s}, \quad h_x = H_x/2Ds.$$

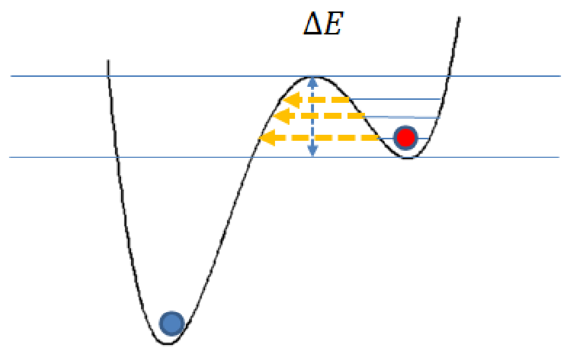


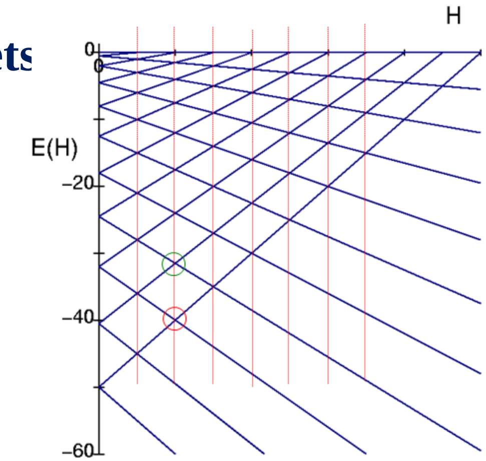
J. L. van Hemmen and A. Suto, in Quantum Tunneling of Magnetism –QTM'94, ed. By L. Gunter and B. Barbara, NATO Advanced Institutes, Ser. E. Vol 301 (Kluwer, Dordrecht 1995) p.19.
S. A. Owerre and M. B. Paranjape: Phys. Rep. 546, 1 (2015).

Quantum effect :
Multiple quantum tunneling with many degree of freedoms

Resonant Tunneling Phenomena: Single molecular magnets

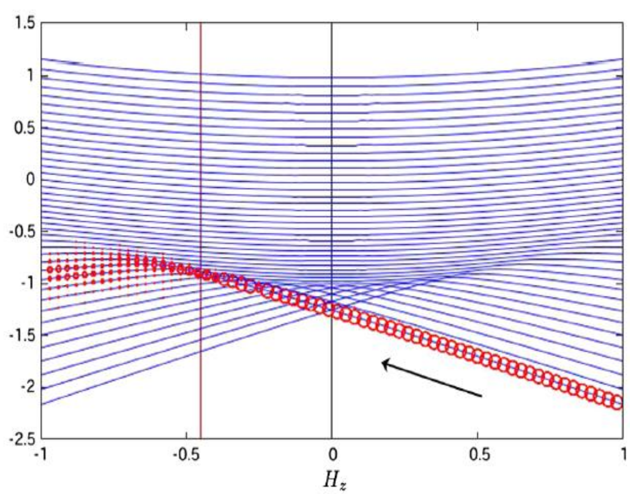






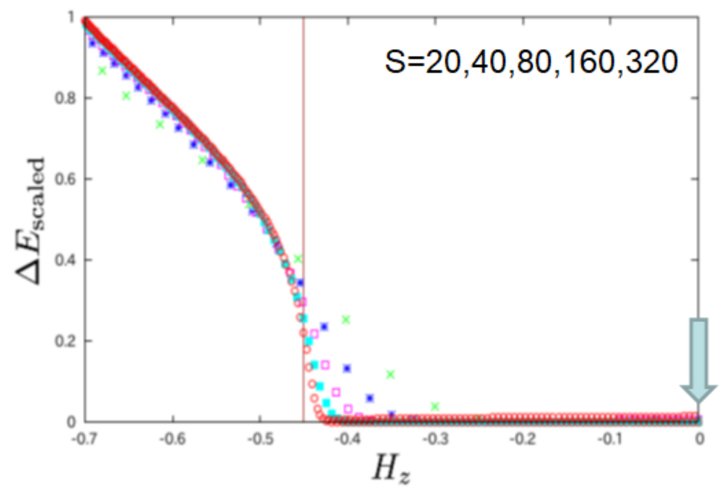
宮下精二：量子スピン系(岩波書店　2006)

Quantum spinodal Phenomena



scaled gap

$$\Delta E_{\rm scaled} = S\Delta E_k^{(S)}$$

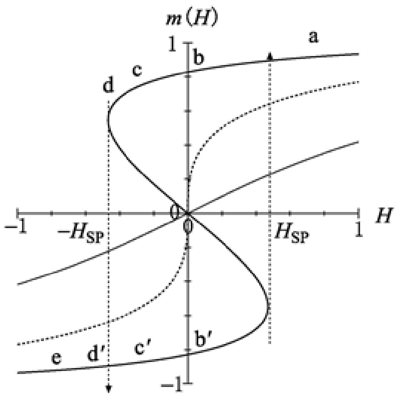


T. Hatomura, B. Barbara and S. Miyashita: Phys. Rev. Lett. 116 037203 (2016).

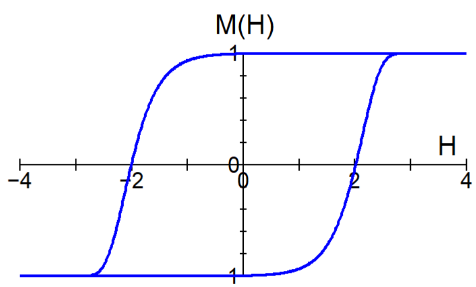
いろいろなヒステリシス

保磁力: Variety of hysteresis loops

Mean-field hysteresis

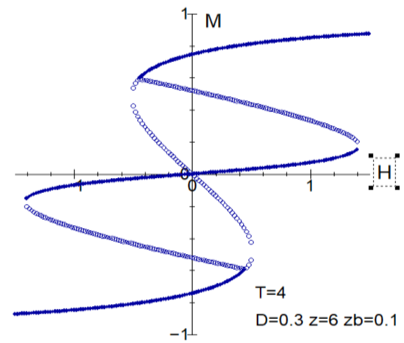


Neel-Arrhenius hysteresis

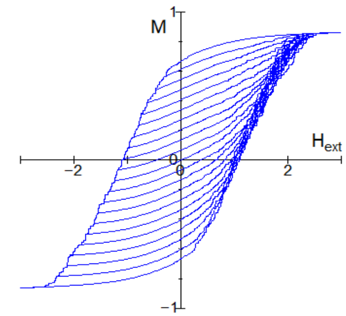


v=1, beta=3 E_0=H_0=1, a=0.1d0

Multi-domain



FORC
(first-order reversal curve)



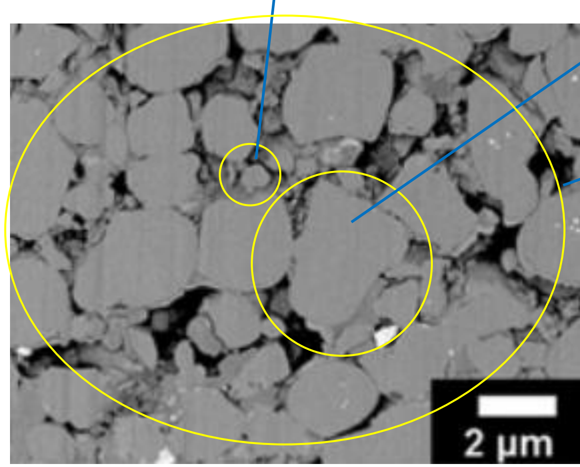
D=0.2, J=0.1, Dtheta=0.1, Dm=0.0, DHc=0.13
+ some reversible contributions from soft magnets

metastability

Thermal activation

Dipole-dipole Int.

Ensemble



Microscopic approach

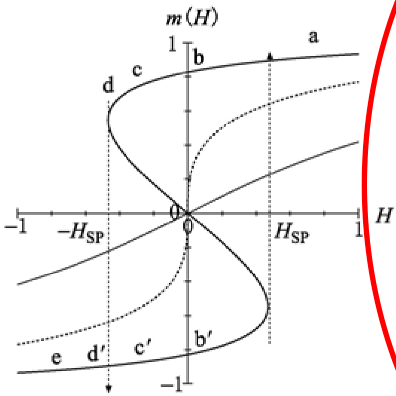
$$\langle A(T)\rangle = \frac{\mathrm{Tr}\, A e^{-\beta\mathcal{H}}}{\mathrm{Tr}\, e^{-\beta\mathcal{H}}}$$

a microscopic Hamiltonian $\mathcal{H}$

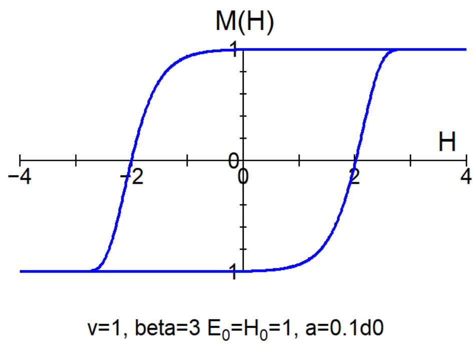
いろいろなヒステリシス

保磁力: Variety of hysteresis loops

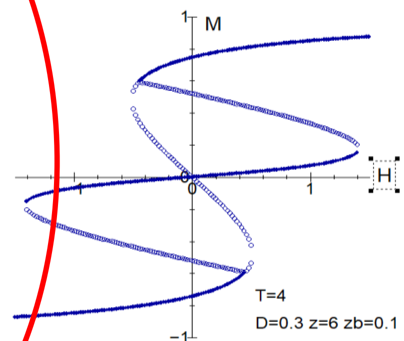
Mean-field hysteresis



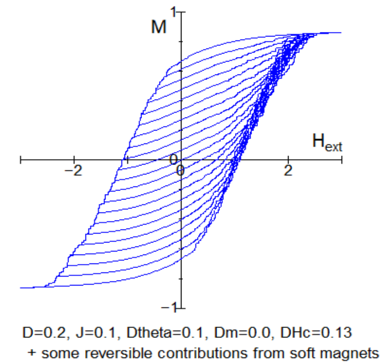
Neel-Arrhenius hysteresis



Multi-domain



FORC
(first-order reversal curve)

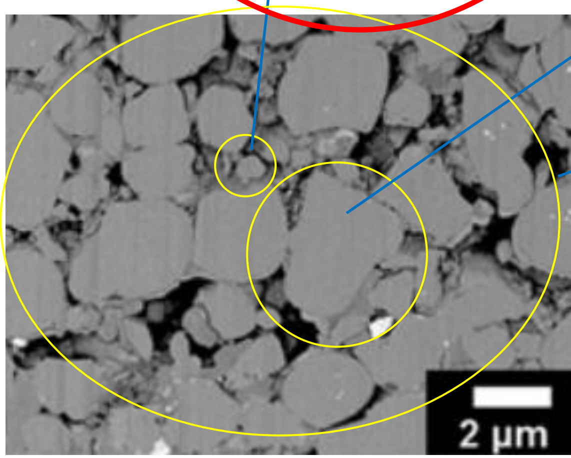


metastability

Thermal activation

Dipole-dipole Int.

Ensemble

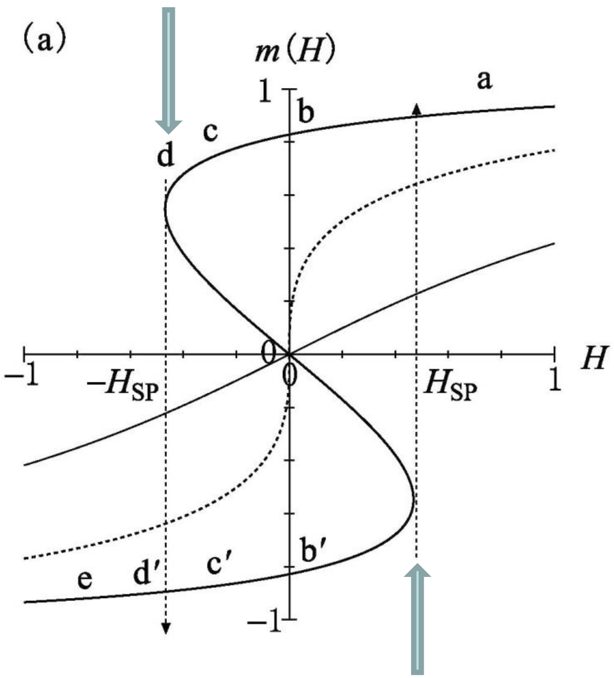


Microscopic approach

$$\langle A(T)\rangle = \frac{\mathrm{Tr}\ A e^{-\beta\mathcal{H}}}{\mathrm{Tr}\ e^{-\beta\mathcal{H}}}$$

a microscopic Hamiltonian $\mathcal{H}$

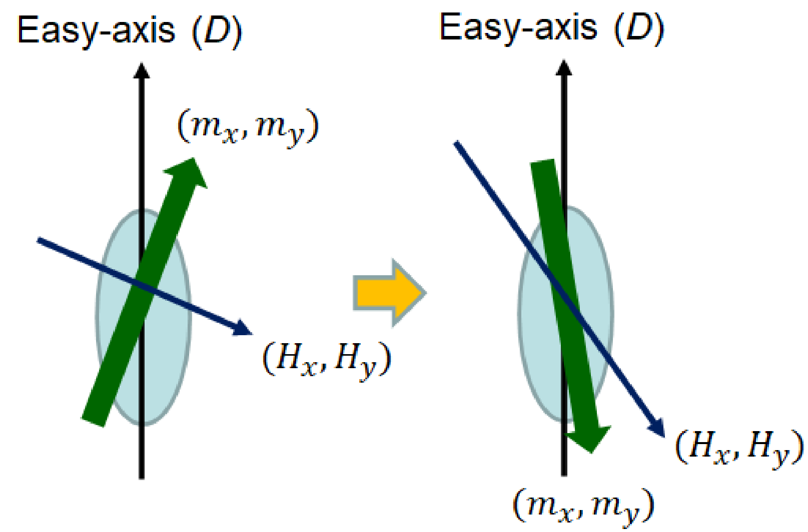
ベクトル模型：一様回転模型



Ising model

$$m = \pm 1$$

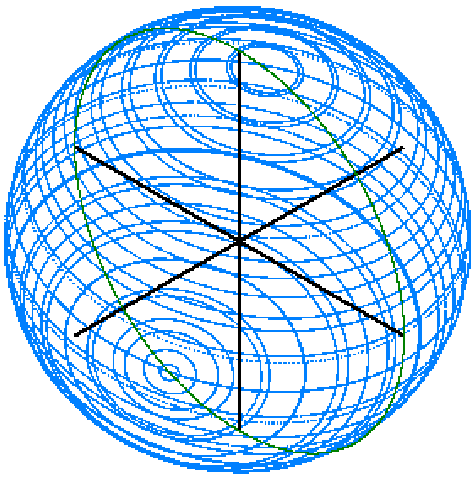
Stoner-Wohlfarth rotation



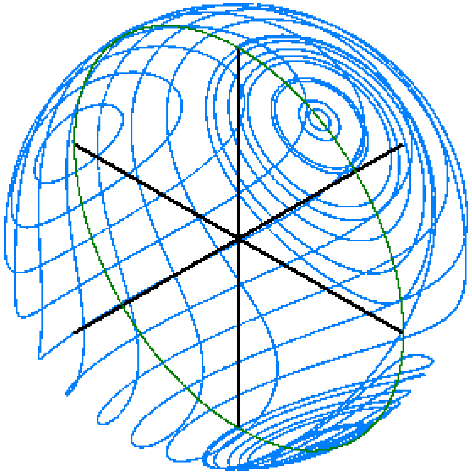
Ising-like Heisenberg model
Easy-axtis-type magnet

$$E = -Dm_z^2 - H_z m_z - H_x m_x,$$
$$m_x^2 + m_y^2 + m_z^2 = 1$$

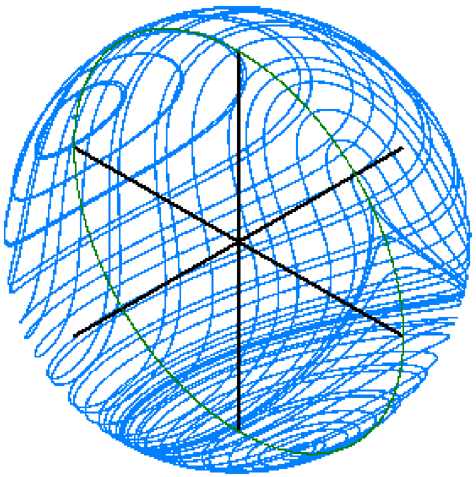
Change of the trajectory with Hz D=1,Hx=1



