

Lecture of Spin current

若手向けスクール

スピントロニクスと
スピン流SPINTRONICS & SPIN CURRENT
September 24th & 25th, 2015

Organized by:

Nano Spin Conversion Science, ERATO Spin Quantum Rectification (ERATO-SQR),
ICC-IMR (International Collaboration Center/Institute for Materials Research)

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実証スピン量子整流プロジェクト

新学術領域研究
「ナノスピン変換科学」 A4 Leaflet (PDF)

Introduction to Magnetism

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意欲ある皆さんの参加を歓迎いたします。

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Microscopic Origin of Magnetism

**WHAT OCCURS WHEN YOU DIVIDE
A MAGNET TO PIECES?**

MICROSCOPIC ORIGIN OF MAGNETISM

What occurs when a magnet is chopped into pieces?

- Even when a magnet is divided, only small magnets appear with the same magnetic charge density on both ends.
- Magnetic charges always appear as pair of opposite polarity, and no magnetic monopole has been observed until now.

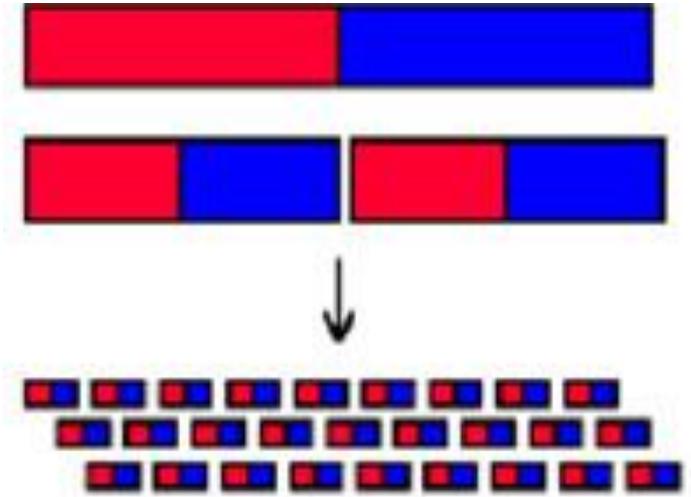


Fig.1 Even if a magnet is divided into pieces, areal density of magnetic charge at both ends of the small magnet remains unchanged.

What occurs when the magnet is cut into pieces of atomic scale?

- Finally even the atomic scale pieces show similar magnetic properties as the magnet.
- No magnetic poles exist on the atom.
- We start with a classical Bohr model of atom, in which an electron is circulating around the a nucleus.

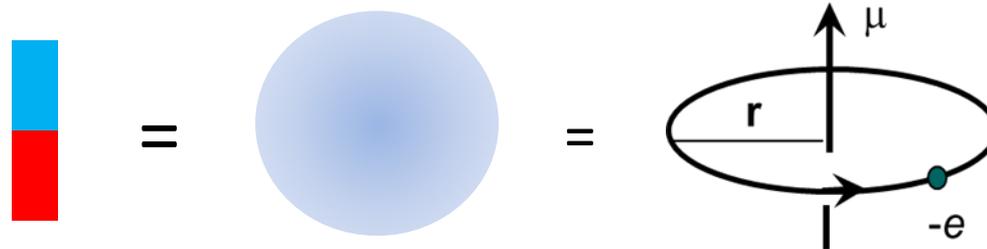


Fig.2 Classic Image of Atom

➤ Classic Picture of Atoms and Atomic Magnet

- According to the classic model of an atom, electrons circulate around a nucleus. Since current of $-ev$ is created when an electron with a charge $-e$ [C] moves with a velocity v , the circular current generates a magnetic moment.
- Equivalence of the magnetic moment generated by the circular current and the one produced by a pair of opposite magnetic charges can be verified by an equivalence of torque when both are placed in the static field.

➤ Torque created by a circular current

- An electron with a charge $-e$ [C] moving with a linear velocity of v [m/s] along a circumference of a circle with a radius r produces a current

$$i = -e/t = -ev/2\pi r \text{ [A]} \quad (1)$$

since time t to go around the circle is expressed by

$$t = 2\pi r/v \text{ [s]}$$

- If the circular current is placed in a homogeneous static magnetic field H [A/m], a force vector dF [N] working on a minute arc ds [m] can be described by the following formula ($[N]=[m \text{ kg/s}^2]$)

$$dF = ids \times \mu_0 H \quad (2)$$

- Since a torque dT working on an arc ds at a position r is expressed by $r \times dF$, a total torque T [Nm] can be obtained by integrating dT along the circumference as follows:

$$\begin{aligned} T &= \oint dT = (i/2) (\oint r \times ds) \times \mu_0 H \\ &= iS \times \mu_0 H \end{aligned} \quad (3)$$

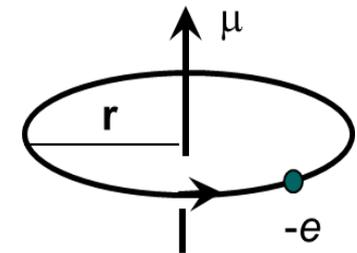


Fig.3 An electron going around a circle generates a magnetic moment

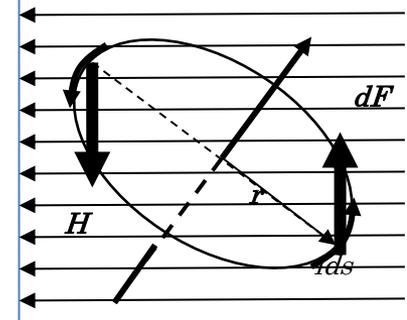


Fig.4 A force working on a circular current in a magnetic field.

➤ Torque working on a pair of magnetic charges placed in a magnetic field

- A torque T [Nm] working on a magnetic moment $\mu=Qr$ [Wbm] consisting of a pair of hypothetical magnetic charges $+Q$ [Wb] and $-Q$ [Wb] placed in a magnetic field H can be expressed by the following equation:

$$T=Qr \times H=\mu \times H \quad (4)$$

Since eq.(4) possesses the same vector product form $T=iS \times \mu_0 H$ in eq.(3), we can obtain an equation for magnetic moment μ [Wbm] by comparison of two equations as eq.(5).

$$\mu=\mu_0 iSn \quad (5)$$

It is found from eq.(5) that circulating current generates a magnetic moment, the value of which is proportional to the current and the area of the circle surrounding by the current, and the direction of which is along the normal of the area of the circle.

➤ Current → Angular momentum

- Using equations $i = -ev/2\pi r$ and $S = \pi r^2$ μ can be expressed as follows:

$$\mu = \mu_0 i S \mathbf{n} = \mu_0 (-ev/2\pi r) (\pi r^2) \mathbf{n} \quad (6)$$

- We replace r and v in eq.(6) by using an angular momentum $\Gamma = \mathbf{r} \times m\mathbf{v}$ to get an equation

$$\mu = \mu_0 (-1/2) e \mathbf{r} \times \mathbf{v} = (-\mu_0 e/2m) \Gamma \quad (7)$$

- In this way we finally express magnetic moment in terms of angular momentum.

➤ Magnetic moment in quantum mechanical expression

- To express a motion of an electron in an atom, it is necessary to treat physical quantity in terms of the quantum mechanics.
- In quantum mechanics angular momentum takes discrete values with a unit of h-bar, and expressed by an equation $\Gamma = \hbar l$, where l is the orbital angular momentum quantum.
- By substitution of Γ into eq.(7), an orbital magnetic moment is obtained as follows:
$$\mu = \mu_0(-e\hbar/2m)l = -\mu_B l \quad (8)$$
- Here $\mu_B = e\hbar/2m$ is a basic unit of magnetic moment called Bohr magneton, and the value is expressed in SI unit (E-H relation)
- $\mu_B = 1.16 \times 10^{-29}[\text{Wbm}] \quad (9)$

➤ Electronic orbital and quantum number

- Electronic states of electrons in an atom can be described using quantum numbers, n , l and $m=l_z$.
- Given the principal quantum n , orbital angular momentum quantum l takes discrete number between 0 and $n-1$ with increment of 1. For example $l=0$ for $n=1$ and $l=0$ or 1 for $n=2$.
- For an orbital angular momentum quantum l , quantization component (magnetic quantum number) $m=l_z$ takes total of $2l+1$ values, as follows:
 $l, l-1, \dots, -l+1, -1$

Table 1 Angular momentum quanta

n	l	m							orbital	Degeneracy
1	0				0				1s	2
2	0				0				2s	2
	1			1	0	-1			2p	6
3	0				0				3s	2
	1			1	0	-1			3p	6
	2		2	1	0	-1	-2		3d	10
4	0				0				4s	2
	1			1	0	-1			4p	6
	2		2	1	0	-1	-2		4d	10
	3	3	2	1	0	-1	-2	-3	4f	14

➤ Shape of electronic distribution corresponding to an orbital quantum number

- Orbitals s, p, d, f represent orbital shape, corresponding to orbital quantum number $l=0, 1, 2, 3$, respectively.
- Fig.5 gives a schematical illustration of spatial distribution of electrons of $1s, 2s, 2p_z, 3d_{xy}, 3d_z, 4f_z$ orbitals.
- As shown in the figure, s-orbital has no constriction, p-orbitals have one constriction, and d-orbitals have two constrictions. In this way orbital angular momentum quantum l represents number of constrictions of electron distribution.
- Magnetic moment obtained from the experiment cannot be explained solely by orbital angular momenta, since electron has not only orbital but also spin angular momentum.

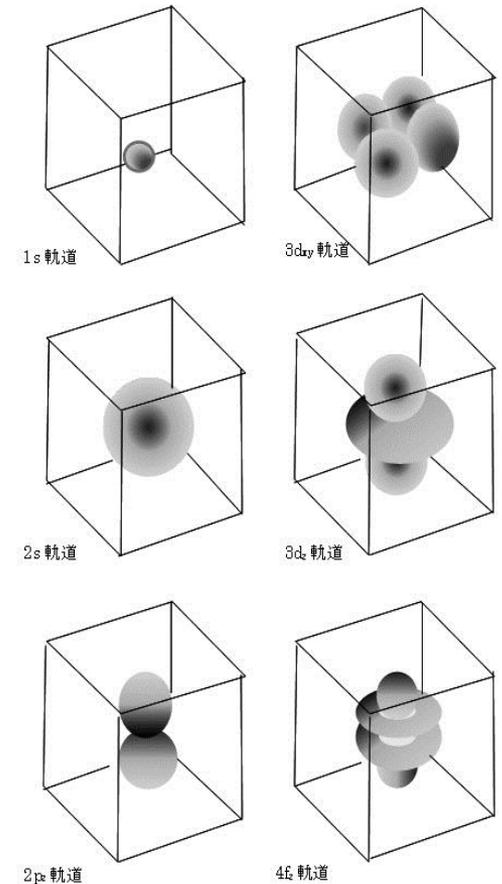


Fig.5 Electron distribution

➤ Spin angular momentum

- Electron possesses charge and spin. Since spin is derived theoretically from Dirac's relativistic wave equation, classical analogy is difficult.
- Spin is an internal degree of freedom of particle and has two eigenvalues corresponding to up-spin (\uparrow) and down spin (\downarrow), ie., right-circular and left circular rotation, respectively.
- Spin angular momentum quantum s takes *only two eigenvalues* $\frac{1}{2}$ and $-\frac{1}{2}$.

➤ Electron possesses spin angular momentum

- The concept that electron has a spin angular momentum was introduced to explain a **Zeeman effect** of D_1 luminescence of Na; i.e., splitting of the luminescence line (598.6nm : $3s_{1/2} \leftarrow 3p_{1/2}$) by a magnetic field.
- Existence of spin angular momentum is supported by **Stern-Gerlach experiment** in which particles (silver atoms in the original experiment) are sent through an inhomogeneous magnetic field to hit a screen, which shows discrete points rather than a continuous distribution, owing to the quantum nature of spin.