D=1, Hx=1, Hz=4

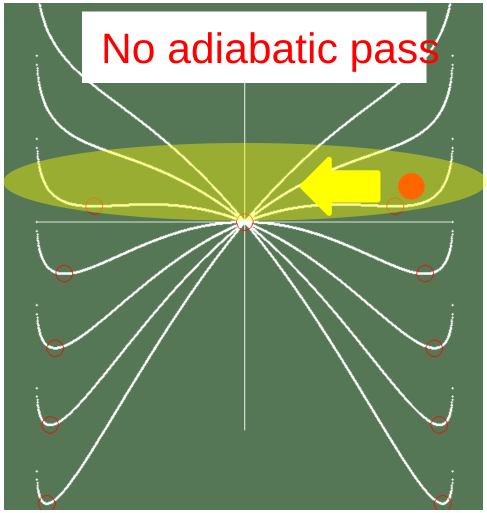


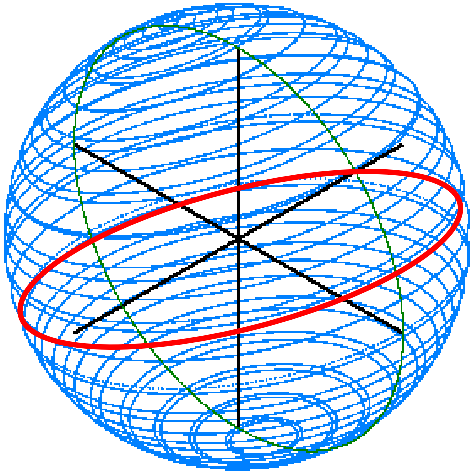
D=1, Hx=1, Hz=0



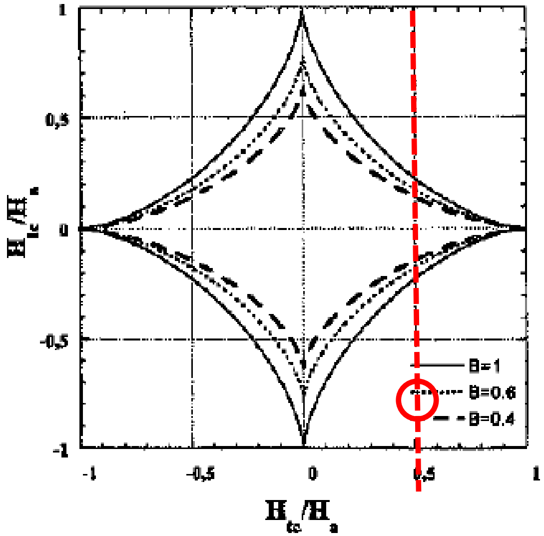
D=1, Hx=1, Hz=hZC=−0.4502

Dynamical process





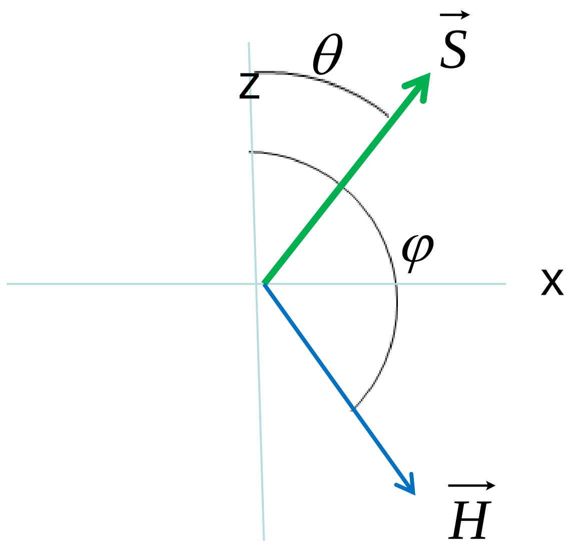
D=1, Hx=1, Hz=hZC=−4

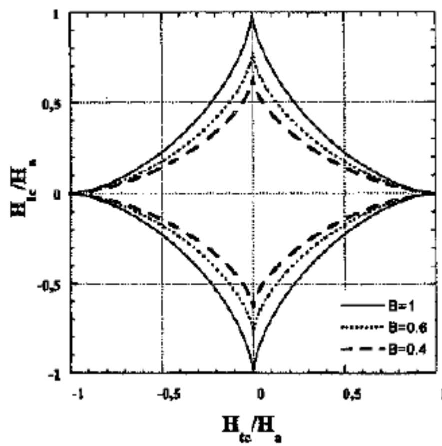


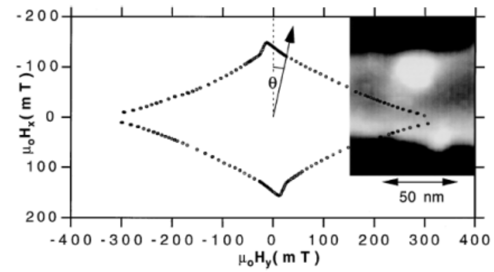
Stoner-Wohlfarth (SW) process

End of the metastability

$$E = -Dm_z^2 - H_z m_z - H_x m_x, \quad m_x^2 + m_y^2 + m_z^2 = 1$$







$$H = \frac{2D}{(\sin^{2/3}\varphi + \cos^{2/3}\varphi)^{3/2}},$$

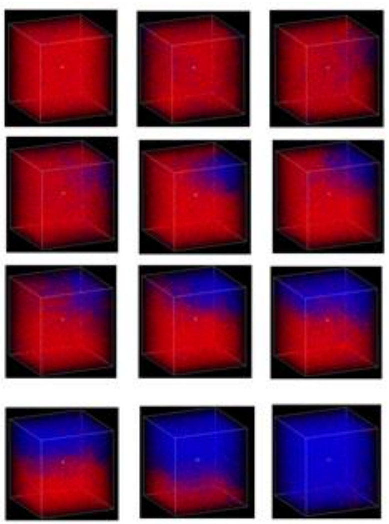
$$\text{i.e.,} \quad (2D)^{2/3} = (H_x)^{2/3} + (H_z)^{2/3}$$

E. C. Stoner and E. P.Wohlfarth, Phil. Trans. R. Soc. A 240, 599 (1948).

Dynamics of grain magnetization at finite temperature
-- real-time approach (stochastic -LLG equation) --

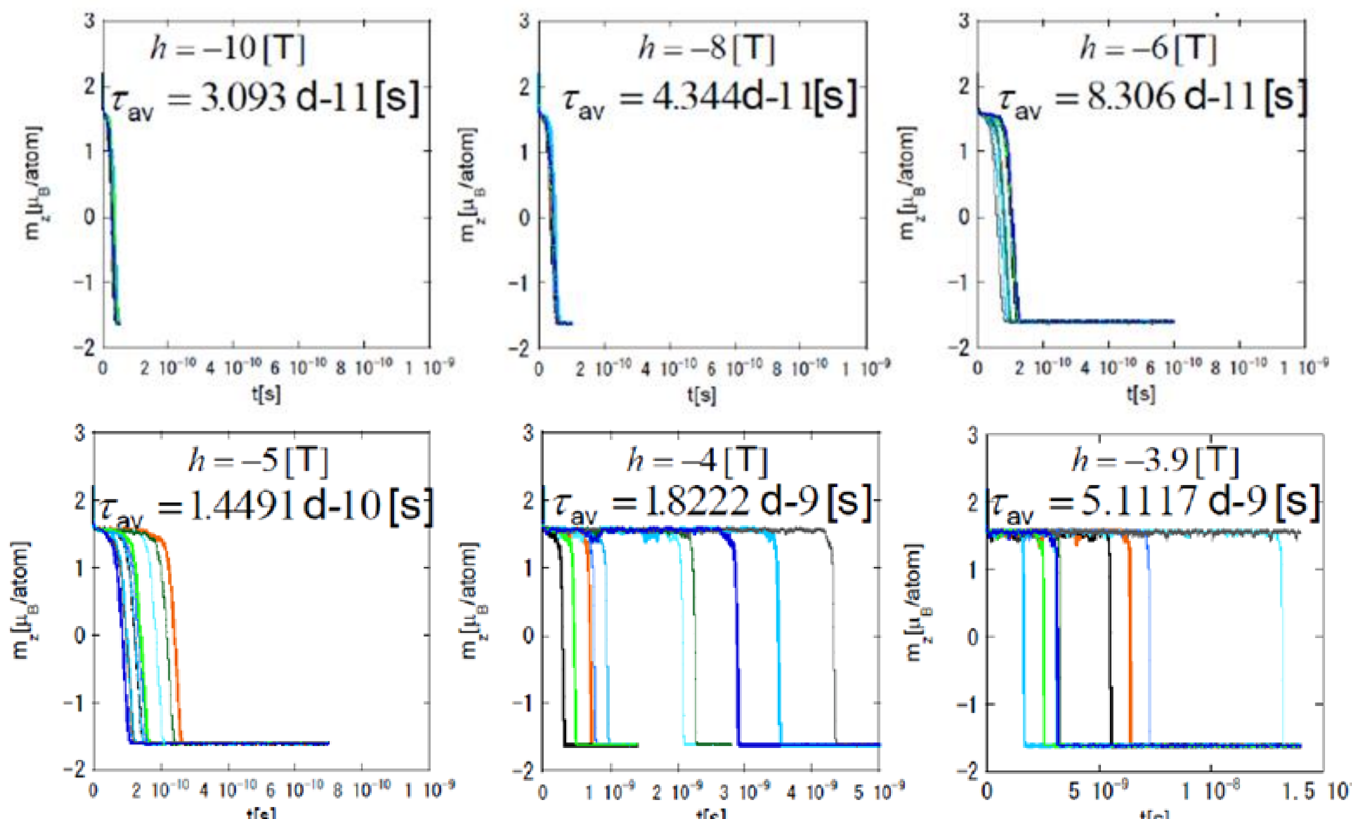
$$\frac{d\mathbf{M}_i}{dt} = -\frac{\gamma}{1+\alpha_i^{\,2}}\mathbf{M}_i \times \mathbf{H}_{\mathrm{eff},i} - \frac{\alpha_i \gamma}{(1+\alpha_i^{\,2})M_{\mathrm{i}}}\mathbf{M}_i \times (\mathbf{M}_i \times \mathbf{H}_{\mathrm{eff},i}), \quad \mathbf{H}_{\mathrm{eff,i}} \rightarrow \mathbf{H}_{\mathrm{eff,i}} + \boldsymbol{\xi}_{\mathrm{fl,i}} \quad i = 1, 2, \cdots, \mathrm{N}$$

$$\left\langle \xi_i^{\alpha}(t) \right\rangle = 0, \ \left\langle \xi_i^{\alpha}(t)\xi_j^{\beta}(s) \right\rangle = 2D_i\delta_{ij}\delta_{\alpha\beta}\delta(t-s). \qquad \frac{\alpha_i}{M_i} = \frac{\gamma D_i}{k_B T}$$



400 [K]

Magnetization reversal time



反転磁化分布の方法

独立核生成プロセス

$$P(t)\Delta t = (1 - p\Delta t)^{t/\Delta t} p\Delta t = pe^{-pt}\Delta t.$$

逆磁場下における磁化の緩和時間（τ）　初期状態all upでt秒間観測

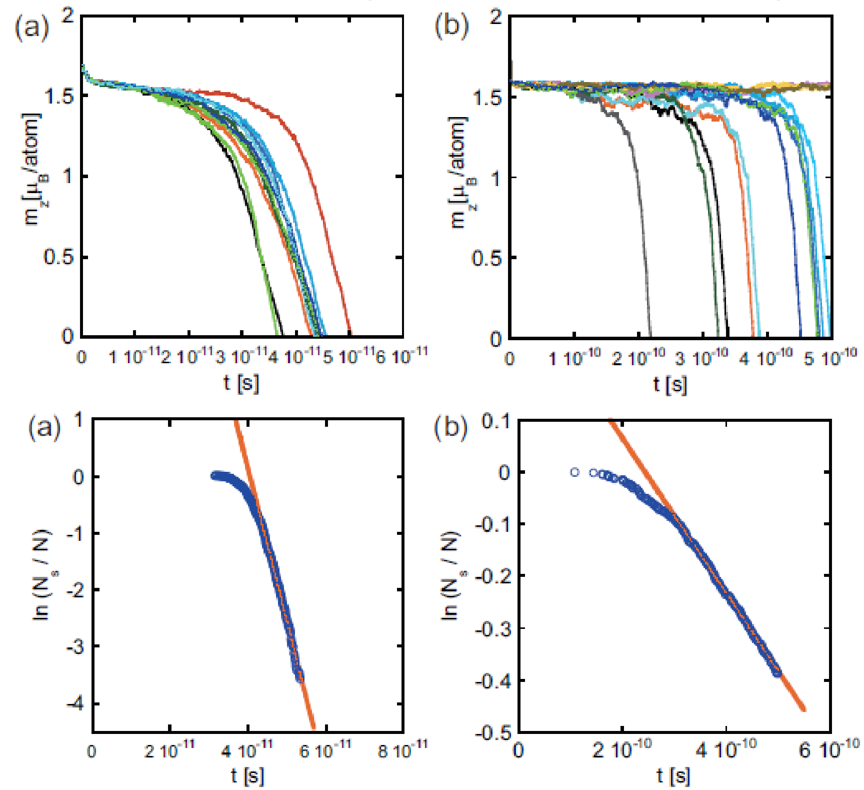
$$\ln(N_S / N) = -pt \quad \tau = 1/p$$

N：サンプル数
Ns：磁化反転せず
生き残ったサンプル数

Merit: easy to remove
initial region

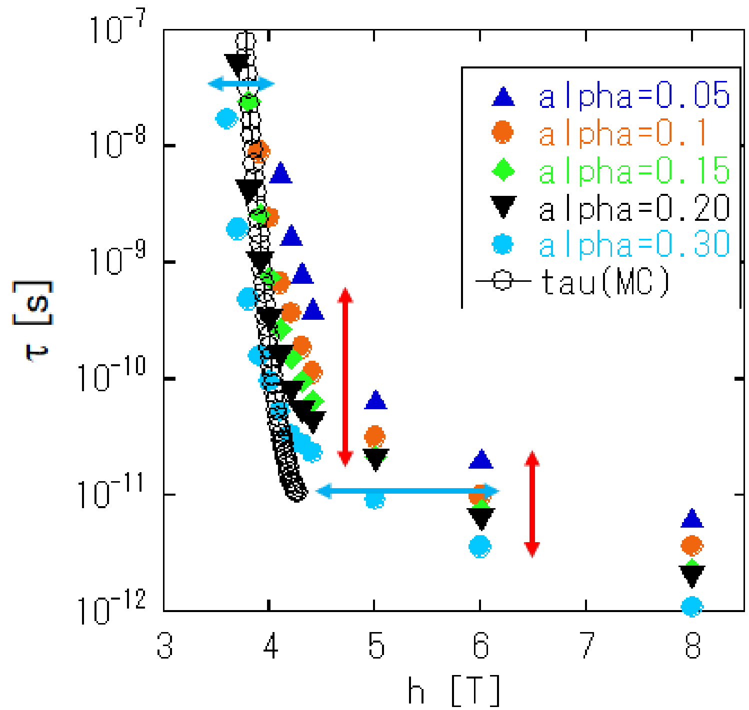
H=8T
deterministic region

H=4.1T
stochastic region



M. Nishino, I. E. Uysal , T. Hinokihara, S. Miyashita, Phys. Rev. B102, 020413(R) (1-5).

Dependence on damping constant (α)



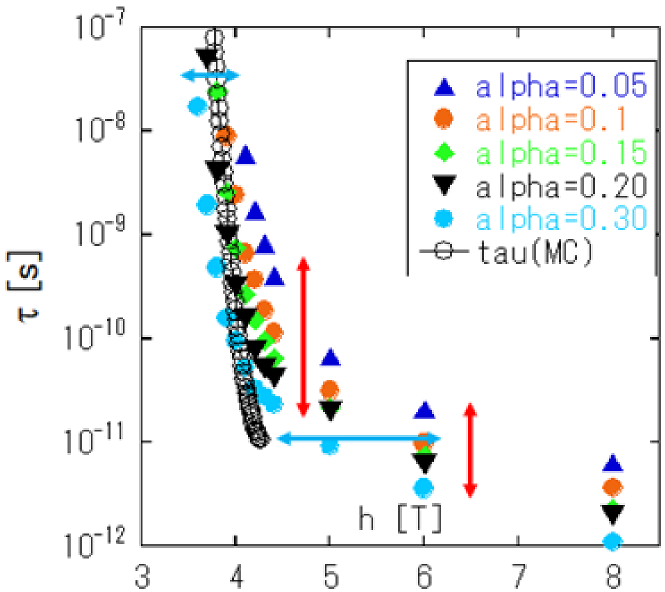
Energy barrier

Stochastic jump

No Energy barrier

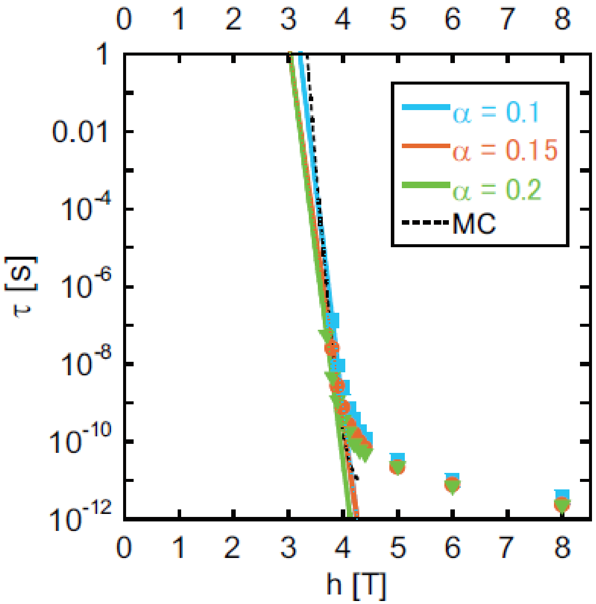
Deterministic Avrami process

Estimation of the coercive field



Large dependence of τ on damping constant (α)

Dynamical phase transition
(sharp increase of the relaxation time τ
between deterministic and stochastic regions)



extrapolation

$$\tau(h) = Ae^{-ah} + Be^{-bh} = Ae^{-ah}\left(1 + Ce^{-dh}\right)$$

$$\chi_2 = \sum_k \frac{1}{\sigma_k^2}\left(\log_{10}(\tau(h_k)) - \log_{10}(Ae^{-ah_k}(1 + Ce^{-dh_k}))\right)^2$$

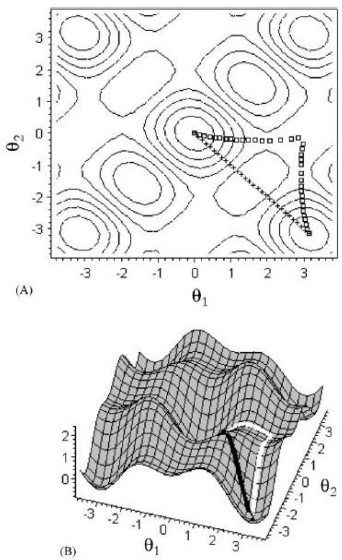
$h_c \cong 3T$

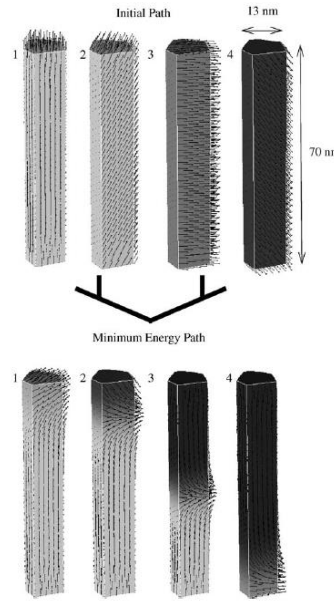
(cf. $h_c \cong 3.3T$ Monte Carlo method Toga et al.)

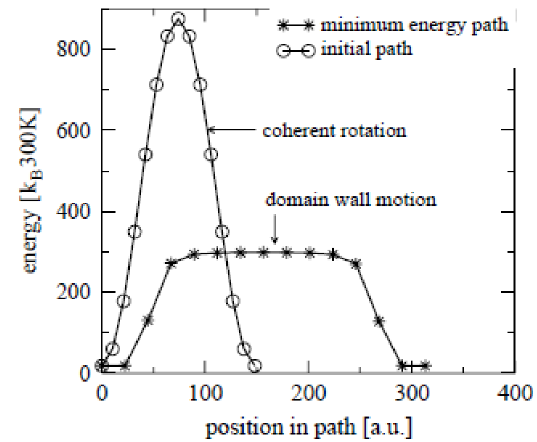
M. Nishino, I. E. Uysal , T. Hinokihara, S. Miyashita, Phys. Rev. B102, 020413(R).

Estimations of the relaxation time by the energy barrier

Minimum energy paths (MEP) method







$$f\,\exp(-\Delta E(H_{\mathrm{C}})/k_{\mathrm{B}}T)=1$$

R. Dittrich, et al., *J. Mag. Mag. Mat.*, **250**, L12 (2002).
E. Weinan, et al., *J. Chem. Phys*. **126**, 164103 2007.

Free-energy landscape method

Wang-Landau method for $F(m)$: (Multi-Canonical MC method)

F. Wang and D. P. Landau, Phys. Rev. Lett. 86, 2050 (2001).

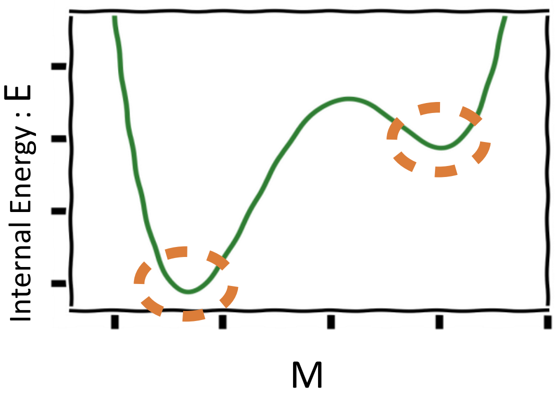
Free energy barrier for metastable state

$$F(T,H;M) = -k_{\rm B}T \ln Z(T,H;M)$$

$$Z(T,H;M) = {\rm Tr} e^{-\beta\mathcal{H}_0 - \beta H \sum_i^N S_i^z} \times \delta(\sum_i^N S_i^z - M)$$

$$P(M) = \frac{Z(T,H;M)}{Z(T,H)}$$

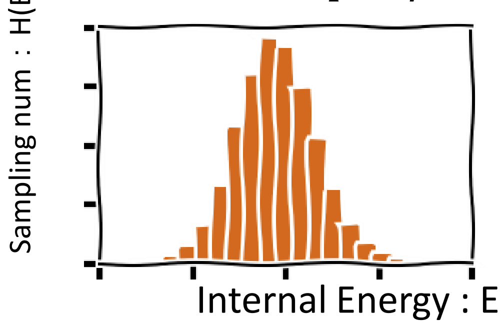
$$F(T,H;M) - F(T,H;0) = -k_{\rm B}T \ln \frac{P(M)}{P(0)}$$



Wang-Landau MC method

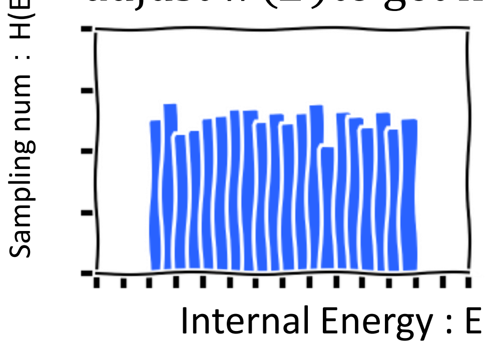
Samples regions around energy minima

$w(E) = \exp(-\beta E)$



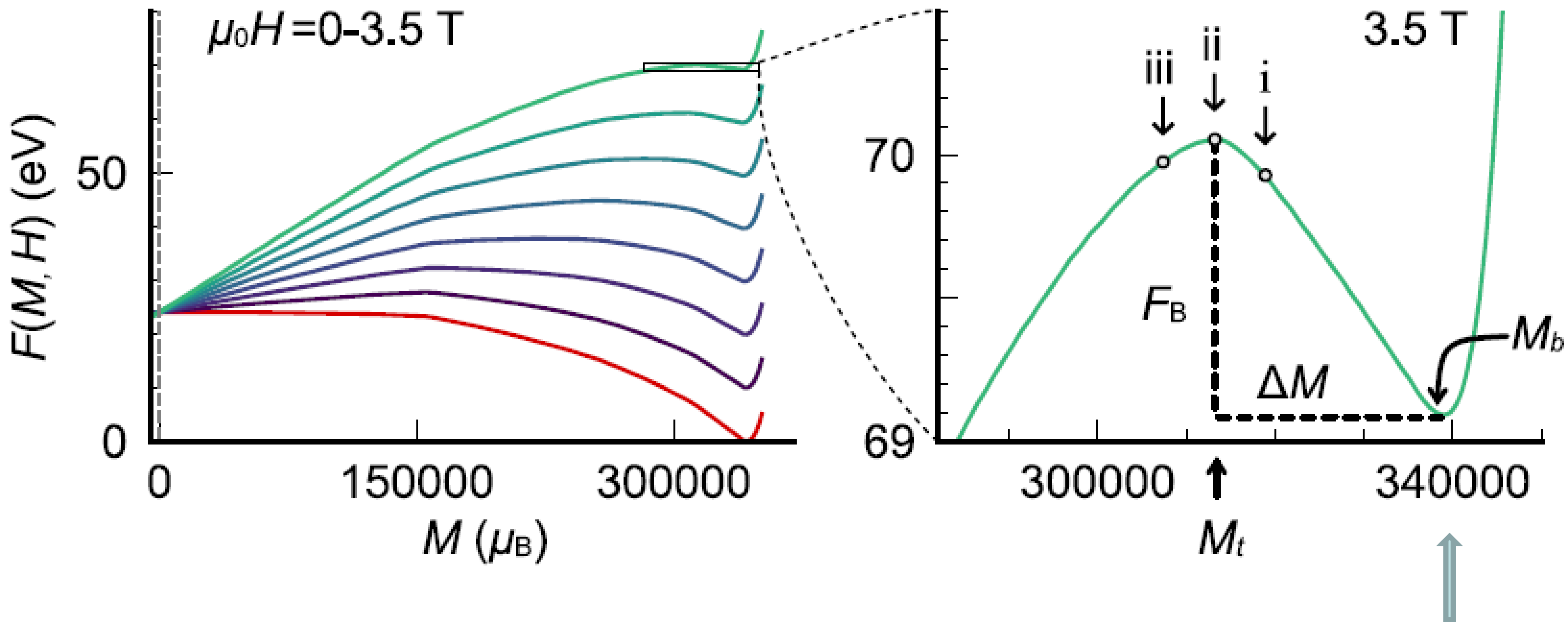
Samples all energy regions

adjust $w(E)$ to get flat $H(E)$



F. Wang and D. P. Landau, Phys. Rev. Lett. 86 2050 (2001).

Free energy as a function of M



Metastable state

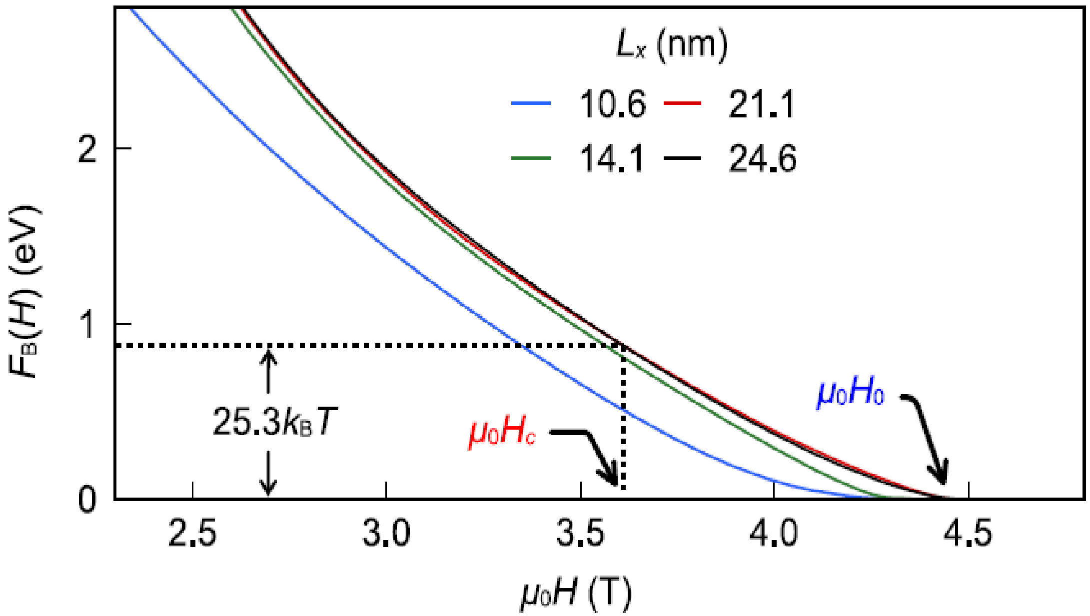
$$F(T, H; M) = F(T, 0; M) - HM$$

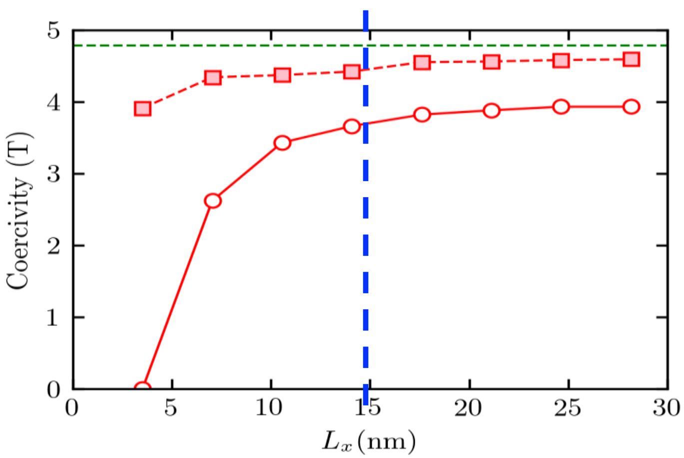
Free energy barrier

$$F_\mathrm{B}(T, H) = F(T, H; M_t) - F(T, H; M_b)$$

Y. Toga, S. Miyashita, A. Sakuma and T. Miyake: Role of atomic-scale thermal fluctuations in the coercivity, Nature partner journal (npj) Computational Materials (2020) 6:67.