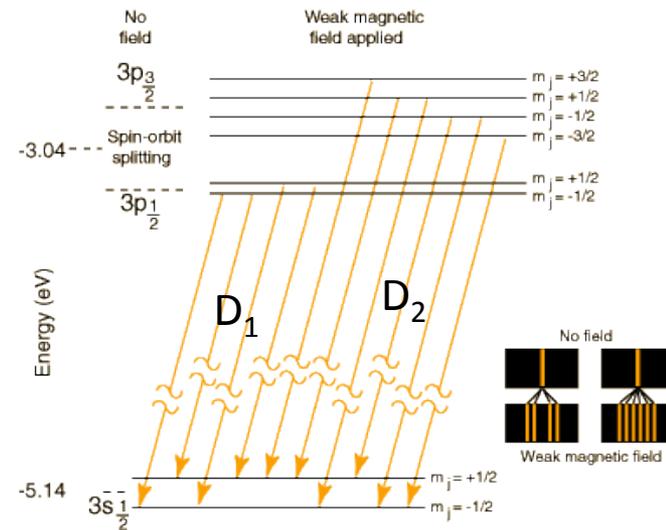


Fig. 6

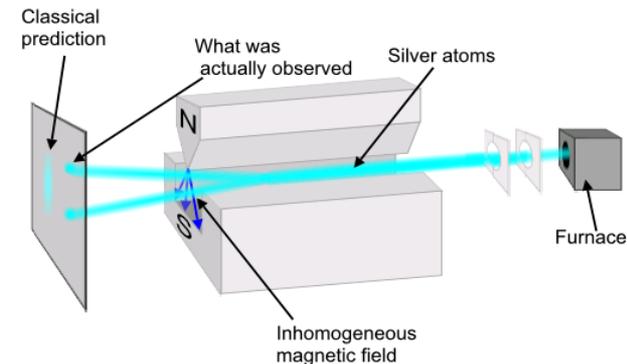


Fig. 7

➤ Composed angular momentum and magnetic moment of multi-electron atom

- Both the orbital angular momentum quantum l and the spin angular momentum quantum contribute to the magnetic moment of an atom.
- In the case of multi-electron atom, we calculate sum of orbital angular momentum quanta $L = \sum_i l_i$, as well as sum of spin angular momentum quanta $S = \sum_i s_i$ and finally we get total angular momentum by a vector addition of both quanta as $J = L + S$.

Composition of total angular momentum

- Relation between total orbital angular momentum and magnetic moment μ_l is expressed by,

$$\mu_L = -\mu_0(e\hbar/2m)L = -\mu_B L \quad (10)$$

On the other hand total spin angular momentum and magnetic moment has a relation expressed by

$$\mu_S = -(e/m)\hbar S = -2\mu_B S \quad (11)$$

Therefore, composed moment μ is described as

$$\mu = \mu_L + \mu_S = -\mu_B(L + 2S) \quad (12)$$

While $L + 2S$ is not reserved, $J = L + S$ is reserved during motion. L and S conserving the relation shown in Fig, 6 and goes round around the axis J .

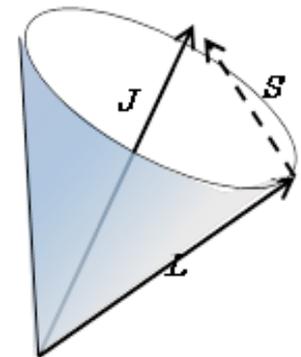


Fig. 8 L and S goes around J keeping vector relations

➤ Lande's g factor

- Magnetic moment μ is a vector parallel to \mathbf{J} with a magnitude which is a \mathbf{J} axis-projection (line OQ) of $\mathbf{L}+2\mathbf{S}$ (line OP) vector and expressed by eq.(13).

$$\mu = -g_J \mu_B \mathbf{J} \quad (13)$$

$$g_J J = |\text{OQ}| = |\text{OP}| \cos \alpha = |\mathbf{L} + 2\mathbf{S}| \cos \alpha = J + S \cos \beta$$

By using $\cos \beta = \mathbf{J} \cdot \mathbf{S} / JS$ and $2\mathbf{J} \cdot \mathbf{S} = J^2 + S^2 - L^2$ we obtain

$$g_J = 1 + (J^2 + S^2 - L^2) / 2J^2 \quad (14)$$

- In quantum mechanics, eigenvalues of L^2 , S^2 , J^2 are $L(L+1)$, $S(S+1)$, $J(J+1)$, respectively, since L, S, J are angular momentum operators.
- Then g_J of eq.(14) can be rewritten as

$$g_J = 1 + \{J(J+1) + S(S+1) - L(L+1)\} / 2J(J+1) \quad (15)$$

g_J is called Lande's g-factor.

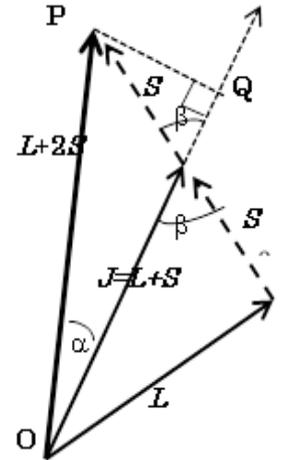


Fig. 9 Projection of OP ($\mathbf{L}+2\mathbf{S}$) on \mathbf{J} , OQ gives a magnitude of magnetic moment vector

Q: Why eigenvalue of L^2 is not L^2 but $L(L+1)$?

- In quantum mechanics any physical quantity corresponds to an operator. Since angular momentum operator L contains a differential operator as in $L=r \times p=r \times (-i\hbar \nabla)$, two operators A, B are not commutative, i.e., $[A,B]=AB-BA \neq 0$.

- Given Cartesian components of L is represented by L_x, L_y, L_z , lift operators L_+, L_- are defined by substitution as

$$L_+ = L_x + iL_y, \quad L_- = L_x - iL_y.$$

- Commutation relations are

$$[L_z, L_+] = L_+, \quad [L_z, L_-] = -L_-, \quad [L_+, L_-] = 2L_z \quad (\text{A1})$$

$$L^2 = L_x^2 + L_y^2 + L_z^2 = L_+ L_- + L_z^2 - L_z = L_- L_+ + L_z^2 + L_z \quad (\text{A2})$$

$$L^2 - L_z^2 - L_z = 0 \rightarrow L^2 = L_z^2 + L_z$$

$$L^2 \Phi_L = (L_z^2 + L_z) \Phi_L = L(L+1) \Phi_L \quad (\text{A3})$$

➤ Electronic configuration of multi-electron atoms

- If many electrons are belonging to an atom, contribution of orbital and spin of each electron to total momentum becomes complicated. A guideline to determine the total momentum was given by Hund and called “Hund’s Rule”.
- The premise of the Hund’s Rule is Pauli principle: i.e., Only one electron can occupy a state specified by a set of quantum numbers (n, l, m_l, m_s) .

➤ Hund's Rule

- Hund's Rule is consisting of following three rules
 1. For a given electron configuration, the term with maximum multiplicity has the lowest energy. The **multiplicity** is equal to $2S + 1$, where S is the total spin angular momentum for all electrons. Therefore, **the term with lowest energy is also the term with maximum S .**
 2. For a given multiplicity, the term with **the largest value of the total orbital angular momentum quantum number L , has the lowest energy.**
 3. For a given term, in an atom with outermost subshell **half-filled or less**, the level with the **lowest value** of the total angular momentum quantum number J , (for the operator $\mathbf{J}=\mathbf{L}+\mathbf{S}$) lies lowest in energy. If the outermost shell is **more than half-filled**, the level with the **highest value** of J , is lowest in energy.

➤ Expression of Multiplets

- In spectroscopy, multiplets are represented by symbols S, P, D, F, G, H, I corresponding to $L=0, 1, 2, 3, 4, 5, 6$ with spin multiplicity $2S+1$ on their left shoulders. Spin multiplicity values are 1, 2, 3, 4, 5, 6 which are called singlet, doublet, triplet, quartet, quintet, sextet corresponding to $S=0, 1/2, 1, 3/2, 2, 5/2$, respectively. And J is added as subscription.
- According to the definition, the ground state of hydrogen is described as ${}^2S_{1/2}$ (doublet S one half) and that of boron ${}^2P_{1/2}$ (doublet P one half) for example.
- In the case of 3d transition metals, only an electronic configuration (orbitals and spins) of incomplete inner shell electrons is sufficient. For example the multiplet of $Mn^{2+}(3d^5)$ with $S=5/2$ ($2S+1=6$), $L=0$ ($\rightarrow S$), and $J=5/2$ is expressed as ${}^6S_{5/2}$ (sextet S five half).

➤ Electronic configuration and magnetic moment in 3d-transition metal ions according to Hund's rule

- Fig. 10 shows how electrons occupy 3d-orbitals in 3d-transition metal ions according to Hund's rule.
- Each level corresponds to either of five $l_z = -2, -1, 0, 1, 2$. Though orbital energy of each level is degenerated, the five orbital levels are described separately for clarity.

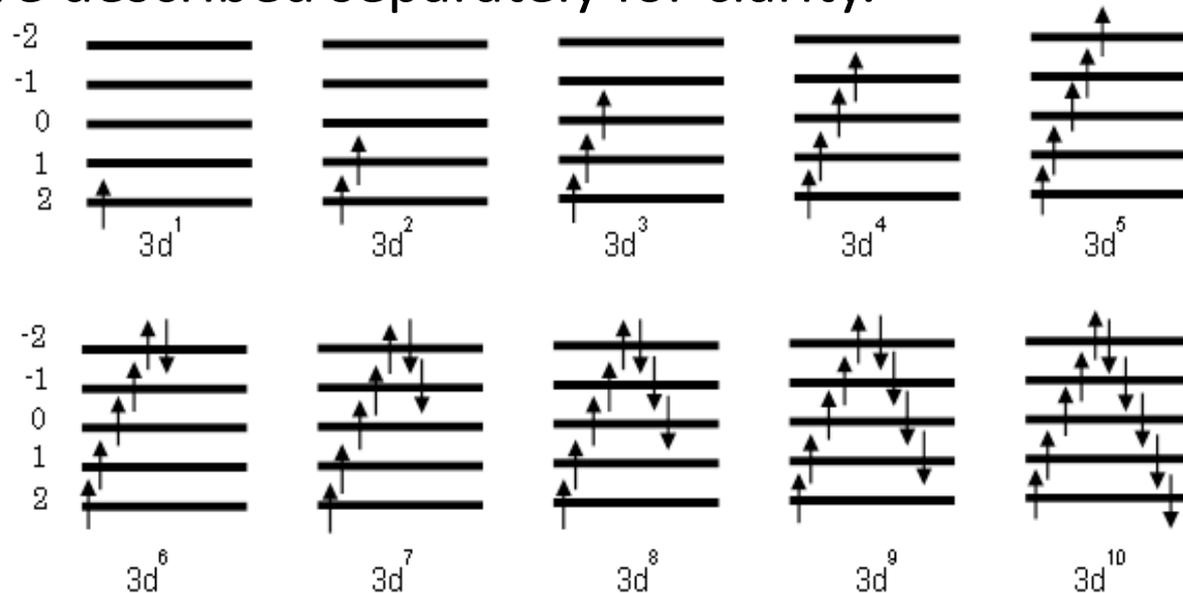


Fig.10 Electronic configuration of 3d transition ions following Hund's rules

Table 2. L, S, J, multiplet, magnetic moment in transition ions

ions	configuration	L	S	J	μ_J	μ_S	exp	multiplet
Ti ³⁺	[Ar]3d ¹	2	1/2	3/2	1.55	1.73	1.7	² D _{3/2}
V ³⁺	[Ar]3d ²	3	1	2	1.64	2.83	2.8	³ F ₂
Cr ³⁺	[Ar]3d ³	3	3/2	3/2	0.78	3.87	3.8	⁴ F _{3/2}
Mn ³⁺	[Ar]3d ⁴	2	2	0	0	4.90	4.8	⁵ D ₀
Fe ³⁺	[Ar]3d ⁵	0	5/2	5/2	5.92	5.92	5.9	⁶ S _{5/2}
Co ³⁺	[Ar]3d ⁶	2	2	4	6.71	4.90	5.5	⁵ D ₄
Ni ³⁺	[Ar]3d ⁷	3	3/2	9/2	6.63	3.87	5.2	⁴ F _{9/2}

Table 2 shows quantum numbers L, S, J for electronic configurations shown in Fig.10. Calculated values of magnetic moment for J and for S only, as well as experimental values are listed in the table.