Thermal activated coercivity

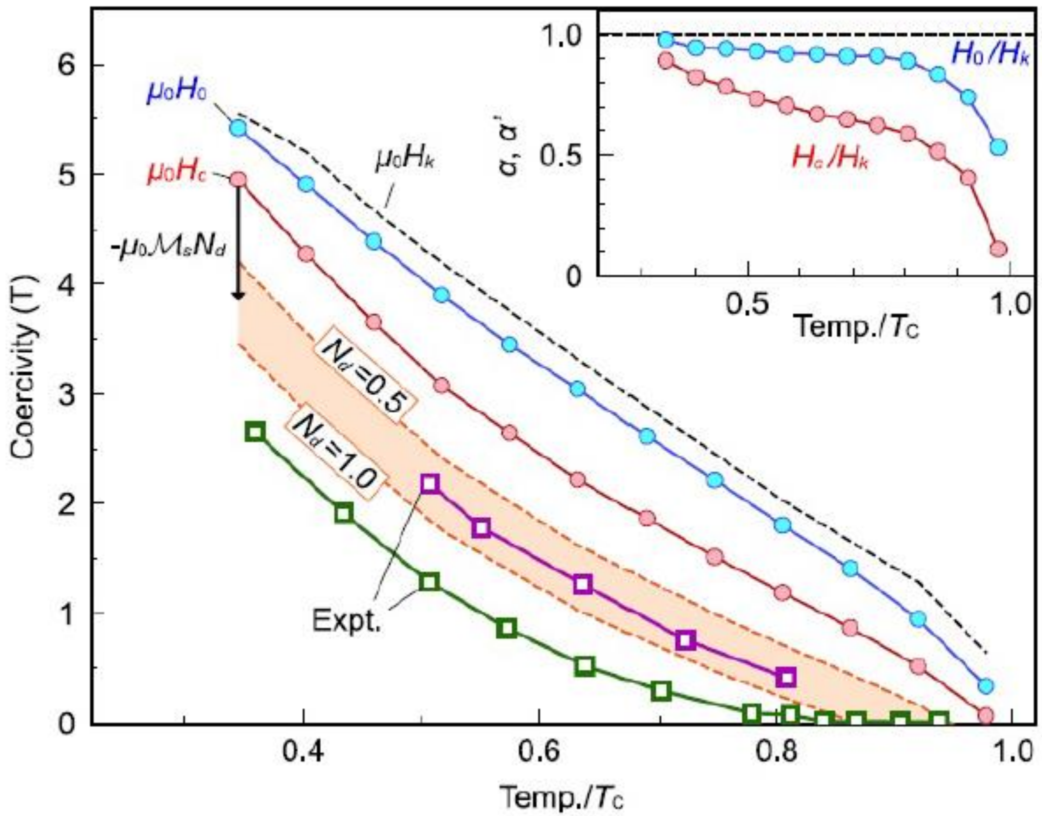




$$r = N_{\mathrm{contact}} e^{-\beta F_{\mathrm{B}}}, \quad \tau = \frac{1}{r} = \frac{e^{\beta F_{\mathrm{B}}}}{N_{\mathrm{contact}}}$$

$$F_{\mathrm{B}} = k_{\mathrm{B}} T \times \ln(10^{11}) = 25.3 k_{\mathrm{B}} T$$

Temperature dependence of coercivities

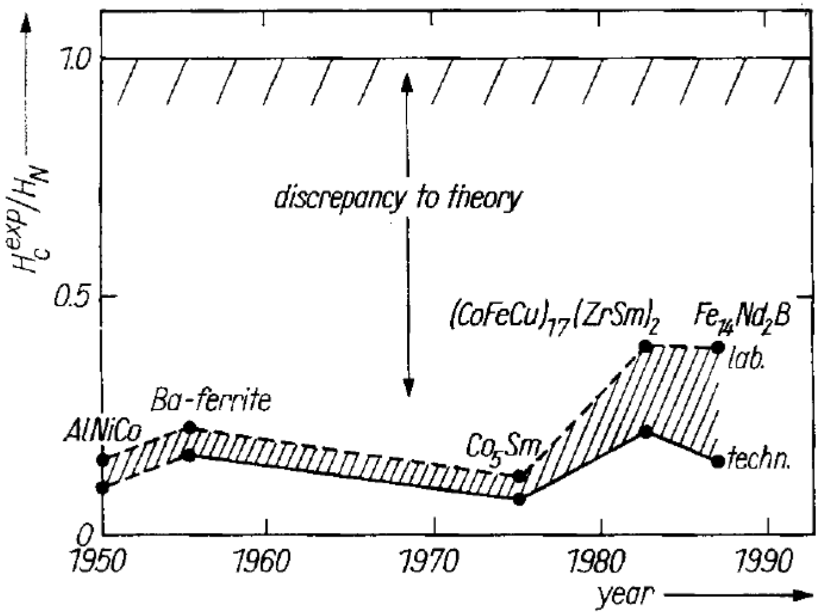


Kronmueller equation

$$H_{\text{coercivity}} = \alpha H_k - H_t - \mathcal{M}_s N_d = \alpha' H_k - \mathcal{M}_s N_d$$

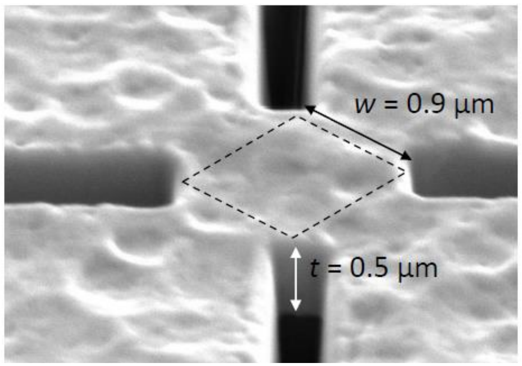
Y. Toga, S. Miyashita, A. Sakuma and T. Miyake: Role of atomic-scale thermal fluctuations in the coercivity, Nature partner journal (npj) Computational Materials (2020) 6:67.

Large reduction of the coercive field

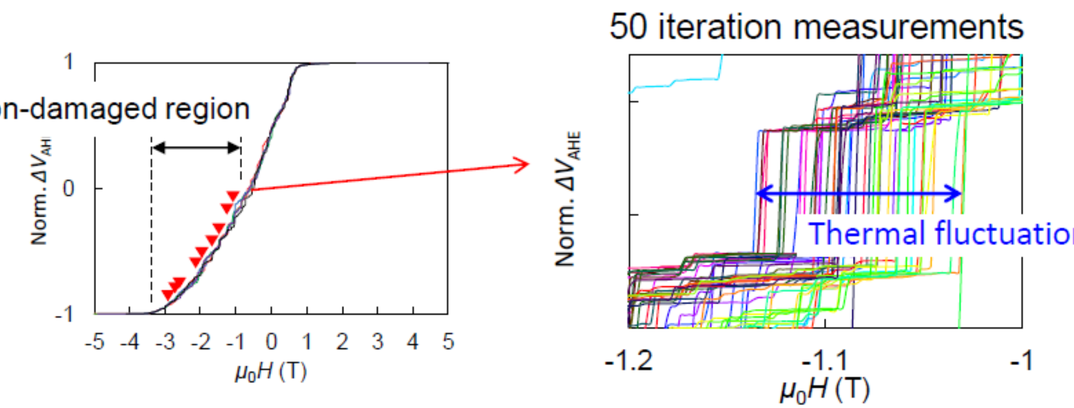


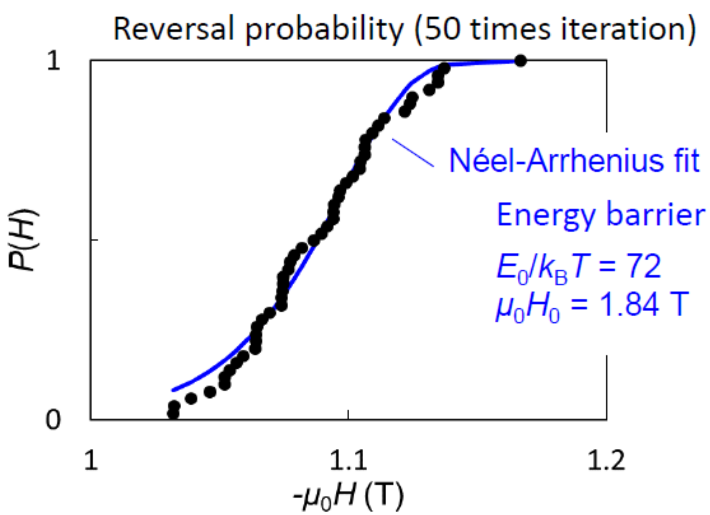
H. Kronmueller, Phys. Stat. Sol. (b) 144 (1987) 385.

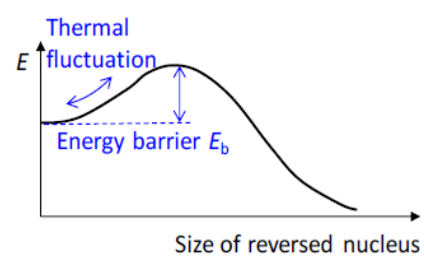
Experimental approach to grain reversal



Approximately 60 grains (*d*300nm, *t*80 nm)





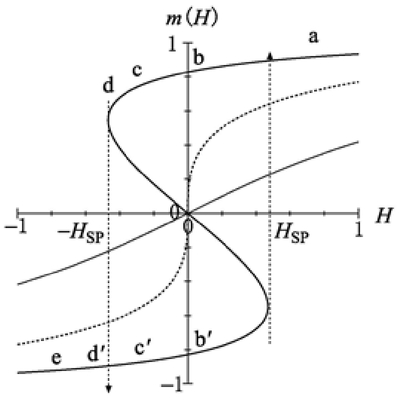


Takahiro Yomogita, Satoshi Okamoto, Nobuaki Kikuchi, Osamu Kitakami, Hossein Sepehri-Amin, Yukiko K. Takahashi, Tadakatsu Ohkubo, Kazuhiro Hono, Keiko Hioki, and Atsushi Hattori, to be published in Acta Mater (2020).

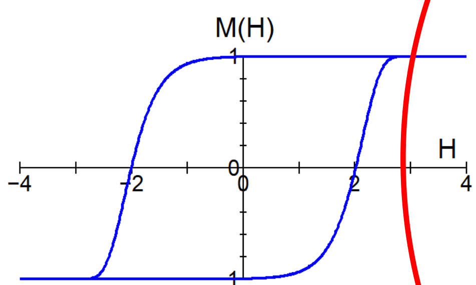
いろいろなヒステリシス

保磁力: Variety of hysteresis loops

Mean-field hysteresis

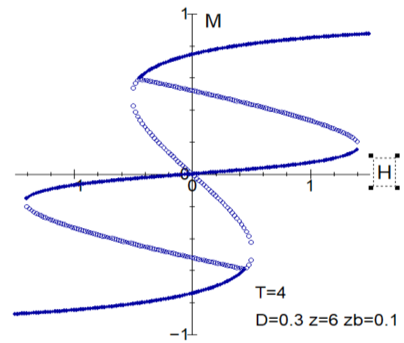


Neel-Arrhenius hysteresis

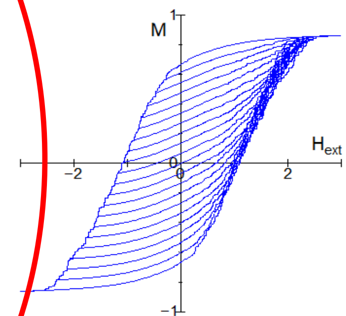


v=1, beta=3 $E_0=H_0=1$, a=0.1d0

Multi-domain



FORC
(first-order reversal curve)



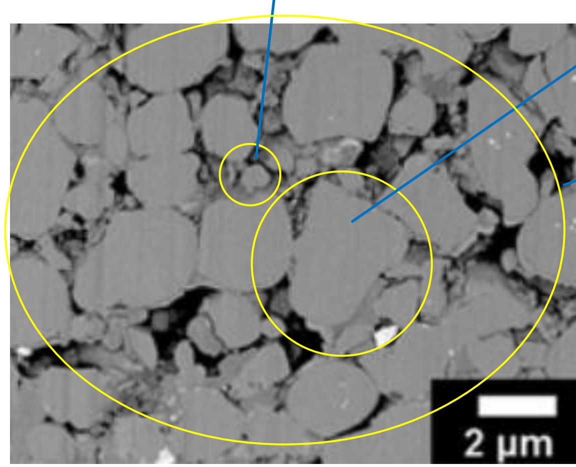
D=0.2, J=0.1, Dtheta=0.1, Dm=0.0, DHc=0.13
+ some reversible contributions from soft magnets

metastability

Thermal activation

Dipole-dipole Int.

Ensemble



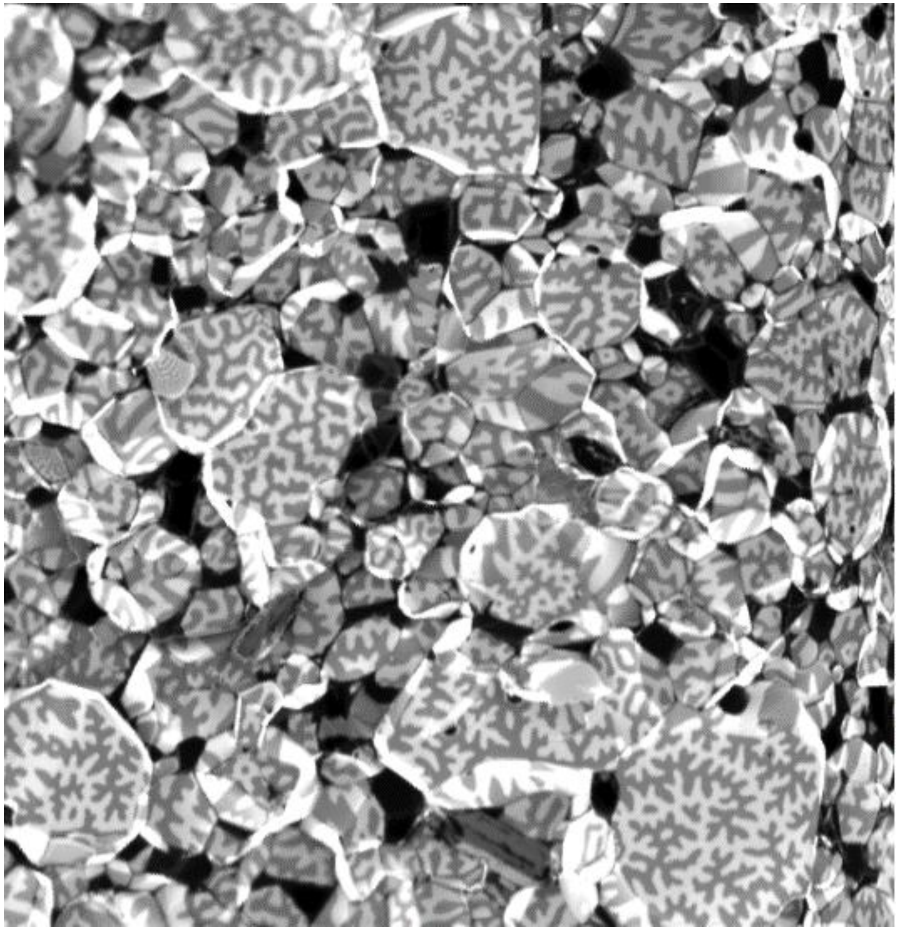
Microscopic approach

$$\langle A(T)\rangle = \frac{\mathrm{Tr}\ A e^{-\beta\mathcal{H}}}{\mathrm{Tr}\ e^{-\beta\mathcal{H}}}$$

a microscopic Hamiltonian
$\mathcal{H}$

Multi-domain structure and coercivity

Thermal demagnetization



Maze pattern in a grain

T. Nakamura, K. Toyoki, M. Suzuki

Effect of the dipole-dipole interaction

$$\hat{H}_{dip} = D\sum_{i<j}\frac{1}{4\pi\mu_0}\frac{1}{r_{ij}^{\ 3}}\left(\mathbf{S}_i\cdot\mathbf{S}_j - \frac{3(\mathbf{r}_{ij}\cdot\mathbf{S}_i)(\mathbf{r}_{ij}\cdot\mathbf{S}_j)}{r_{ij}^{\ 2}}\right),$$

Computational cost: (NxN) ⇨ Algorithms for faster simulation

Stochastic Cut-off method (SCO)：

$$\overline{V}(S_i,S_j) = V(S_i,S_j) - \beta^{-1}\log\left[1-P(S_i,S_j)\right]$$
$$P(S_i,S_j) = \exp\left[\beta\left(V(S_i,S_j)-V^*\right)\right]$$

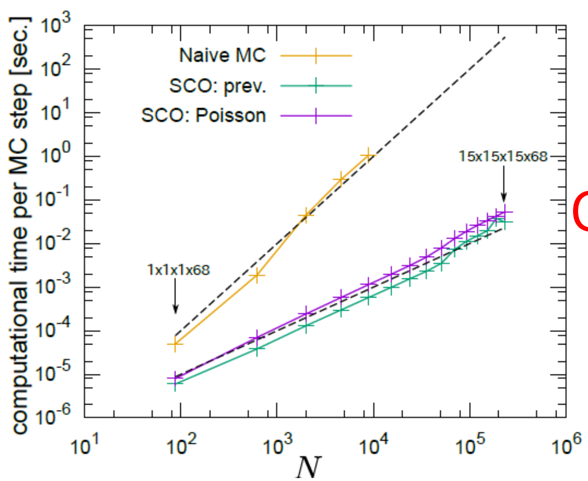
M. Sasaki and F. Matsubara: J. Phys. Soc. Jpn. **77** 024004 (2008).

Walker's algorithm：

K. Fukui and S. Todo: J. Comput. Phys. 228 2629 (2009).

A Hybrid method：

CPU/MCS

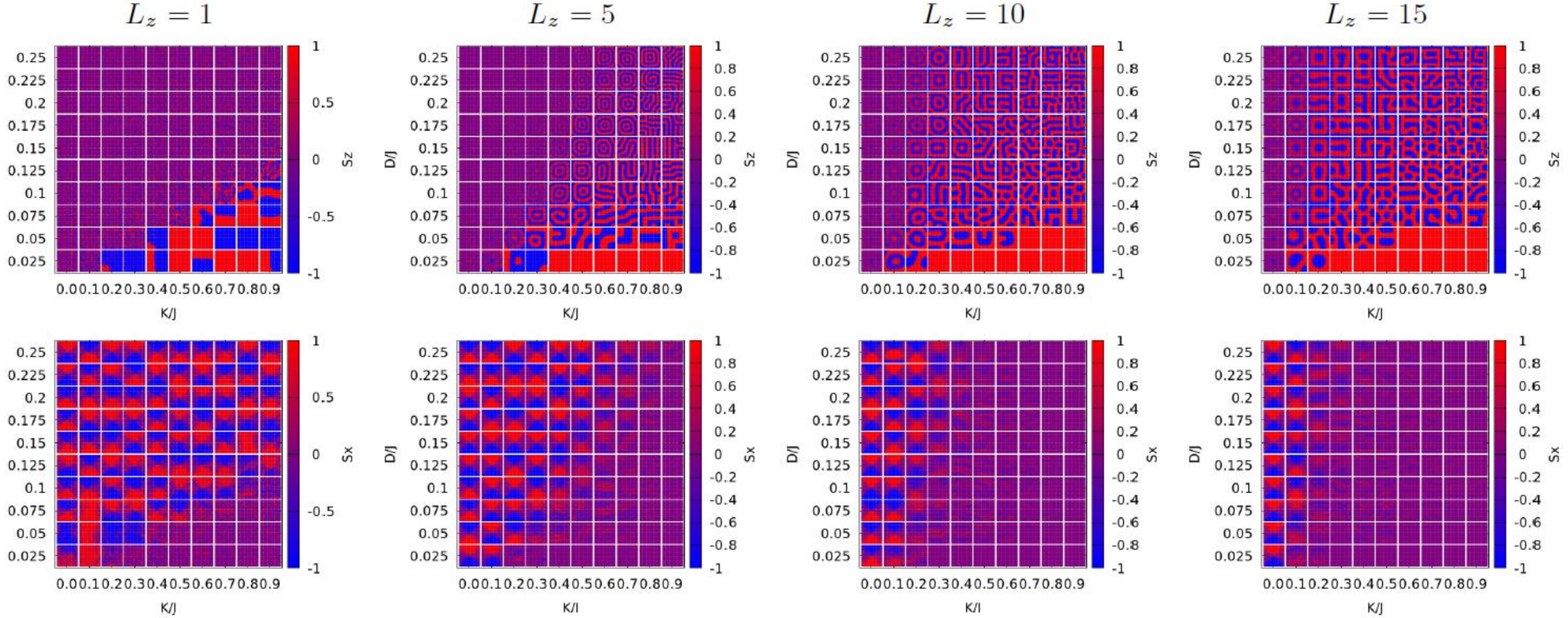


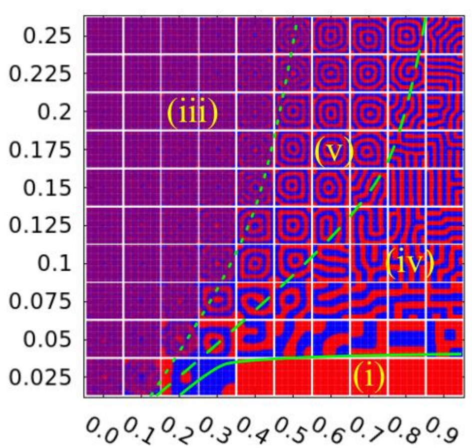
O(NlogN)

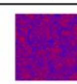
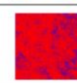
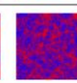
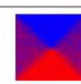
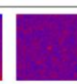
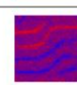
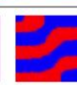
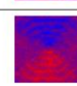
T. Hinokihara, M. Nishino, Y. Toga, and S. Miyashita, Phys. Rev. B97, 104427 (2018).

Paramerter (K/J, D/J) depencence of magnetic patterns in thin films

Multidomain structure (64x64;Lz): thermal-quech



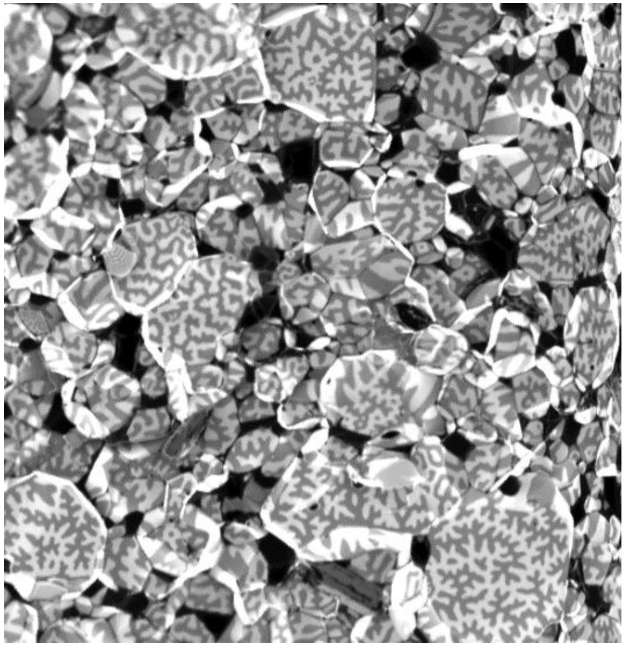


	S_x	S_z	K/J	D/J	L_z
(i)			0.8	0.025	5
(ii)			0.1	0.025	1
(iii)			0.1	0.225	5
(iv)			0.7	0.075	5
(v)			0.6	0.175	5

T. Hinokihara and S. Miyashita: arXive:2009.11574, Phys. Rev. B(2021) to be published.

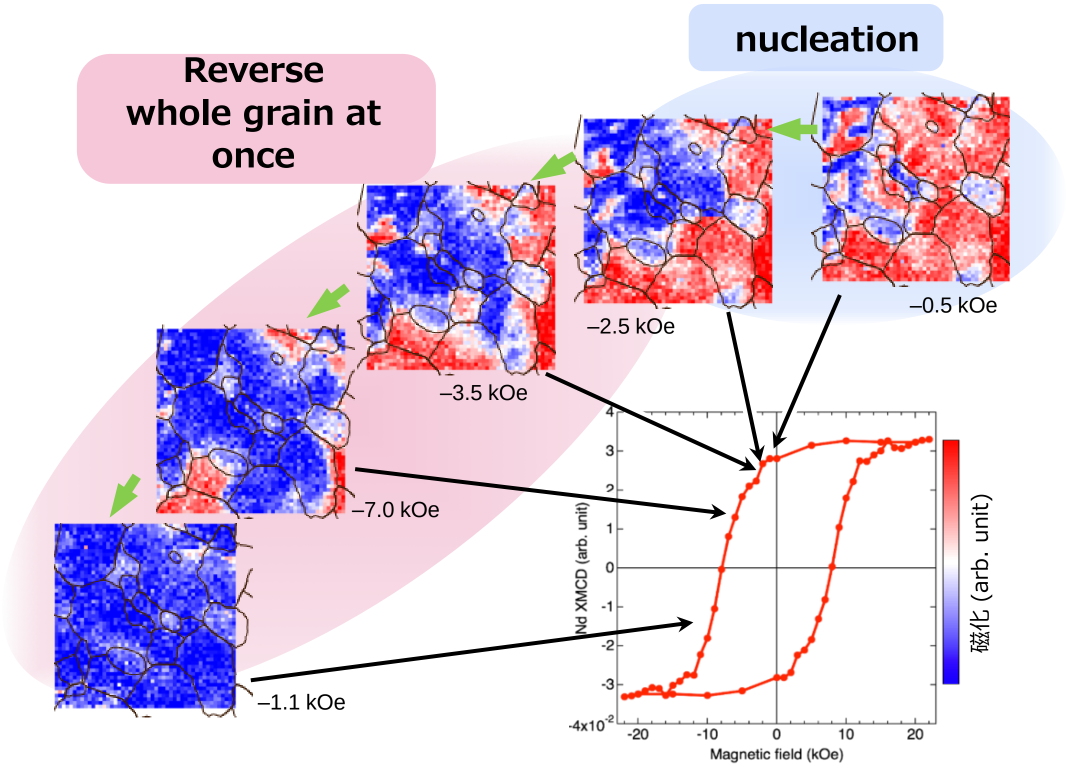
Multi-domain structure and coercivity

Thermal demagnetization



Maze pattern in a grain

From magnetized state



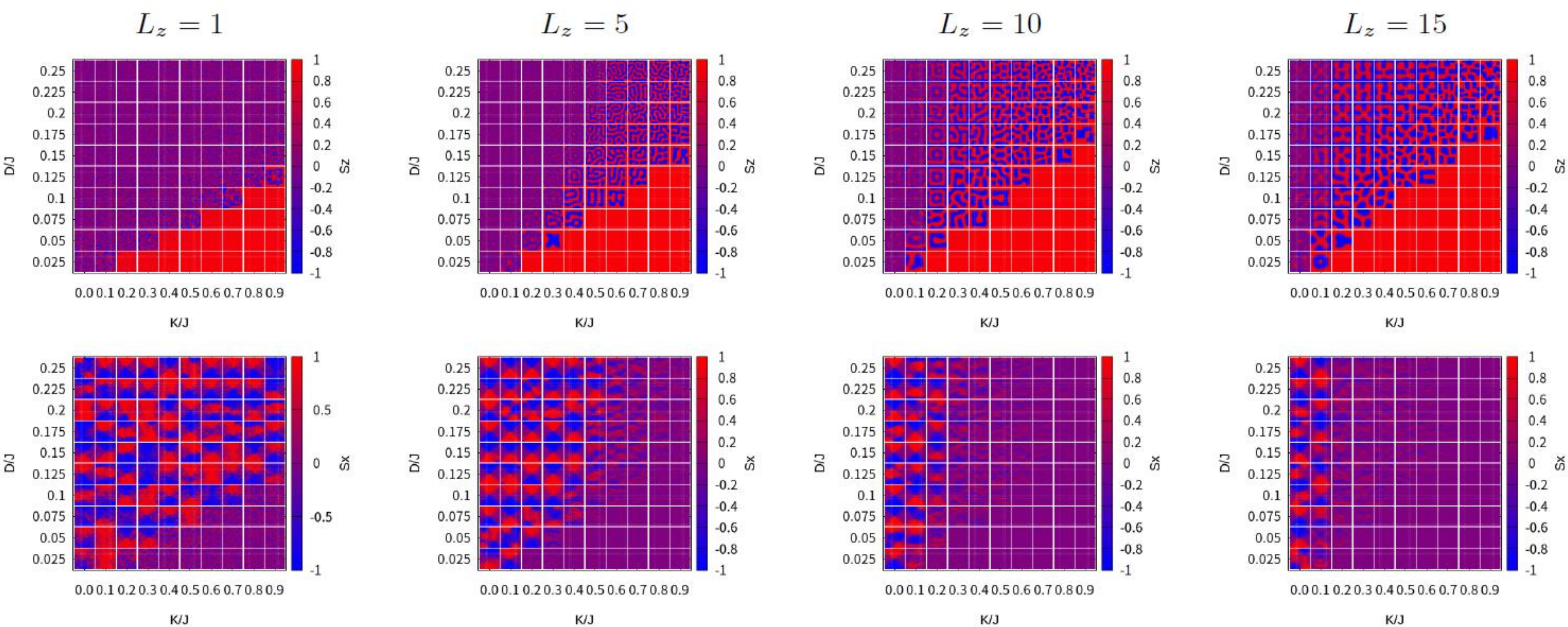
T. Nakamura, K. Toyoki, M. Suzuki

M. Suzuki *et al.*, Acta Mat.**106**, 155 (2016).

Metastability of ferromagnetic state in large grain?