➤ Contribution of orbital and spin angular momentum quantum number to magnetic moment

Magnetic susceptibility of paramagnets χ is inversely proportional to temperature T according to Curie law;

$$\chi = C/T \quad (16)$$

Here C is Curie constant and can be described using total angular momentum quantum number J,

$$C = N g_J^2 \mu_B^2 J(J + 1) / 3k. \quad (17)$$

In this equation N is number of ions, and k Boltzmann constants. If the susceptibility obeys the Curie law inverse of susceptibility is proportional to T. From the slope of the curve C is obtained and effective magnetic moment $\mu = g_J \sqrt{J(J + 1)}$ can be obtained.

➤ Paramagnetism of transition metals and rare earth ions

- As shown in Table 2, calculated and experimental values of magnetic moment of 3d-transition ions are listed.
- Fig.11(a) shows experimental values by open circles for TM. While calculated values for μ with J cannot explain the experiments while those with S fit the experiment.
- Fig.11(b) shows experimental value of magnetic moment of RE ions by open circles.
- The experimental magnetic moment of RE ions are all accounted for by calculated values for μ with J .

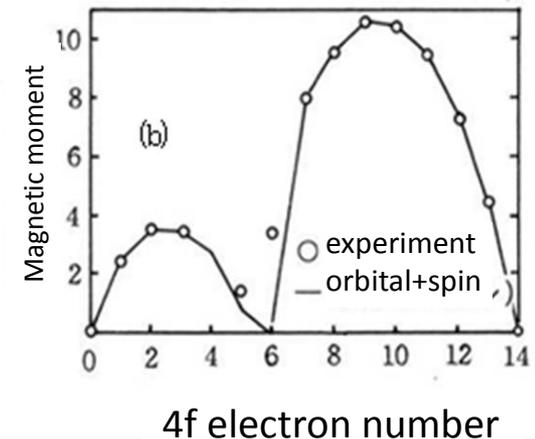
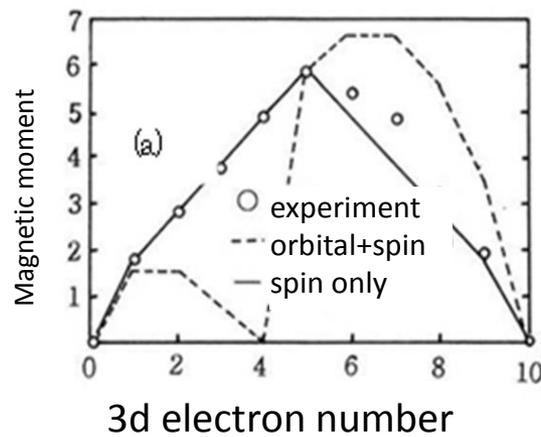


Fig.11 calculated and experimental values of paramagnetic effective moment for TM and RE ions

WHY IS IRON FERROMAGNETIC?

➤ Magnetic moment of Fe cannot be explained by magnetic moment of Fe atom

- Most people will remember the iron by a magnet. Nevertheless it has been a long mystery why magnetism of iron is so strong.
- As described in the last section magnetic moment is originated from orbital and spin angular momentum of electrons.
- Without force to align the atomic moment each other, direction of atomic moments align randomly, which does not lead to a spontaneous moment. Application of magnetic field aligns the moment slightly toward the field direction, inducing a net magnetic moment. This is a mechanism of paramagnetism.
- Temperature dependence of susceptibility of a paramagnet including 4f-RE ion is well explained by total magnetic moment, while that of 3d-TM ion can be explained considering only spin angular momentum.

➤ Exchange Interaction

- Materials will become ferromagnetic if the force acts mutually align the orientation of the magnetic moment in the same direction between adjacent atoms magnet.
- On the other hand, it will become antiferromagnetic if the force acts mutually align inversely.
- The force by which atomic magnets mutually align is brought by electrons and called “exchange interaction”.
- Ferromagnetic materials loose a spontaneous magnetization when temperature exceeds Curie temperature, which means that thermal fluctuations overcome exchange interaction.

➤ Magnetic moment per one Fe atom

- How much is the magnetic moment per a Fe atom, if magnetism of Fe is generated by alignment of atomic moment?

- Since Fe atom has an electronic configuration Ar closed shell $[1s^2 2s^2 2p^6 3s^2 3p^6] + 3d^6 4s^2$. Only outer shell electrons contribute magnetic moment.

- Since spin moment of the two 4s electrons cancel out, only spin moment of 3d TM contributes magnetization of Fe.

- Electron configuration is shown in Fig.12, from which $S = 4 \times 1/2 = 2$ is obtained, so that $\mu = 2S\mu_B = 4\mu_B$.

- However, **experimental Fe moment per atom is as small as $2.219\mu_B$** . Not only Fe, Co ($1.715\mu_B$) and Ni ($0.604\mu_B$) also show smaller moment than expected from calculation.

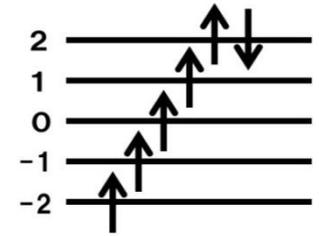


Fig.12 Electronic configuration of $3d^6$ according to Hund's rule.

➤ Itinerant Electron Model

- In metallic ferromagnet the magnetic moment per atom is reduced from the one expected from localized model and takes a non-integer value.
- This phenomenon can only be explained by an **itinerant electron model** or **band electron model** in which electrons are not confined in the atomic position but extend over many atoms in the metal to form the energy band structure.

➤ Energy band structure of non-magnetic metals

In metals conduction band is partially filled with electrons. The highest energy of filled band is called Fermi energy E_F .

(a) is DOS (density of states) of alkali metals.

(b) is DOS of 3d-transition metals without magnetic moment; In addition to the s-band, there appears a d-band with high DOS near the Fermi energy.

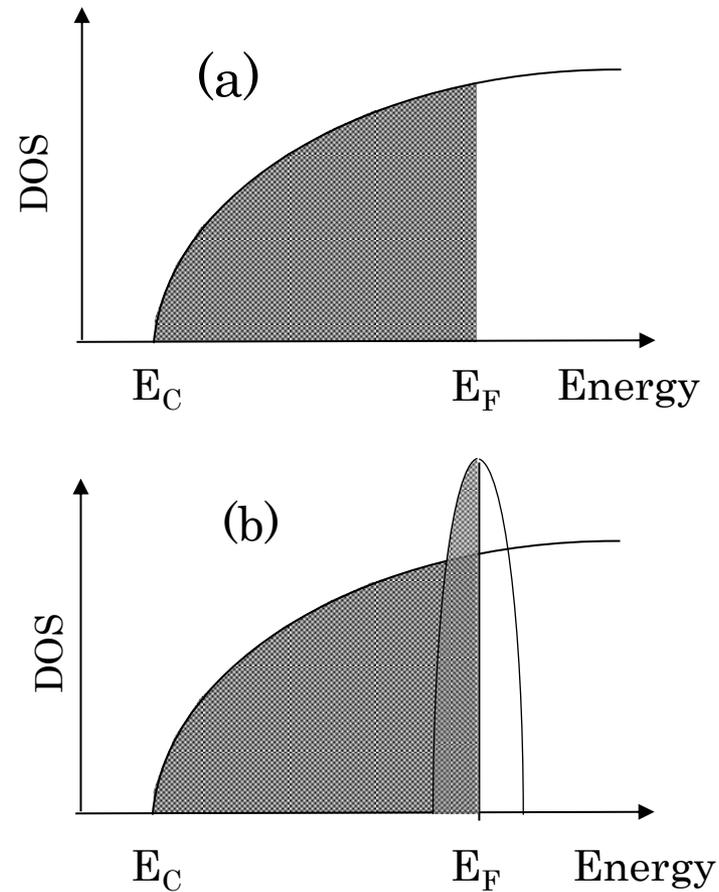


Fig.13 Density of states of metallic band
(a) Alkali metal, (b) 3d transition metal

➤ Band DOD of paramagnetic and ferromagnetic metals

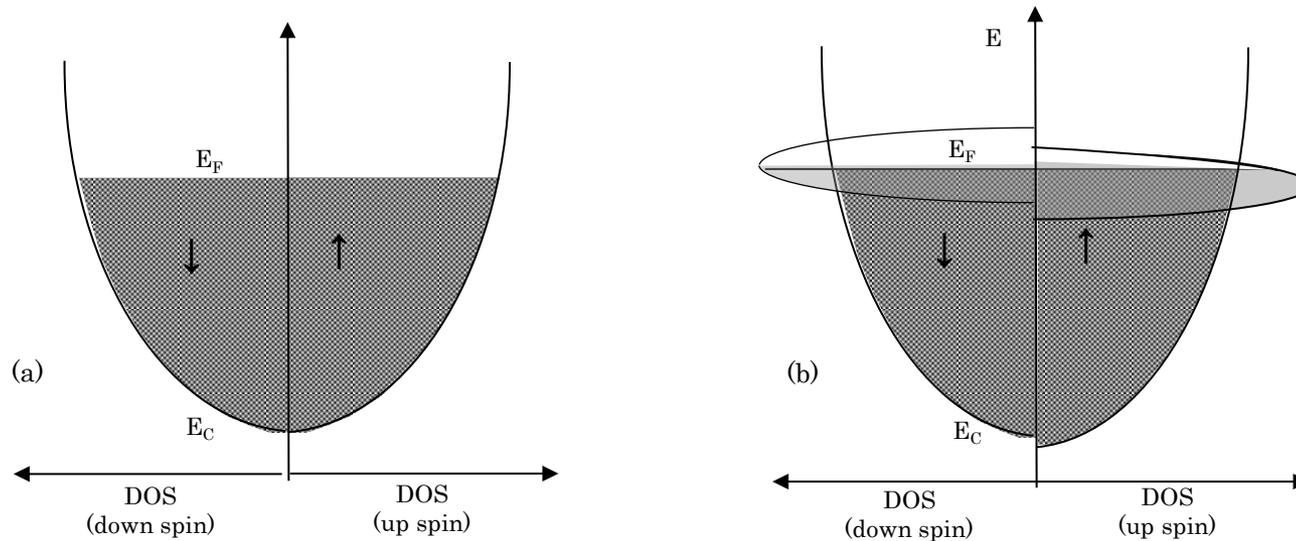


Fig.14

- Energy band of ferromagnetic metals is different for each spin. As shown in the DOS curves of the right half represents up spin and the left half represents down spin.
- (a) In paramagnetic metals DOS curves of up-spin band and down-spin band are symmetric.
- (b) In ferromagnetic metals up-spin band and down-spin band shows a shift in energy. Exchange splitting of the two bands is larger in 3d-band than in sp-band.

Magnetic moment of Fe can be explained in terms of electronic filling

Slater-Pauling Curve

- Magnetic moment per atom for various transition metal (TM) alloys plotted against number of electrons in the alloy.
- Plots are on the half lines with a 45° slope starting Cr or on the line with a -45° slope starting from Fe₃₀Co₇₀ to Ni₆₀Cu₄₀.