Paramerter (K/J, D/J) depencence of magnetic patterns in thin films

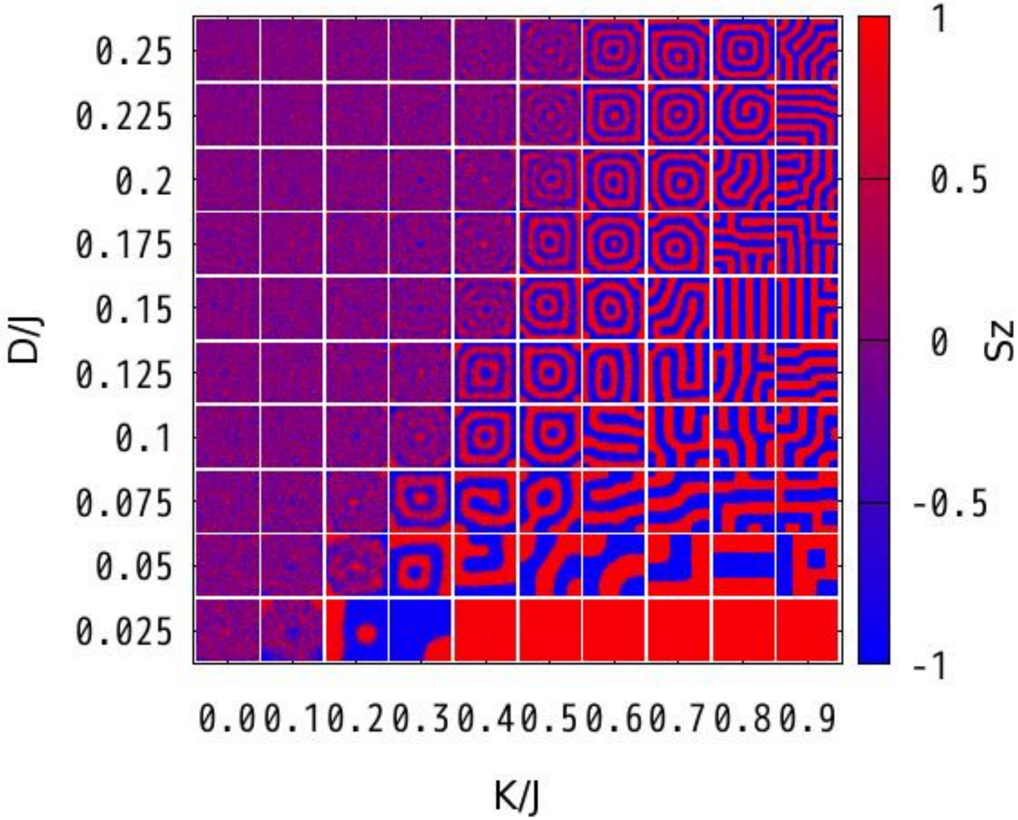
Multidomain structure (64x64;Lz): field-quench



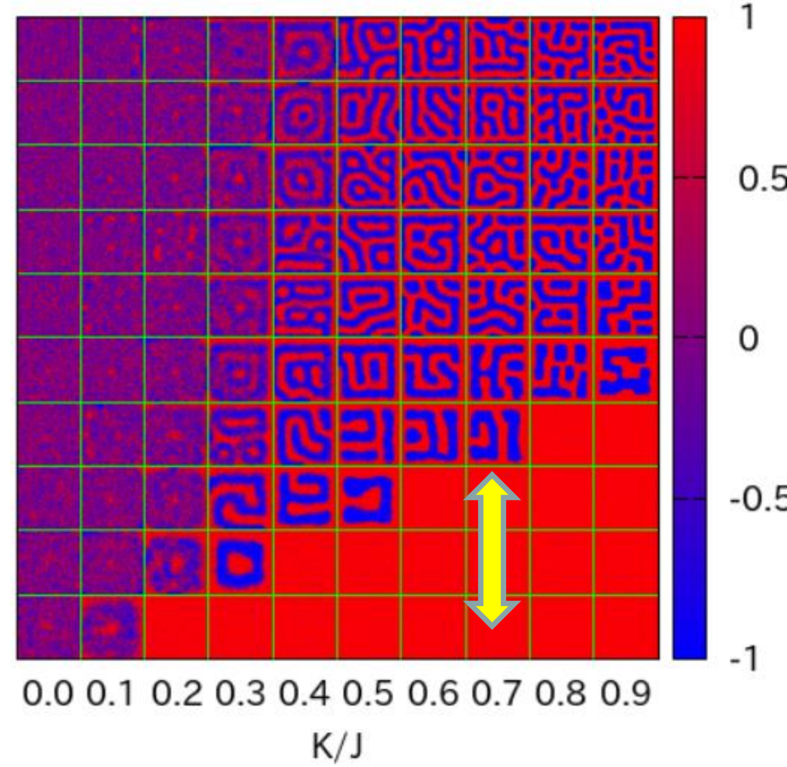
T. Hinokihara and S. Miyashita: arXive:2009.11574.

Hysteresis of large (multi-domain) grains

Random start



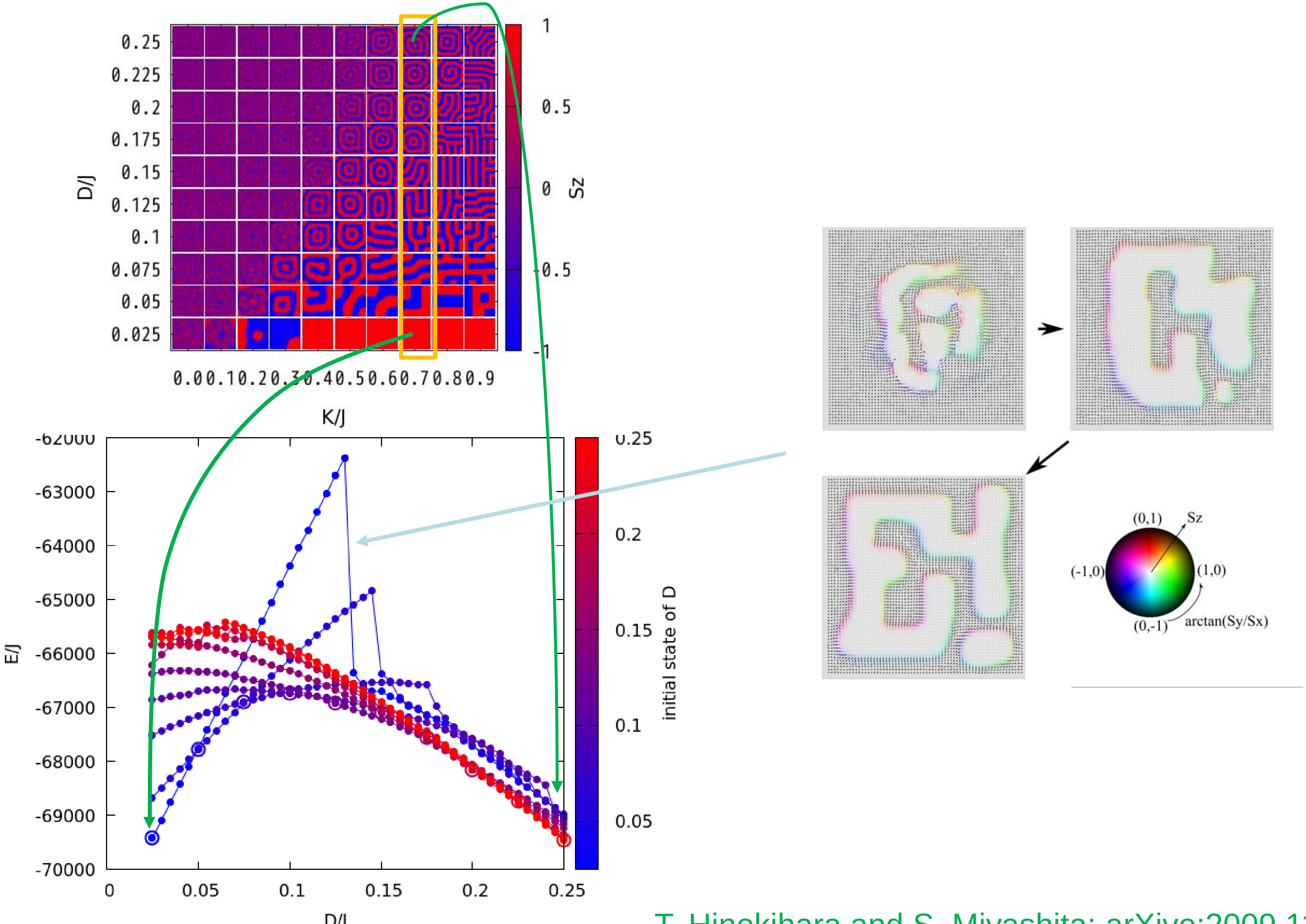
ferro start



D/J: strength of the dipole-dipole interaction (size effect)
K/J: strength of the anisotropy energy.

T. Hinokihara and S. Miyashita: arXive:2009.11574.

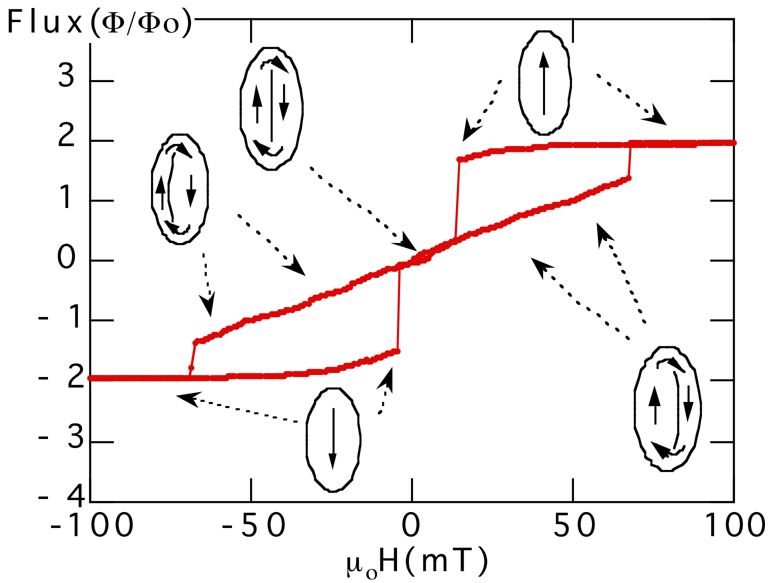
Collapse of metastability by DDI



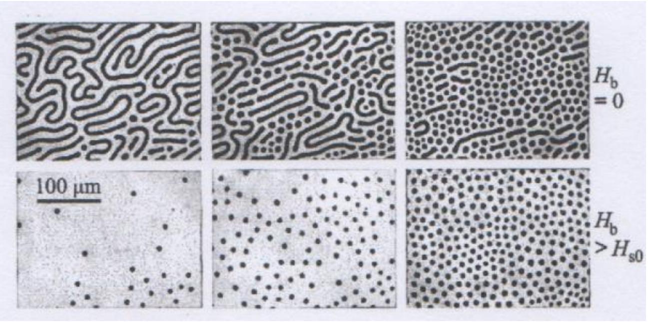
T. Hinokihara and S. Miyashita: arXive:2009.11574.

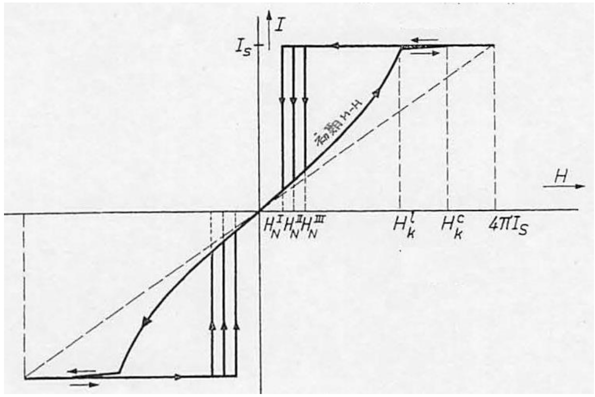
Hysteresis with multidomain

one single particle elliptic Co particle of 300 nm x 200 nm x 35 nm



Worfgang Wernsdorfer KIT Lecture,
PRB 53, 3341 (1996) and PRB 53, 6536 (1996).