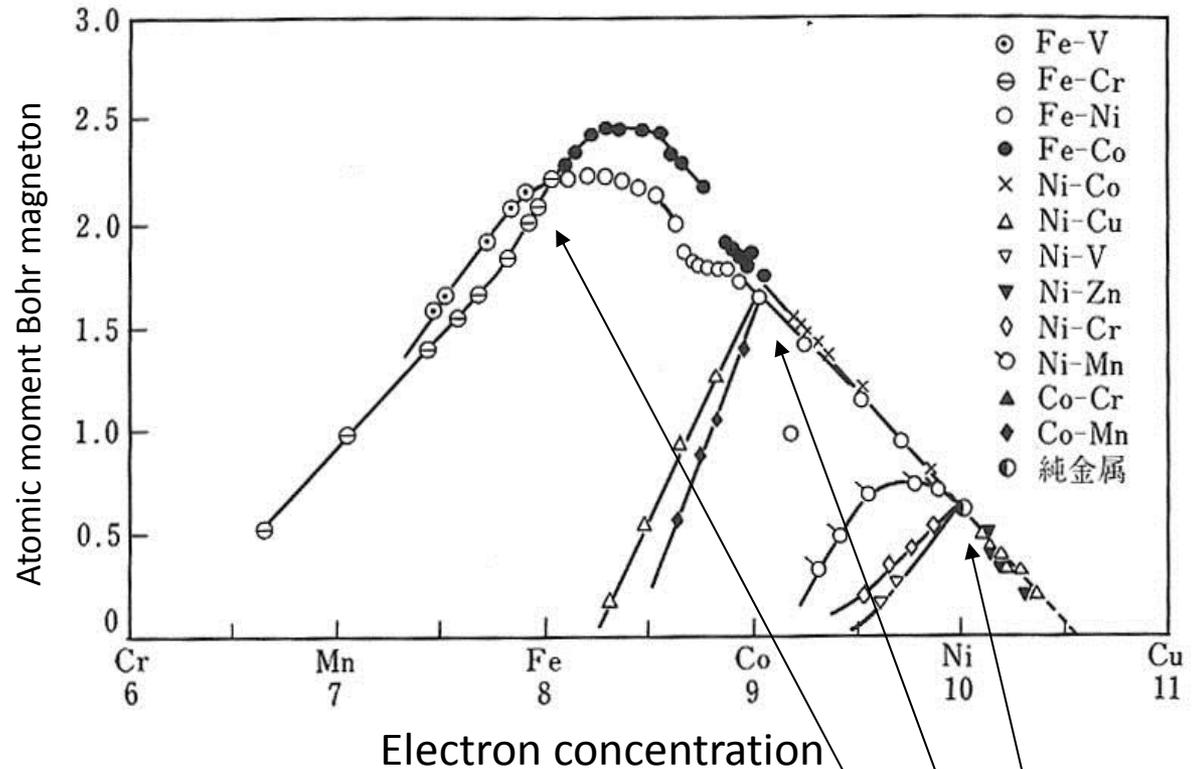
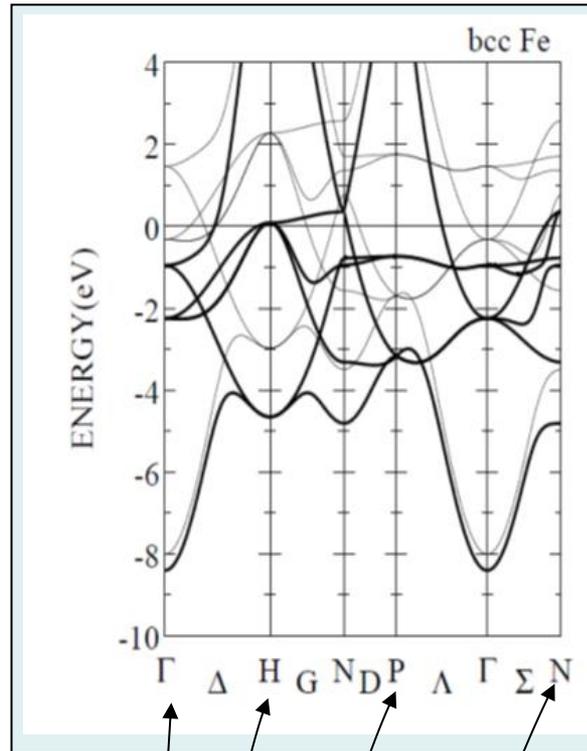
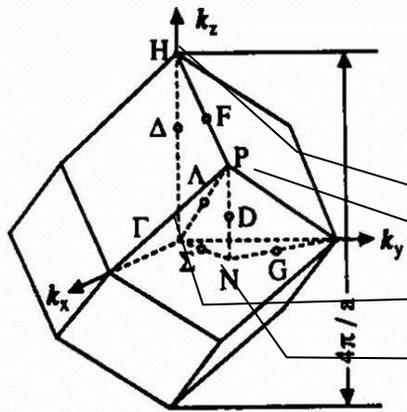


Fig.15 Slater Pauling curves of ferromagnetic alloys
Magnetic moment of Fe, Co and Ni is 2.2, 1.7 and 0.6 μ_B , which are smaller than atomic values. (Bozorth)

➤ Spin-polarized energy bands of Fe

Brillouin Zone of bcc lattice



Difference of occupied DOS between \uparrow spin and \downarrow spin bands gives a magnetic moment

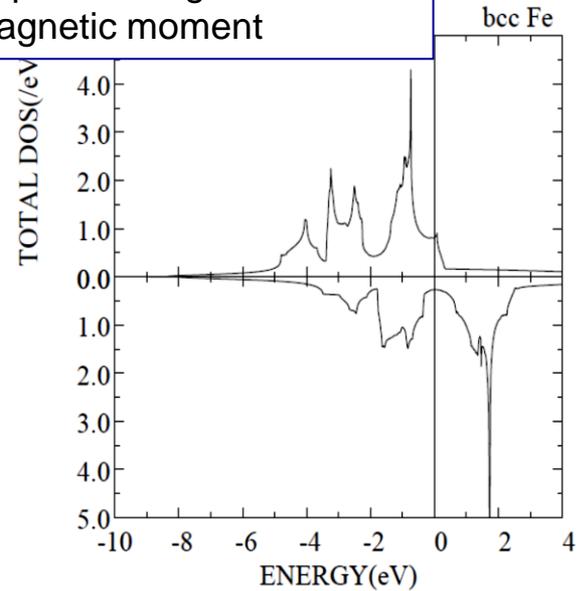
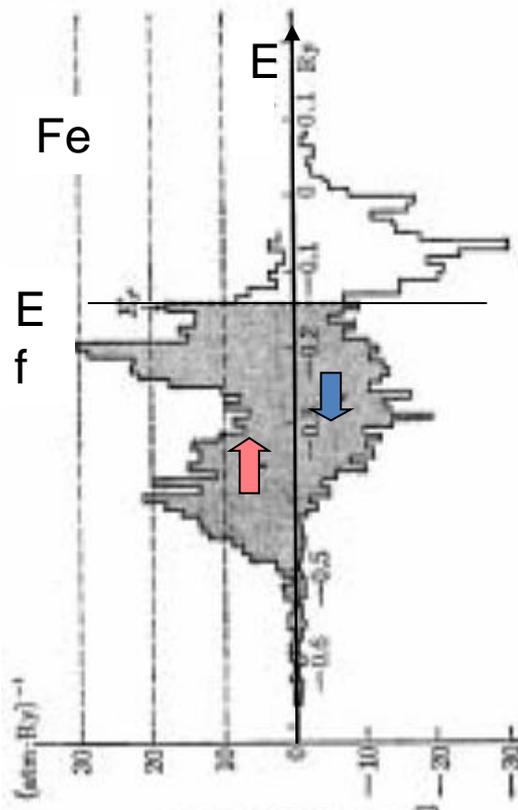


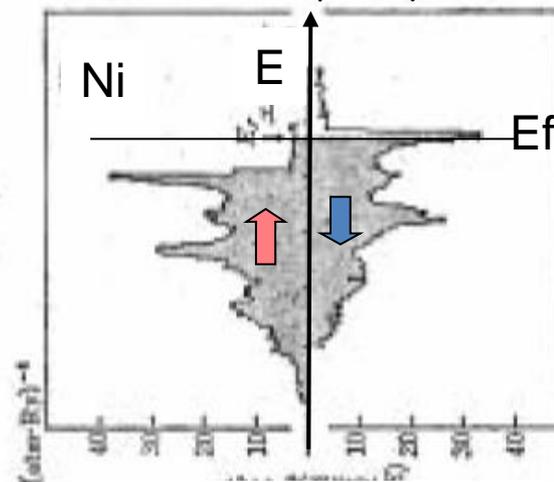
Fig. 16 Spin-polarized band and spin-polarized DOS of Fe

➤ DOS of Fe and Ni

- DOS of \uparrow band is smaller than \downarrow band in Fe giving $\Delta n = n_{\uparrow} - n_{\downarrow} = 2.2$
- \uparrow band is fully occupied and \downarrow band has only small amount of holes in Ni giving $\Delta n = n_{\uparrow} - n_{\downarrow} = 0.6$



Spin-polarized DOS



Spin-polarized DOS

- If \downarrow band has 0.6 holes, s-electrons flow from Cu into Ni, moment vanishes when Cu content becomes 40%.

Fig. 17 Spin-polarized DOS of Fe and Ni

➤ Mechanism of appearance of spontaneous magnetization: localized electron model

- In magnetic insulators such as iron oxides a spontaneous magnetization can be explained when atomic magnetic moments align mutually along the same direction.
- This mechanism is first introduced by Weiss in terms of **mean-field approximation** (or **molecular field approximation**).

Mean-field Approximation by Weiss

- We treat the exchange field as equivalent to a magnetic field H_E . The magnitude of the exchange field may be as high as $\mu_0 H_E = 10^3 [T]$.
- In the mean-field approximation we assume each magnetic atom experiences a field proportional to the magnetization:

$$H_E = AM \quad (18)$$

where A is expressed as $A = 2zJ_{ex}/(N(g\mu_B)^2)$ by quantum treatment.

- Consider the paramagnetic phase: an applied field H will cause a finite magnetization and this in turn will cause a finite exchange field H_E . Effective field is a sum of applied field H and exchange field H_E :

$$H_{eff} = H + H_E \quad (19)$$

- Below T_c we use the complete Brillouin expression for the magnetization:

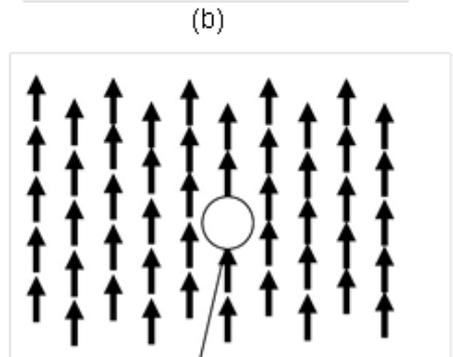
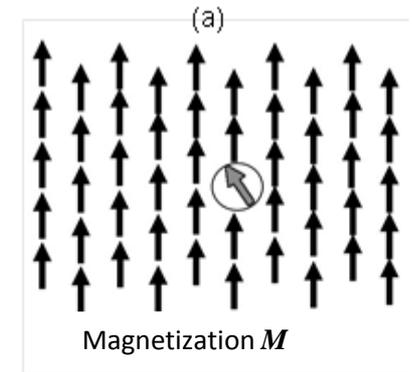
$$M = Ng\mu_B J B_J(g\mu_B H_{eff} / kT)$$

- Spontaneous magnetization appears without applied field: $H=0$. By substitution we get

$$M = Ng\mu_B J B_J(Ag\mu_B JM / kT) \quad (20)$$

- By assuming $M_0 = Ng\mu_B J$ eq.(20) becomes

$$M/M_0 = B_J((2zJ_{ex}J^2 / kT)M/M_0) \quad (21)$$



Mean field from surrounding moments

Fig. 18 Mean field approximation of Weiss

➤ Condition for existence of a spontaneous moment

By substitution $y=M/M_0$ and $x=(2zJ_{\text{ex}}J^2/kT)M/M_0$, eq.(21) becomes

$$y = (kT/2zJ_{\text{ex}}J^2)x \quad (22)$$

$$y = B_J(x). \quad (23)$$

Fig.18 illustrates eqs.(22) and (23). The curves in Fig.18 represent eq.(23) for $J=1/2, 3/2, 5/2$, while lines in Fig.18 represent eq.(22) whose slope is proportional to T .