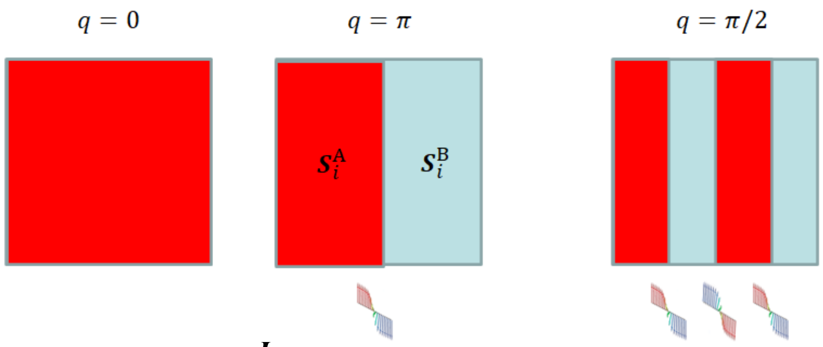


A. Hubert and R. Schaefer: Magnetic Domains (Springer (1998)

C. Kooy and U. Enz: Philips Res. Repts 15, 7 (1960)

Simplified mean-field analysis

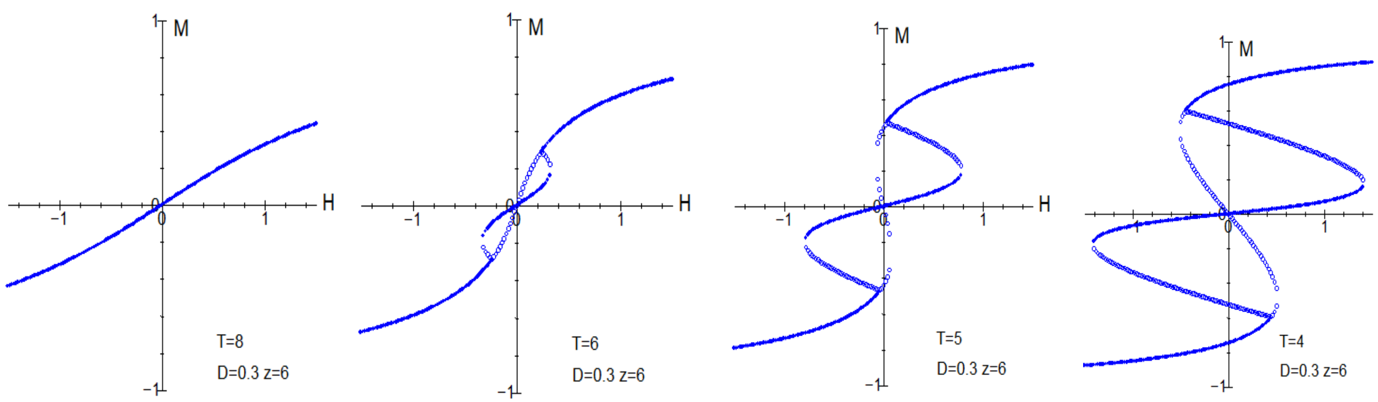
$$\mathcal{H} = -J\sum_{<ij>} \boldsymbol{S}_i^{\mathrm{A}}\cdot\boldsymbol{S}_j^{\mathrm{A}} - J\sum_{<ij>} \boldsymbol{S}_i^{\mathrm{B}}\cdot\boldsymbol{S}_j^{\mathrm{B}} + D\sum_{ij}\frac{\boldsymbol{S}_i^{\mathrm{A}}\cdot\boldsymbol{S}_j^{\mathrm{A}} - 3(\boldsymbol{S}_i\cdot\boldsymbol{r})(\boldsymbol{S}_j\cdot\boldsymbol{r})}{r^3} - \sum_i H\cdot\boldsymbol{S}_i$$

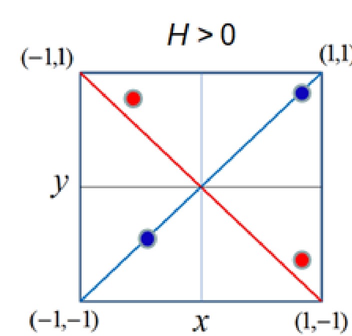


$$x = \langle S_i^{z\mathrm{A}}\rangle$$
$$y = \langle S_i^{z\mathrm{B}}\rangle$$
$$M = \langle S_i^{z\mathrm{A}}\rangle + \langle S_i^{z\mathrm{B}}\rangle$$

$$F(x,y) = \frac{Jz}{2}x^2 - k_{\mathrm{B}}T\ln[2\cosh^2(\beta Jzx - \beta Dy + \beta H)] + \frac{Jz}{2}y^2 - k_{\mathrm{B}}T\ln[2\cosh^2(\beta Jzy - \beta Dx + \beta H)]$$

M. Nishino and S. Miyashita, Phys. Rev. B **88**, 014108 (2013).

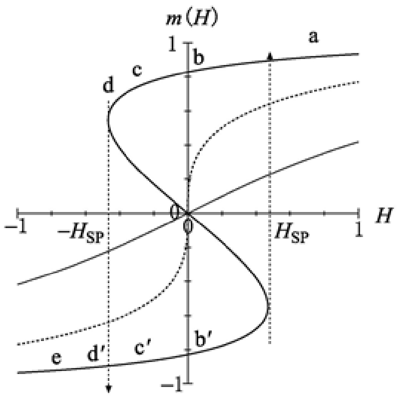




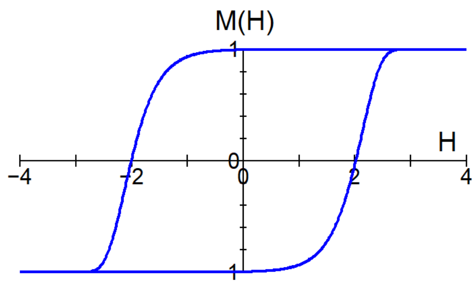
いろいろなヒステリシス

保磁力: Variety of hysteresis loops

Mean-field hysteresis

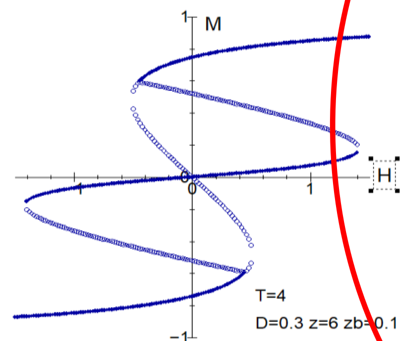


Neel-Arrhenius hysteresis

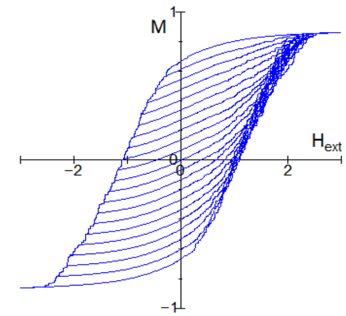


v=1, beta=3 E_0=H_0=1, a=0.1d0

Multi-domain

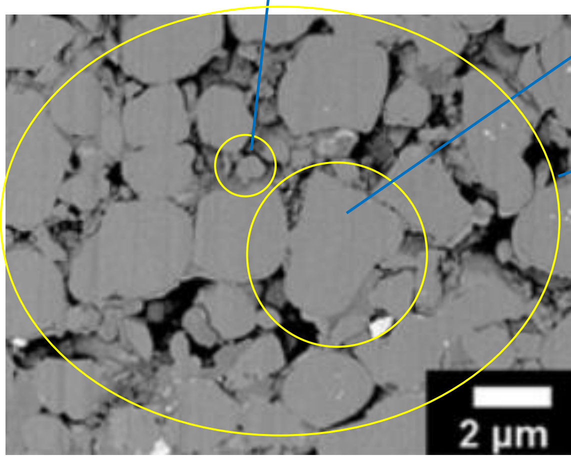


FORC
(first-order reversal curve)



D=0.2, J=0.1, Dtheta=0.1, Dm=0.0, DHc=0.13
+ some reversible contributions from soft magnets

metastability Thermal activation Dipole-dipole Int. Ensemble

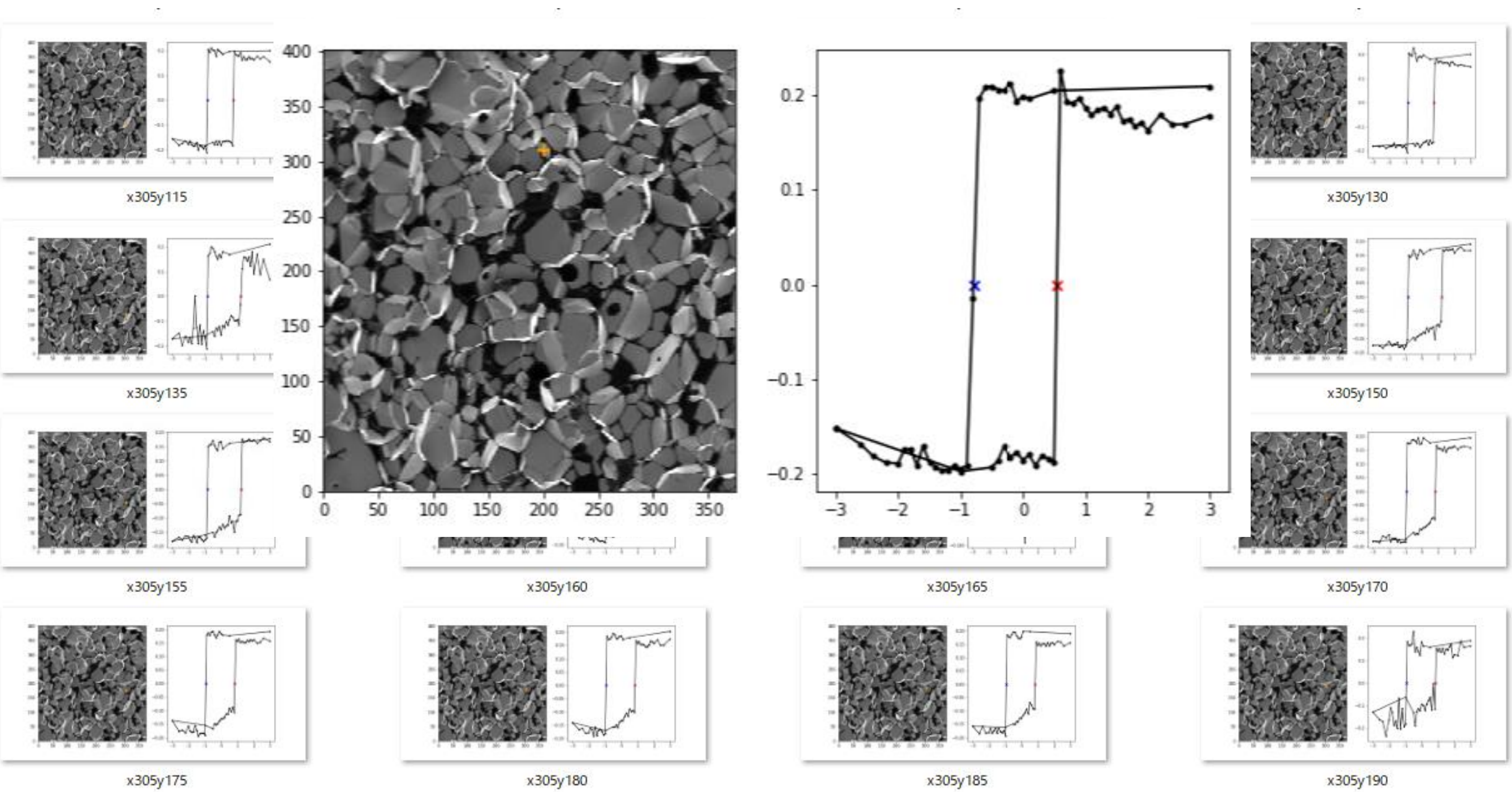


Microscopic approach

$$\langle A(T)\rangle = \frac{\mathrm{Tr}\ A e^{-\beta\mathcal{H}}}{\mathrm{Tr}\ e^{-\beta\mathcal{H}}}$$

a microscopic Hamiltonian $\mathcal{H}$

Distribution of hysteresis loop (interacting Preisach model)

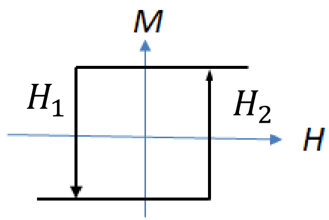


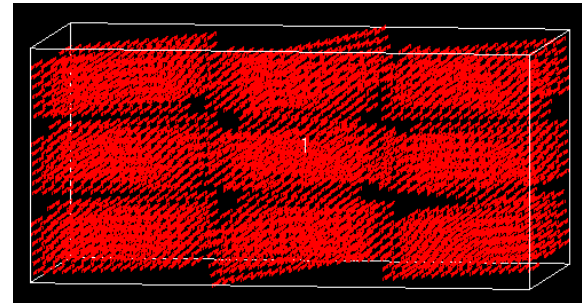
K. Toyoki, private communication

Preisach model and FORC analysis

Hysteresis of each grain

hysteron



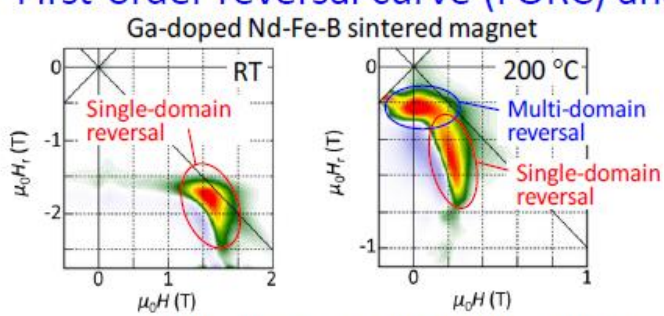


Distribution P(Ui, Vi) and FORC(First-order reversal curve)

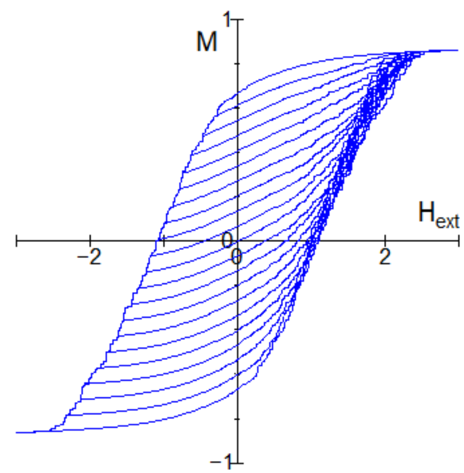
$$P(H_1, H_2) = M(H_1+\epsilon_1, H_2+\epsilon_2) + M(H_1, H_2) \\ -M(H_1+\epsilon_1, H_2) - M(H_1, H_2+\epsilon_2)$$

$$= \frac{\partial^2 M(H_1, H_2)}{\partial H_1 \partial H_2}$$

- First-order reversal curve (FORC) analysis



[Miyazawa and SO, ACTA Mater. (2019)]
[SO, ACTA Mater. (2019)]



D=0.2, J=0.1, Dtheta=0.1, Dm=0.0, DHc=0.13
+ some reversible contributions from soft magnets

FORC analysis for archaeology

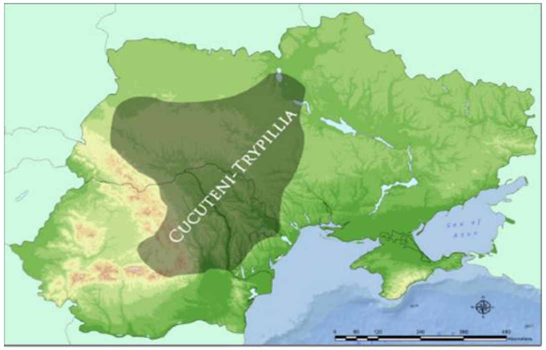
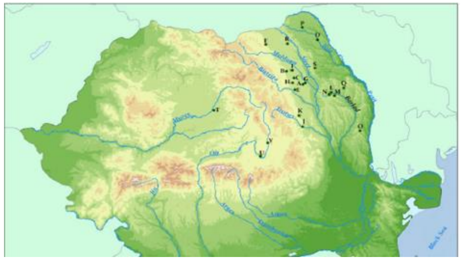


Fig. 1. The location of the Cucuteni–Trypillia civilization in Romania, Ukraine and the Republic of Moldova.