The lines rise up more steep for higher temperatures than for low temperature.

Spontaneous magnetization appears when the curve (23) and the line (22) have an intersection.

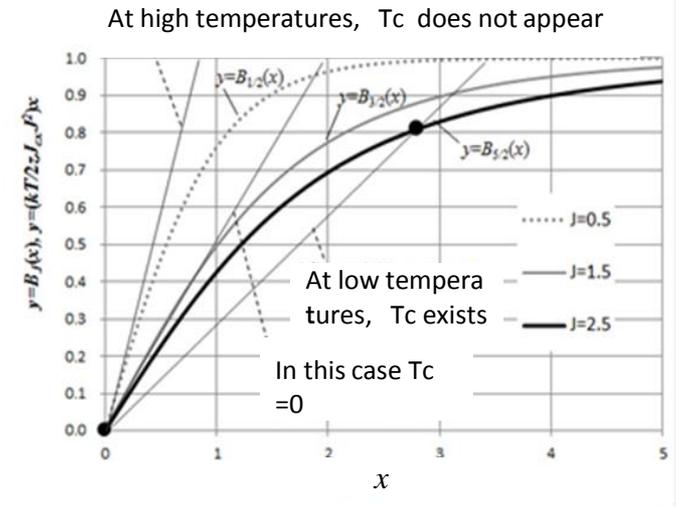


Fig. 19 Existence of a spontaneous moment in the mean-field theory

At low temperatures intersection exists since slope is small, while at high temperatures no intersection exists which leading to disappearance of spontaneous magnetization

Temperature-dependence of Magnetization

- In Fig.20 Magnetization obtained from intersections using Fig.19 for different temperature.
- Experimental M-T curves of Most ferromagnetic materials can be explained by the mean-field theory of Weiss, even they are itinerant magnets.

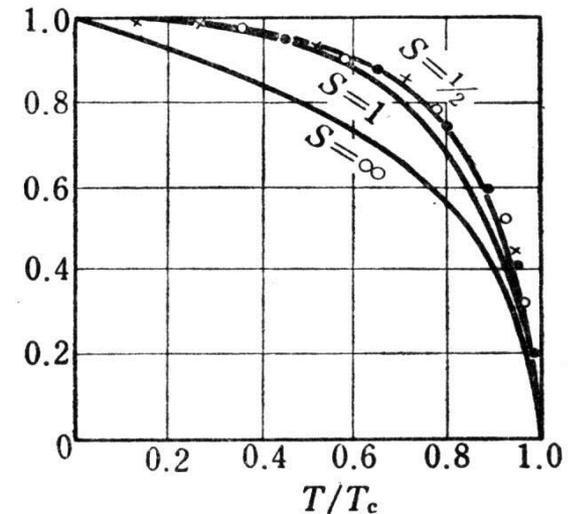


Fig.20 Temperature dependence of spontaneous magnetization
Experimental values are \times Fe, \bullet Ni, \circ Co.
Curves are plots for $J=S=1/2, 1, \infty$

➤ Curie Weiss Law

The susceptibility $\chi = M/H$ of a paramagnetic material, in which no interaction exists between magnetic moments can be expressed by Curie's $\chi = C/T$. (24)

Therefore if the line of $1/\chi$ is plotted against T intersects at the origin the material is paramagnetic. Above the Curie temperature of a ferromagnetic material the magnetic moments fluctuate randomly leading to paramagnetism.

In this case susceptibility is given by the Curie-Weiss law as follows

$$\chi = C/(T - \theta_p). \quad (26)$$

Here θ_p is called "paramagnetic Curie temperature". In this case $1/\chi$ plots against T passes through Y-axis at θ_p . If the value is positive the material is ferromagnetic, while if it is negative the material is antiferromagnetic.

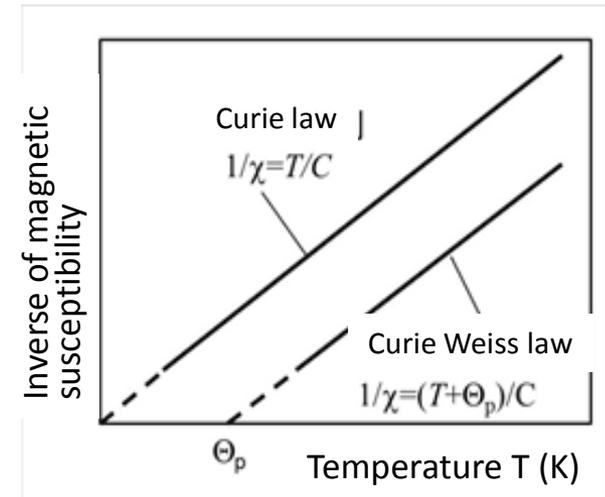


Fig.21 Curie law and Curie Weiss law

➤ Explanation of Curie Weiss Law by Mean field Approximation

- Effective field is expressed by eq.(19)

$$H_{eff} = H + H_E$$

and the exchange field H_E is proportional to M as given by eq (18)

$$H_E = AM$$

- In $T > T_c$ Curie law is satisfied for effective field:

$$\chi = M/H_{eff} = M/(H + H_E) = C/T$$

Here C is a constant.

From two equations we get

$$M/(H + AM) = C/T$$

from which $M = CH/(T - AC)$ is obtained.

Then paramagnetic moment can be written as

$$\chi_p = M/H = C/(T - \Theta) \quad (27)$$

Here $\Theta = AC$ is paramagnetic Curie temperature.

Hysteresis and Coercivity

MYSTERY OF MAGNETICS

WHAT IS HYSTERESIS?

➤ Magnetic Recording and Hysteresis

- In the computer storage HDD (hard disk drive) is used, in which information is recorded on a circular magnetic medium.
- Fig.22 shows an MFM (magnetic force microscope) image of recorded states, showing arrays of magnetic poles N and S aligned along the circumference on the disk surface.
- Schematically permanent magnets with different NS direction align along the circumference with a magnetic moment directed perpendicular to a disk surface.
- The mechanism of magnetic recording is supported by the hysteresis of the magnetic recording medium.

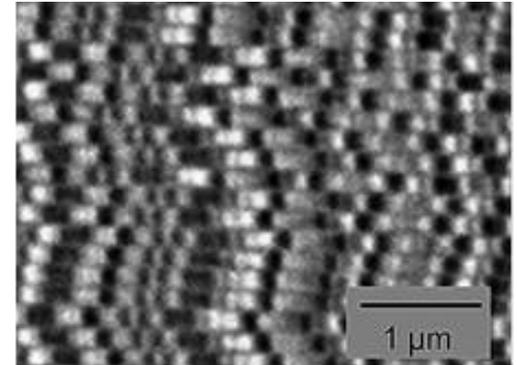


Fig. 22 MFM image of recorded domains on the perpendicular recording disk
Courtesy of Prof. Futamoto (Chuo Univ.)

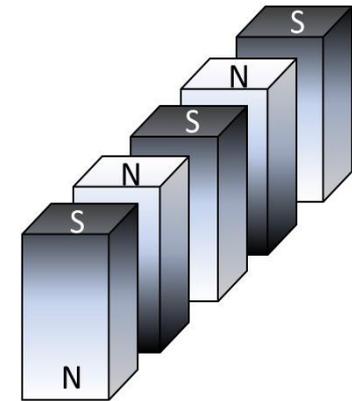


Fig. 23 Schematic illustration of recorded domains on perpendicular recording disk.

Magnetic Hysteresis Curve

- Fig. 24 provides a typical hysteresis curve of magnetization against applied magnetic field.
- Since hysteresis loop provides two values at zero field a magnetic recording is possible by assigning the two values to 1 and 0.

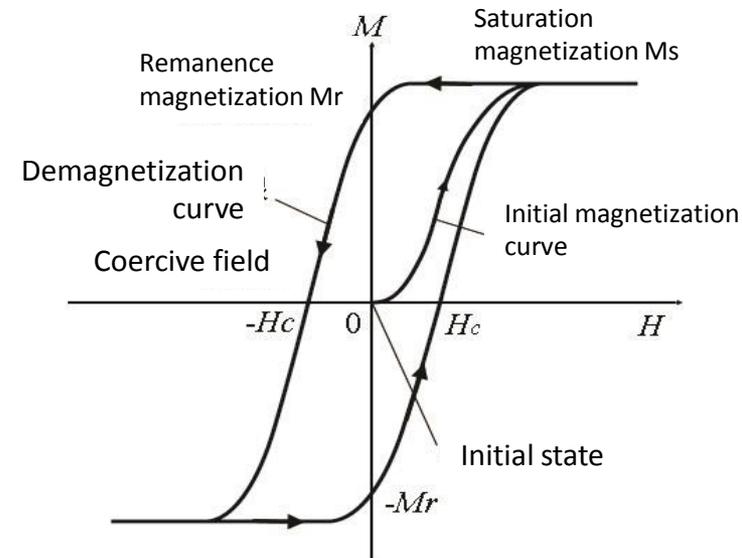


図4.3 強磁性体の典型的な磁化曲線

Hysteresis curves in ferroelectric materials

- Hysteresis is also observed between polarization P and electric field E in ferroelectric materials.
- Fig.24 shows a dielectric hysteresis loop in a ferroelectric materials TGS. In this figure vertical axis represents an electric displacement $D = \epsilon_0 E + P$ and horizontal axis is electric field.
- A ferroelectric memory device (FeRAM) makes use of residual polarization P_r to record information.
- The material has two different stable states with a potential barrier between two states. Transition between the states needs to surpass the potential barrier. Such a situation is called “bistable” and may show a hysteresis.

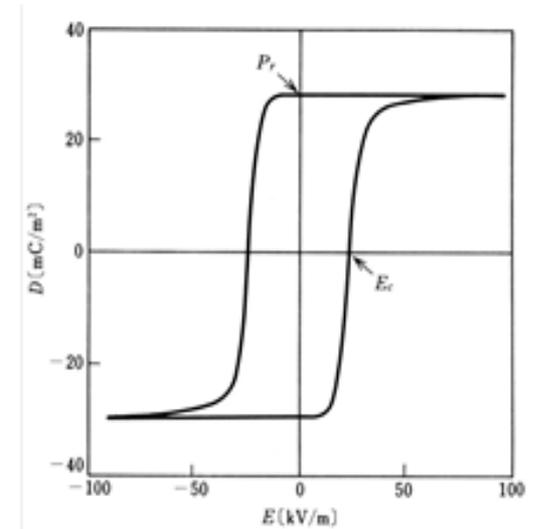


Fig. 24 A typical hysteresis curve of a ferroelectric material

Hysteresis in mechanical system

- Hysteresis phenomenon can be also observed mechanical system. Gear 2 follows Gear 1 when Gear 1 is rotated toward left, but follows after the “backlash” when inversely rotated.
- This system is also bistable since two states (Gear 1 is attached to right wall and left wall of Gear 2) with a threshold of the backlash.

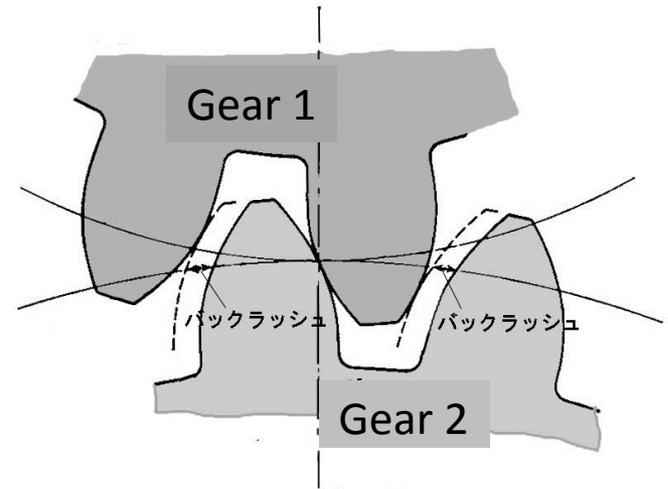


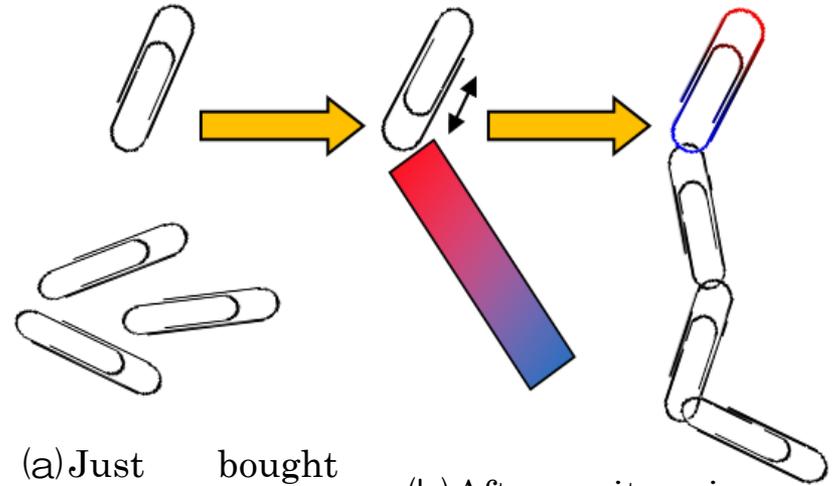
Fig.25 Gears show hysteresis

Etymology of “hysteresis” is a Greek word expressing a delay, meaning a slow response. Hysteresis is often confused with history.

WHAT IS MAGNETIC DOMAIN?

Just bought clips and magnetized clips

- A just bought clip cannot lift other clips by touch. However, once the clip is rubbed by a magnet, it attracts other clips as if the clip is a magnet.
- Why the clip became a magnet?



(a) Just bought clip cannot attract other clips.

(b) After it is rubbed by a magnet, the clip become attracting other clips

Fig.26 Magnetize a clip by rubbing with a magnet

Initial magnetic state of iron

- A polarization microscope image shows that iron is divided in different areas with different contrast. Each area is called the “magnetic domain”. Total magnetization of domains are cancelled. This is the reason why virgin iron shows no magnetization.
- When the magnetic field is applied the domain parallel to the field increases its area and do not recover the initial state even after the field is removed.

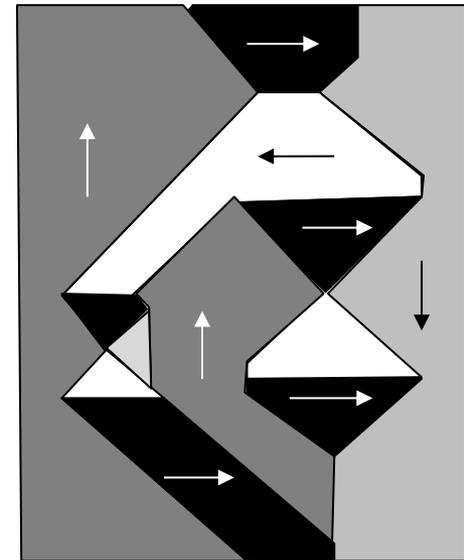


Fig.27 Schematic illustration of polarization microscope image showing a magnetic domain structure before application of magnetic field.

Demagnetization field

- Atomic magnets are aligned in the same direction to bring total magnetization.
- The pole N of a moment is attached to the pole S of the adjacent magnetic moment and internal poles are cancelled out and only the poles in the both sides remain.

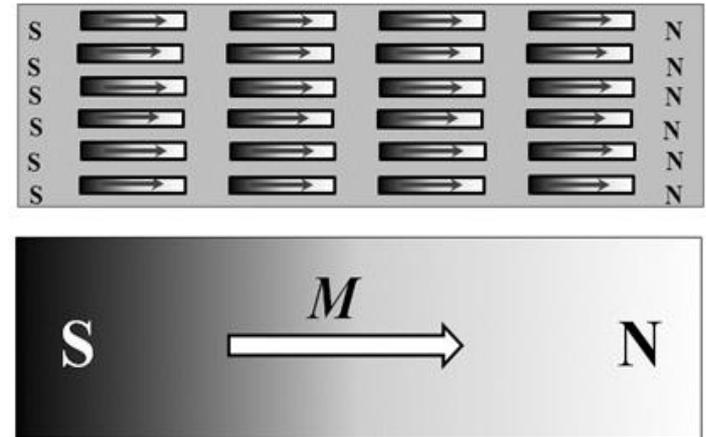


Fig.28 There are number of atomic moment exist in the magnet and magnetic poles of adjacent moments are cancelled out and poles in the both remain.

Demagnetization field come from magnetic poles

- Since magnetization M and magnetic flux density B are continuous, inside and outside magnetic flux lines are connected continuously as shown in Fig.29.
- On the other hand magnetic force line flows from the pole N and is absorbed into a pole S as shown in Fig.30 regardless of inside or outside of the magnet.
- Since the magnetic field outside the magnet is $H=B/\mu_0$, the magnetic force line and magnetic flux line share the same direction, while the field direction inside magnet is opposite to the magnetization direction. This opposite field H_d is called “demagnetization field”.

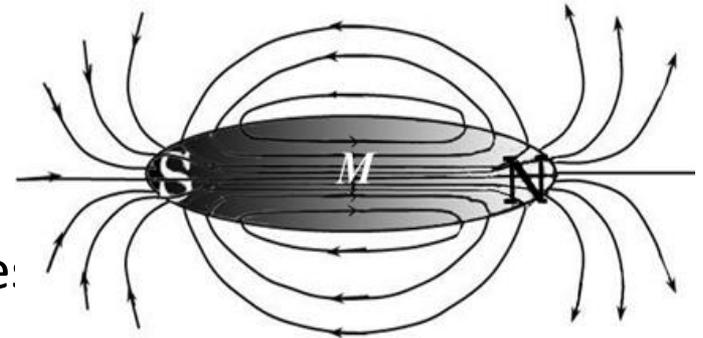


Fig.29 Flux line is continuous to magnetization

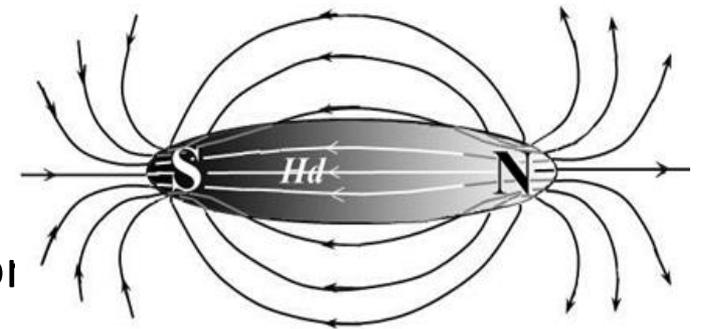


Fig.30 Force lines spring out from the pole N and flow to the pole S .

Coefficient of demagnetization depends on the shape of a magnet

- Demagnetization field H_d [A/m] is proportional to magnetization M [T], since the field is generated from magnetic poles.

$$\mu_0 H_d = -NM \quad (28)$$

- The coefficient N is called “demagnetization field coefficient”. Since both demagnetization field and magnetization are vectors \mathbf{H}_d , \mathbf{M} , respectively, the coefficient should be expressed by a tensor \mathbf{N} and is described as:

$$\mu_0 \begin{pmatrix} H_{dx} \\ H_{dy} \\ H_{dz} \end{pmatrix} = - \begin{pmatrix} N_x & 0 & 0 \\ 0 & N_y & 0 \\ 0 & 0 & N_z \end{pmatrix} \begin{pmatrix} M_x \\ M_y \\ M_z \end{pmatrix} \quad (29)$$

Demagnetization coefficient depends on the shape and direction of a magnet

	N_x	N_y	N_z
Sphere	1/3	1/3	1/3
Infinite cylinder	1/2	1/2	0
Circular disk	0	0	1

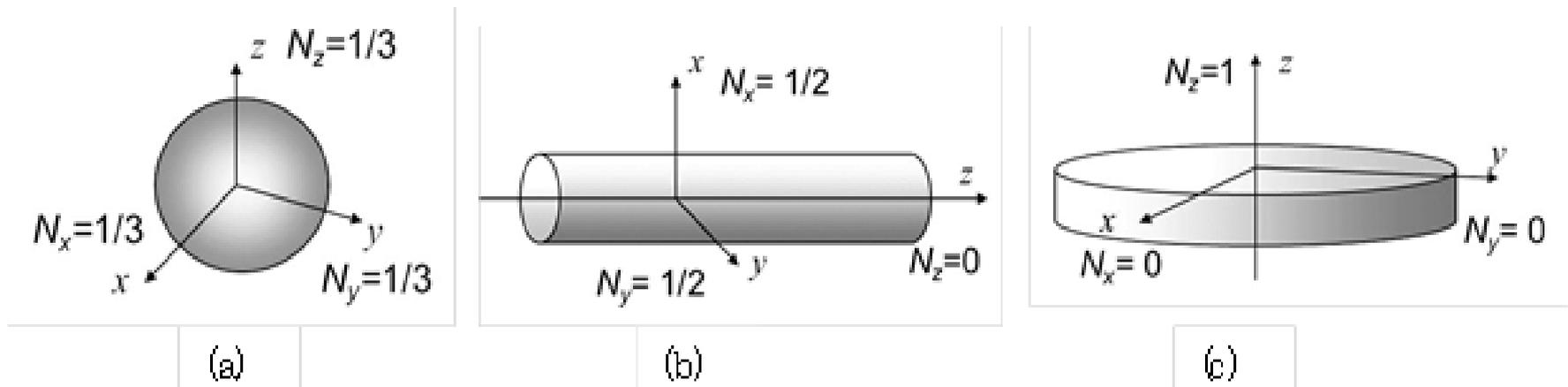


Fig.31 Table of demagnetization coefficient

unit : SI + E-H correspondence

Reason why a magnet is divided into

- The atomic moment N faces the edge N-pole and atomic moment S faces the edge S-pole as shown in Fig.32 and become unstable due to loss of static magnetic energy.
- If total area is divided into right-pointing and left-pointing areas in the stripe as shown in Fig.33, demagnetization fields cancel out and stabilized by lowering of the static magnetic energy. This is the reason why a magnet is divided in magnetic domains.

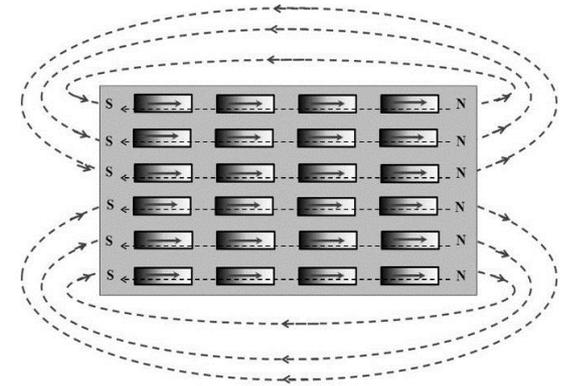


Fig.32 Inner atomic moments are unstable due to demagnetization field.