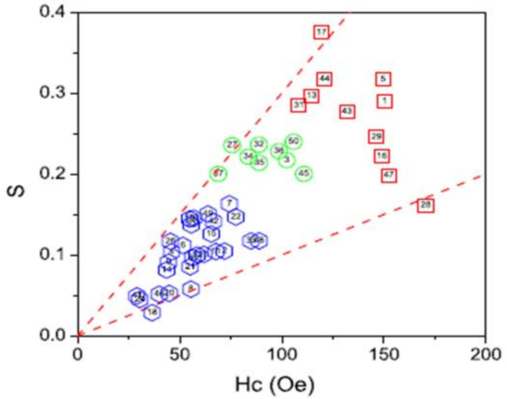
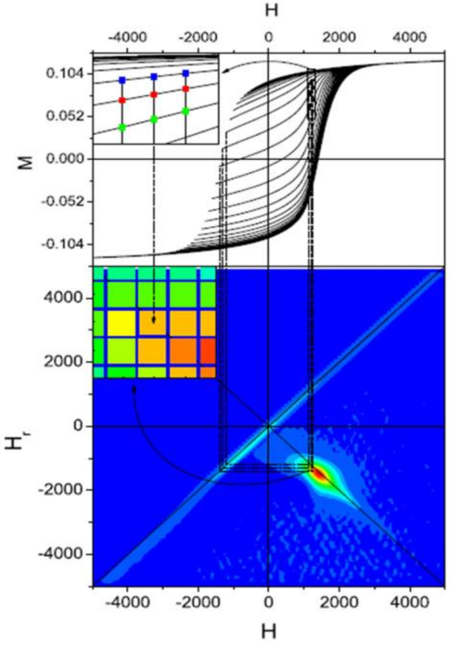
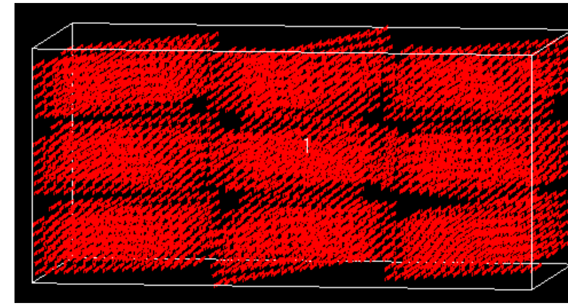
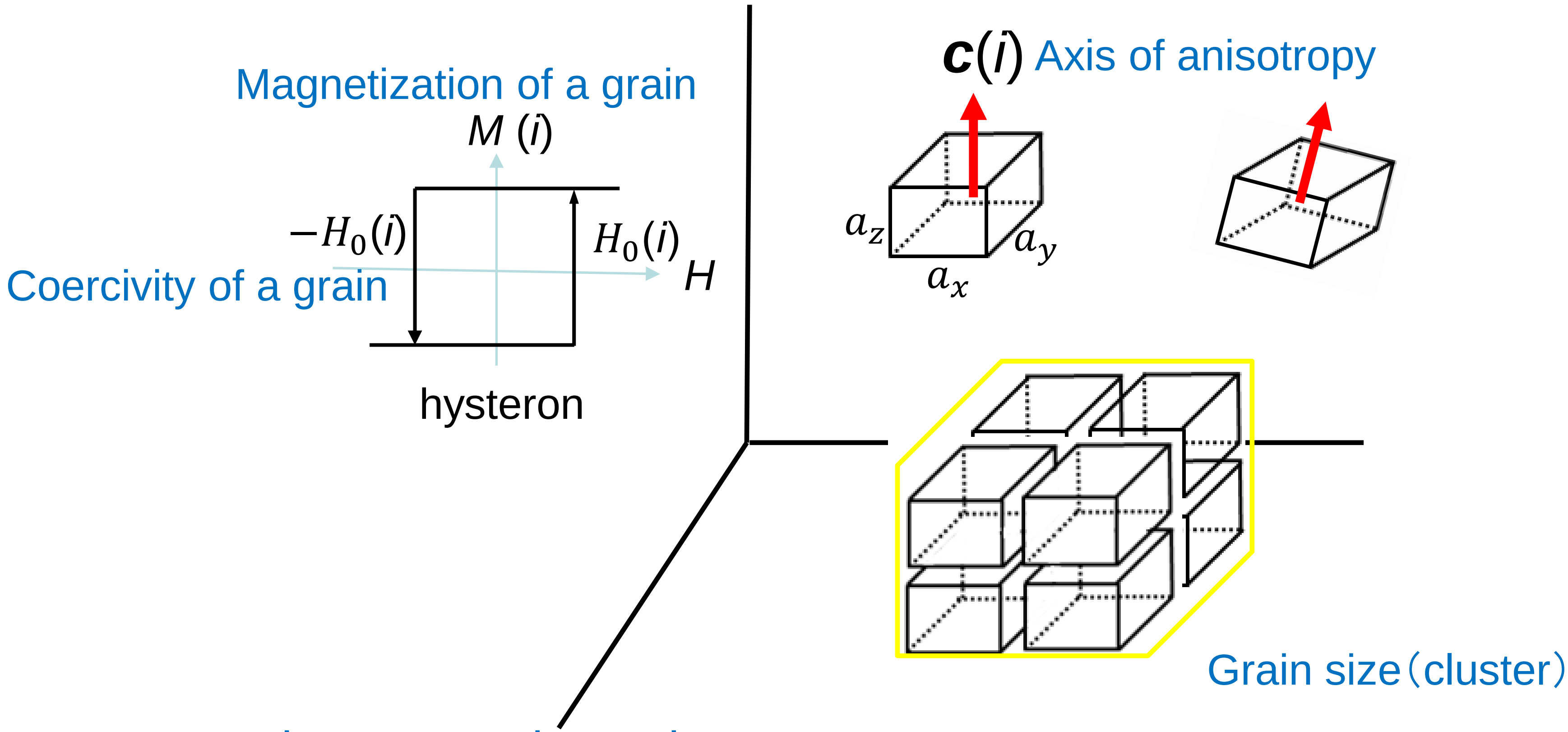


Fig. 8. The squareness (*S* is the ratio between the remanent and the saturation magnetic moment measured on the major hysteresis loop) *versus* coercive field diagram for the Cucuteni potshards.



Interacting Preisach model





Interaction among the grains

Dipole-dipole interaction (moving model)

e.g., C. R. Pike, et.al. PRB **71**, 134407 (2005)

Interaction with the grain boundary

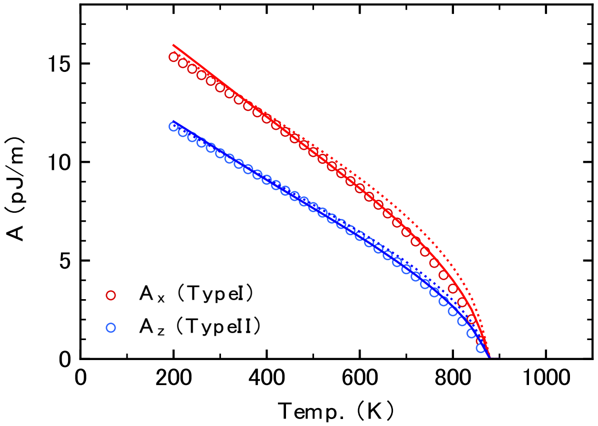
Trial to a renormalized Hamiltonian for finite temperatures to study larger system.

Continuum model: Temperature dependence of stiffness constant.

$$E_{\mathrm{cont}} = \int d\boldsymbol{r}\, \boxed{A_{\mathrm{ex}}(T)} [\nabla \boldsymbol{M}(T)]^2 + \int d\boldsymbol{r}\, [\boxed{K_1(T)} \sin^2\theta + \boxed{K_2(T)} \sin^4\theta + \boxed{K_4(T)} \sin^6\theta]$$

$A_{\mathrm{ex}}(T)$: Exchange Stiffness const.

$K_u(T)$: Magnetic Anisotropy const.

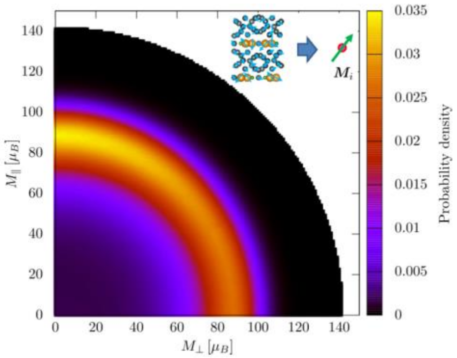


Y. Toga, M. Nishino, S. Miyashita, T. Miyake, and A. Sakuma, Phys. Rev. B98, 054418 (2018).

Renormalized Hamiltonian with a variable on-site magnet

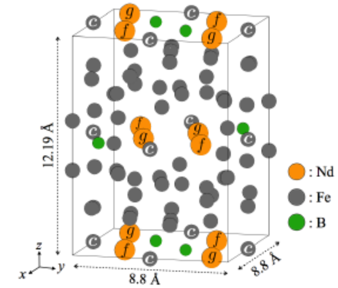
$$P(M) \propto e^{-\beta E_{\mathrm{self}}(\boldsymbol{M})} = \int_{\mathrm{microscopicconfigurations}} e^{-\beta \mathcal{H}} \times \delta\left(\boldsymbol{M} - \sum_i \boldsymbol{S}_i^2\right)$$

$$\mathcal{H}_{\boldsymbol{M}} = \sum_{ij} J_{ij} \boldsymbol{M}_i \cdot \boldsymbol{M}_j + \sum_i E_{\mathrm{self}}(\boldsymbol{M}_i)$$



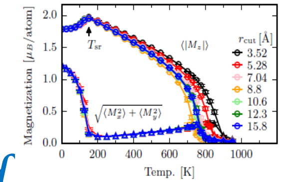
Choice of interaction form is difficult

$Nd_2Fe_{14}B$ magnet at finite temperatures

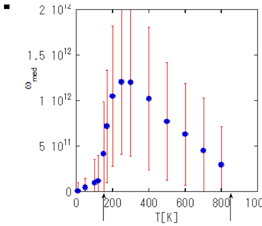


Thermodynamic properties $$\langle A(T)\rangle = \frac{\mathrm{Tr}\, A e^{-\beta\mathcal{H}}}{\mathrm{Tr}\, e^{-\beta\mathcal{H}}}$$

Response function $$I^{\alpha}(\tau) = \frac{1}{T}\int_{t0}^{t0+T} dt \left\langle \bar{S}^{\alpha}(t)\bar{S}^{\alpha}(t+\tau)\right\rangle$$



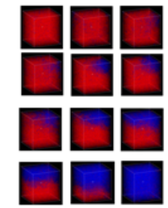
All properties can be obtained by a microscopic Hamiltonian $\mathcal{H}$.

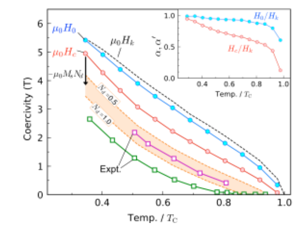


Coercivity : relaxation from a metastable state

Single grain : Arrhenius law:

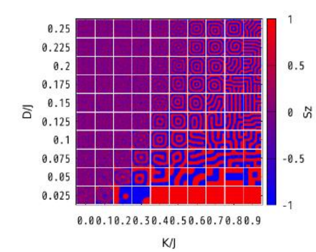
LLG, Minimum energy path, Free-energy method





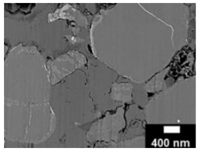
Dipole-dipole interaction for large size:

Stochastic cutoff method, Time quantified MC method

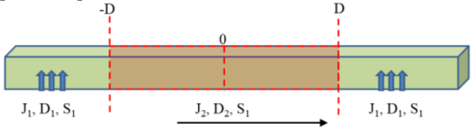


Ensemble of grains :

Preisach model, FORC diagram

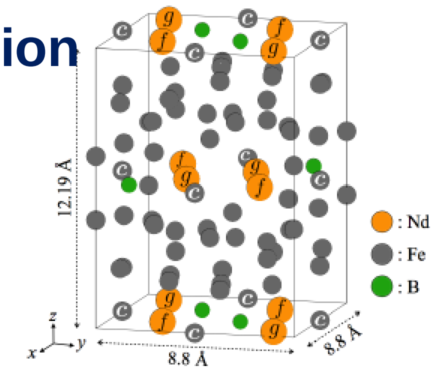


Effects of grain boundary phase and surface properties



論文リスト:

Finite temperature property, Atomic specification



新規開発アルゴリズム

LLG: finite temperature (西野)、

MC: Constrained, Free energy landscape, (栂)

MC: dipole-dipole interaction, SCO, TQMC (檜原)

論文

[1] M. Nishino and S. Miyashita, Phys. Rev. B91, 134411(1-13) (2015).

[2] S. Mohakud, S. Andraus, M. Nishino, A. Sakuma, S. M., Phys. Rev. B 94, 054430 (2016).

[3] Y. Toga, M. Matsumoto, S. Miyashita, H. Akai, S. Doi, T. Miyake, and A. Sakuma: Phys. Rev. B 94, 174433 (2016). & Phys. Rev. B 94, 219901 (2016).

[4] M. Nishino, Y. Toga, S. M., H. Akai, A. Sakuma, and S. Hirosawa, Phys. Rev. B 95, 094429 (2017).

[8] Y. Toga, M. Nishino, S. Miyashita, T. Miyake, and A. Sakuma, Phys. Rev. B98, 054418 (2018) .

[5] T. Hinokihara, M. Nishino, Y. Toga, and S. Miyashita, Phys. Rev. B97, 104427 (2018).

[6] S. Hirosawa, M. Nishino, S. Miyashita, Adv. Nat. Sci.: Nanosci. Nanotechnol. 8 (2017) 013002.

[7] S. Miyashita, M. Nishino, Y. Toga, T. Hinokihara, T. Miyake, S. Hirosawa and A. Sakuma, Scripta Materialia, 154,259 (2018).

[8] M. Nishino, S. Miyashita: Phys. Rev. B100, 020403(R) (2019).

[9] I. E. Uysal, M. Nishino, S. Miyashita, Magnetic fields threshold for nucleation and depinning in of the neodymium permanent magnet Nd2Fe14B, Phys. Rev. B101,094421 (2020).

[10]Y. Toga, S. Miyashita, A. Sakuma and T. Miyake, Temperature dependence of the coercive force – activation energy for magnetization reversal –, Nature Partner Journal Computational Materials 6, 67 (2020).

[11]M. Nishino, I. E. Uysal , T. Hinokihara S. Miyashita, Dynamical aspects of magnetization reversal in the Neodymium permanent magnet by a stochastic LLG simulation at a finite temperature -- real time dynamics and a quantitative estimation of coercive force --, B102,020413(R) (2020)

[12]T. Hinokihara and S. Miyashita: Multidomain structure and coercive force, Phys. Rev. B(2021) to be published.

T. Hinokihara, Y. Okuyama, M. Sasaki, and S. Miyashita: Time Quantized Monte Carlo Method for Long-range Interacting Systems 投稿中

M. Nishino, I. E. Uysal , S. Miyashita: Effect of the magnetic anisotropy of the surface neodymium atoms on the coercivity 投稿中

S. Miyashita, et al, Atomistic theory of thermally-activated magnetization processes in rare earth permanent magnets投稿中

Thank you very much