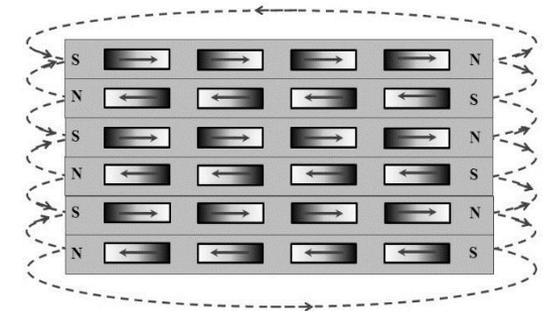


Fig.33 By division to stripe domains atomic moments become stable

➤ Domain Wall

- Although static magnetic energy is lowered by an introduction of domains, an exchange energy at the boundary is increased.
- Therefore, atomic moment cannot abruptly change direction as large as 180° , but gradually change over a few atomic layers. This transition region is called “Domain Wall”.

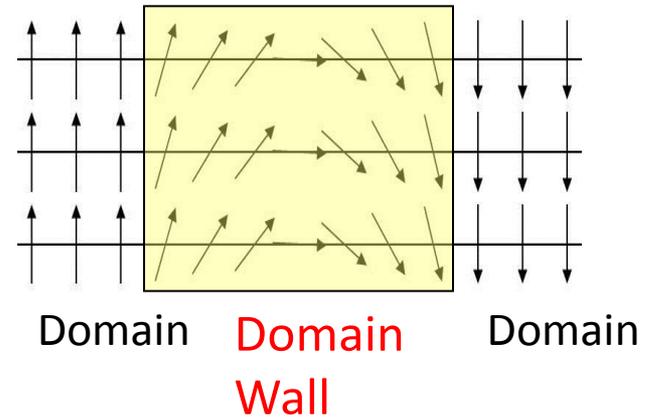


Fig.34 In the domain wall atomic moments gradually rotate to connect the magnetization of adjacent domains.

Various domains

- Closure domain: Due to an magnetic anisotropy inherent to the magnetic materials, a magnetization in a domain has tendency to be directed along a specific crystallographic orientation, eg., Since six directions $[100]$, $[010]$, $[001]$, $[-100]$, $[0-10]$, $[00-1]$ are equivalent in a cubic crystal, magnetization directions in domains are determined to circulate flux flow. This structure is called “closure domain”.
- Vortex: A magnet with a small magnetic anisotropy shows a volute magnetic structure when the size below a certain value. This structure is called “vortex”.

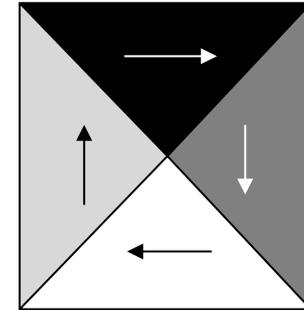


Fig.35 Closure domain

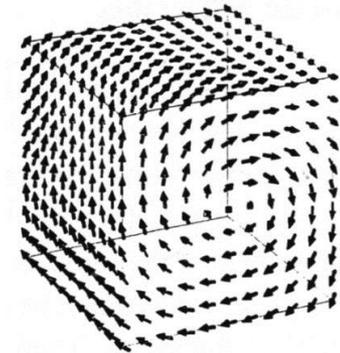


Fig. 36 Vortex

MFM Images of magnetic structure of small magnetic dots

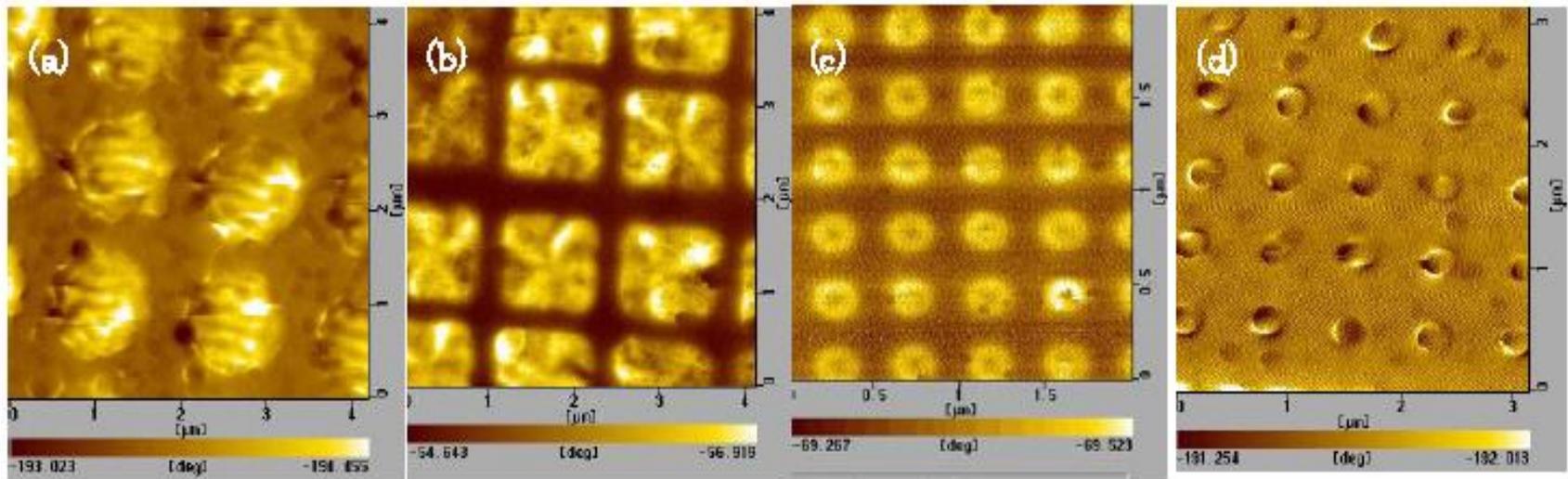


Fig.37 Magnetic structure of small magnetic dots: (a) Stripe (Co circular dot $1.2\mu\text{m}\phi$), (b) Closure (Permalloy square dot $1.2\mu\text{m}$ in edge), (c) Vortex (Permalloy circular dot $300\text{nm}\phi$), (d) Single domain (Co circular dot $100\text{nm}\phi$)

MAGNETIZATION CURVE AND MAGNETIC DOMAINS

Initial magnetization curve and magnetic domains

- At the origin of the graph in Fig.38, total magnetization vanishes due to an introduction of the domain structure.
- Imagine a case in which magnetic field is applied along the easy-axis. For applied magnetic field weaker than B in the initial magnetization curve, the magnetization gradually increases with an applied field. The change of $A \rightarrow B$ (initial region) is reversible, i.e., magnetization become zero when applied field is decreased to zero.
- By application of a field larger than H_B , a steep rise of magnetization is observed. In this region irreversible change of magnetization occurs, since domain wall traverses over the potential barriers and the wall cannot go back by decrease of the field. This region $B \rightarrow C$ is called “discontinuous magnetization region”.
- Above H_C increase of magnetization becomes slower. In this region magnetization rotates in the domain. The region is called “Magnetization rotation region”.

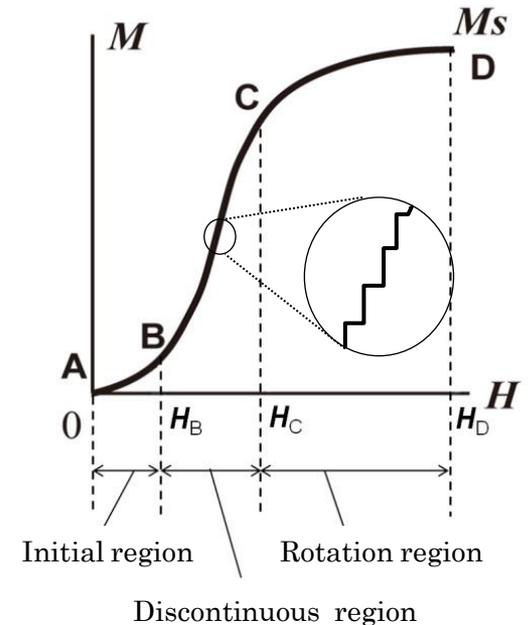


Fig.38 Initial Magnetization

Magneto-Optical Image of Domains in the Initial Magnetization Curve

- In the initial state total magnetization vanishes by domain-formation as in (a). In the $A \rightarrow B$ region domain walls move to extend the domain parallel to the field as shown in (b).
- At the steep rising region $B \rightarrow C$ irreversible domain walls motion occurs as in (c).
- Beyond H_C magnetization in a domain occurs as in (d).
- Magnetization saturation corresponds to single wall state as in (e).
- Once magnetization is saturated it never return along the initial magnetization curve but remains at a remanent state.

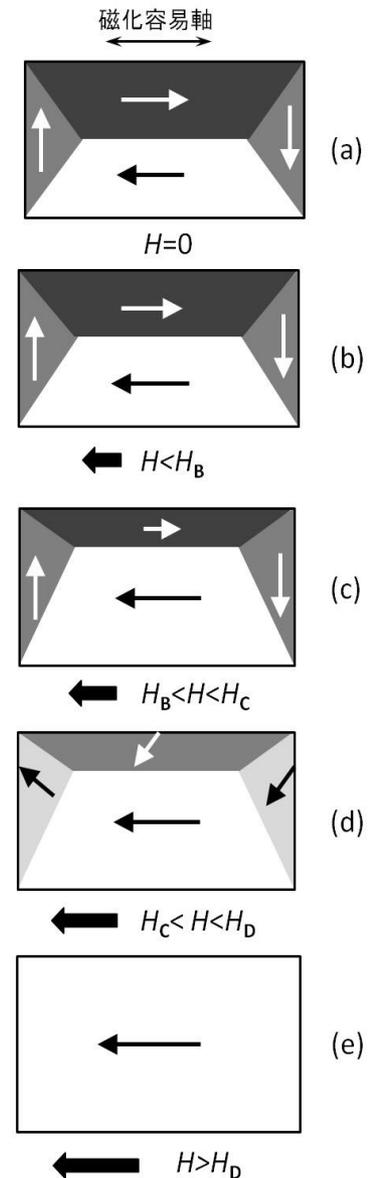


Fig.39 Domain wall motion and magnetization rotation images

Magnetic Anisotropy

- Most important reason for an irreversible magnetization process observed in the magnetic hysteresis curve is a “magnetic anisotropy”.
- There are several physical origins of magnetic anisotropy: shape anisotropy, magneto-crystalline anisotropy, induced anisotropy.

Shape Anisotropy

- Magnetic anisotropy occurs due to dependence of the demagnetization field on shape and orientation.
- Needle-like crystal has an easy axis along axial direction, since it shows a negligibly small demagnetization field along the needle axis.
- In thin films easy axis lies in plane parallel to the film surface, since demagnetization does not work in plane but work strongly perpendicular to the surface of the film.

Magneto-crystalline Anisotropy

- In crystals a specific crystal axis often becomes an easy axis of magnetization, due to magneto-crystalline anisotropy.
- Since Co crystallize in hexagonal lattice, c-axis of the hexagon becomes an easy axis.
- Fe crystallizes in body centered cubic (BCC) lattice, it is isotropic in most physical properties such as dielectric constant and conductivity. On the other hand magnetization of Fe is subjected to anisotropy with an easy axis along $\langle 100 \rangle$, and hard axis along $\langle 111 \rangle$ direction.

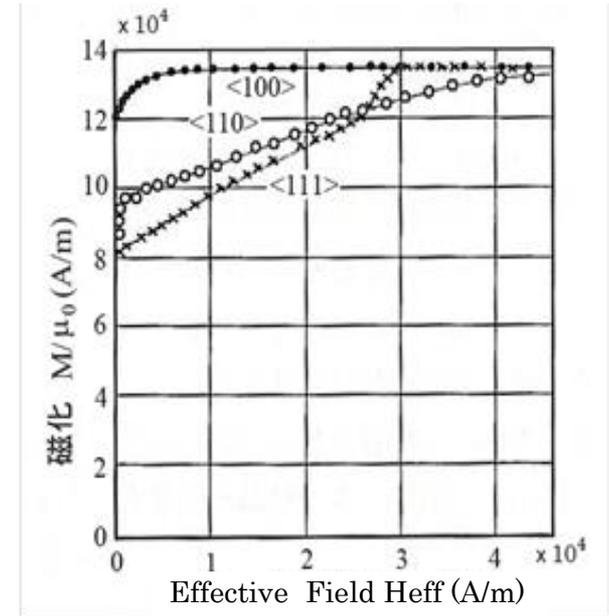


Fig.40 Crystal axis dependence of magnetization curves of Fe

Why magnetization of Fe shows anisotropy?

- Domain wall motion is different for crystalline axis.
- Application of a magnetic field along [100] axis expands domains (colored in black) to form a single domain structure as shown in Fig.41 (saturation). Such domain wall motion occurs with a small energy. This corresponds to $\langle 100 \rangle$ curve of Fig.40
- Application of a field along [110] direction 45 deg from [100] axis, two equivalent domains expand to fill the area with a weak field to realized a magnetization $M_s/\sqrt{2}=0.71M_s$. Increase of the field induces a magnetization rotation leading to saturation as shown in Fig.42. This correspond to the $\langle 110 \rangle$ curve of Fig.40.

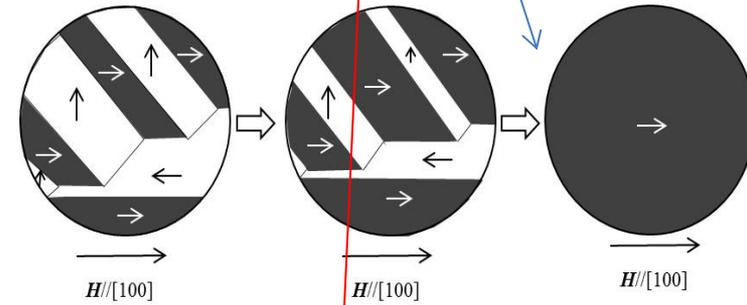
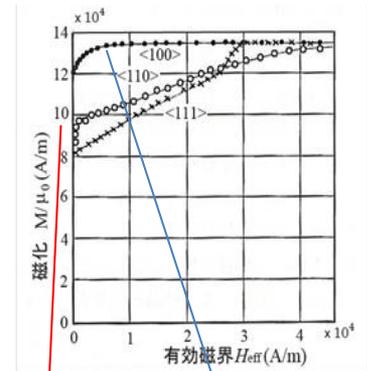


Fig.41 Domain wall motion when magnetic field is applied along $\langle 100 \rangle$

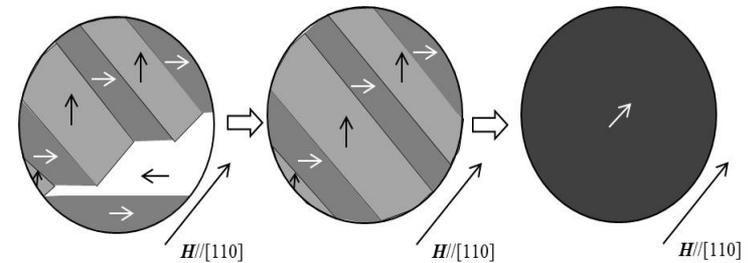


Fig.42 Domain wall motion when magnetic field is applied along $\langle 110 \rangle$ direction.

↑ and → domains are equivalently fill to realize a magnetization of $M_s/\sqrt{2}$, followed by magnetization rotation leading to saturation

Induced anisotropy

- This type of anisotropy is induced during preparation of materials. It occurs in cases as (1) growth in the applied magnetic field, (2) lattice-mismatch between the film and substrate, (3) Specific atom pair is formed during sputter deposition.
- For example, amorphous rare earth (RE)-transition metal (TM) alloy films such as TbFeCo employed for magneto-optical recording show perpendicular anisotropy. This anisotropy is caused in part by a pair of elements generated during sputter-deposition and also by a single atom anisotropy inherent to RE.

Magnetic Anisotropy Energy

Magnetic anisotropy energy is an energy necessary to orient a magnetization from the easy axis to the hard axis.

Anisotropy energy E_u of a uniaxial magnet when a magnetic field is applied with an angle θ from easy axis is expressed by

$$E_u = K_u \sin^2 \theta. \quad (30)$$

Here K_u is an anisotropy constant in unit $[J/m^3]$

Fig.43 shows a plot of the anisotropy energy as a function of θ . It takes a minimum value at $\theta=0^\circ, \pm 180^\circ$ ($\langle 100 \rangle$ direction) and a maximum value at $\theta=90^\circ, -90^\circ$ ($\langle 110 \rangle$ direction) for $K_u > 0$.

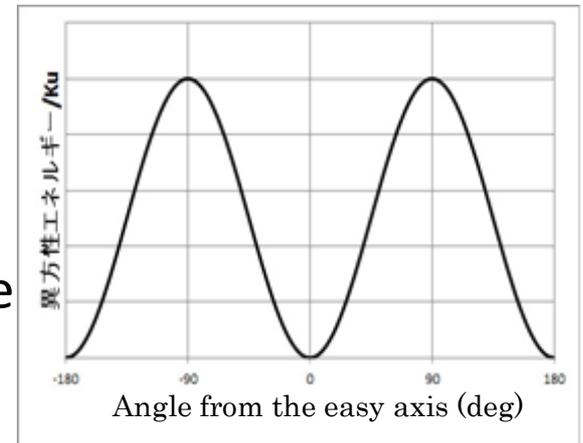


Fig.43 Magnetic Anisotropy as a function of angle from the easy axis

Anisotropy Field H_K

Restoring force when a magnetic field is applied to a magnet with anisotropy K_u with a small angle $\Delta\theta$ from the easy axis is expressed as

$$F = \partial E_u / \partial \theta = K_u \sin 2\Delta\theta \sim 2K_u \Delta\theta. \quad (31)$$

Anisotropy field H_K is defined as a magnetic field working on the magnetization M_0 along the direction inclined at a small angle $\Delta\theta$ from the easy axis necessary to give the same restoring force as given by eq.(31). This force is expressed as

$$F = \partial E / \partial \theta = - \partial M_0 H_K \cos \Delta\theta / \partial \theta = M_0 H_K \sin \Delta\theta \sim M_0 H_K \Delta\theta \quad (32)$$

By comparison of eq.(31) and eq.(32) we get as anisotropy field

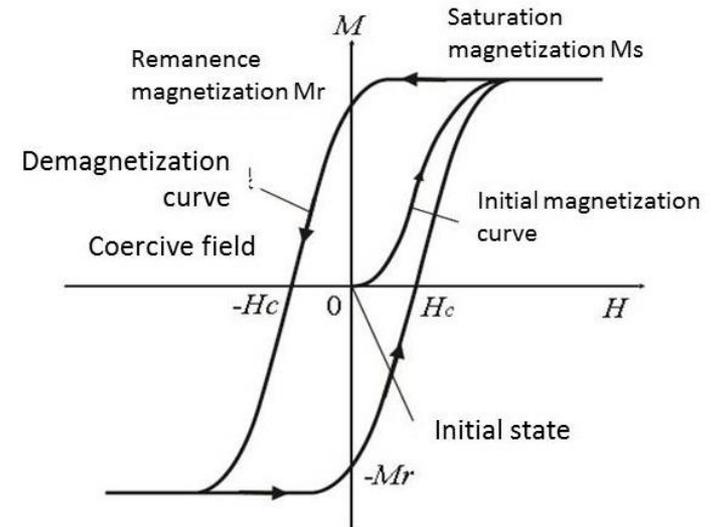
$$H_K = 2K_u / M_0. \quad (33)$$

Anisotropy field of single crystalline particle of Co is estimated as $H_K = 5.06 \times 10^5$ [A/m] using the anisotropy constant of Co, $K_u = 4.53 \times 10^5$ [J/m³], and magnetization of $M_0 = 1.79$ [Wb/m²]

MYSTERY OF COERCIVE FORCE

What is coercive force?

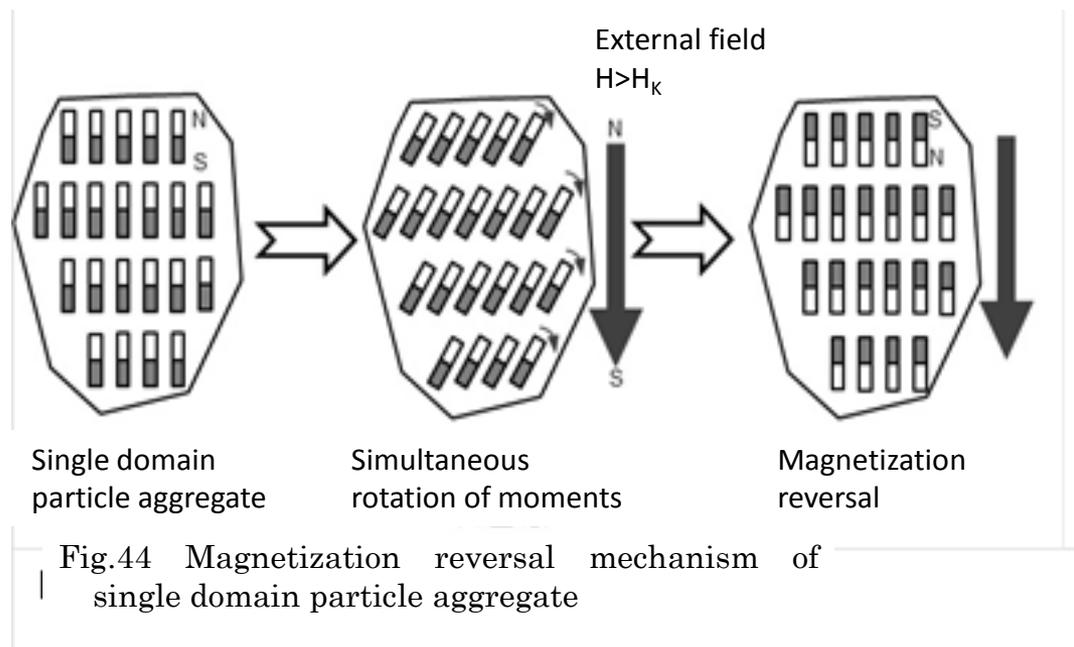
- Intersection of demagnetization curve and horizontal axis is called “coercive field” or “coercive force” and denoted as H_c . The term coercive stands for compulsive or forced. Therefore it means the field necessary to make magnetization vanish.
- In general coercive field H_c is much smaller than the anisotropy field H_K .
- **Coercive force depends on the preparation process and “structure sensitive”. The mechanism of coercive force has not still been fully understood**



Coercive Force of single domain nanoparticle aggregate

Magnetic nanoparticle shows a single-domain structure. Consider an aggregate of such nanoparticles. According to the Stoner–Wohlfarth model all the magnetic moments rotate simultaneously and the system can be represented by a vector \mathbf{M} . In this case the coercive force is given by the anisotropic field H_K , and can be expressed by

$$H_c = \frac{2K_u}{M_0} \quad (34)$$



Coercive force in the presence of a domain structure

Once a domain wall is introduced, it can smoothly move in the magnet, leading to an easy magnetization reversal. This occurs even in a highly anisotropic magnet. In an ideal case the external field necessary to generate a reverse domain is equal to the anisotropic field. However, in reality reduction of anisotropic field at some point on the grain boundary, or local enhancement of demagnetization field occur, leading to a reduction of H_c from H_K as expressed by eq. (35)

$$H_c = \alpha H_K - NM_0 \quad (35)$$

Here α is a reduction factor ($\alpha < 1$) of anisotropic field and N coefficient of demagnetization influence by adjacent grains.

Suppression of domain wall nucleation is a crucial issue in development of a permanent magnet. For this purpose in Nd-Fe-B magnet, Dy which possesses a high anisotropy field is diffused at the grain boundary.

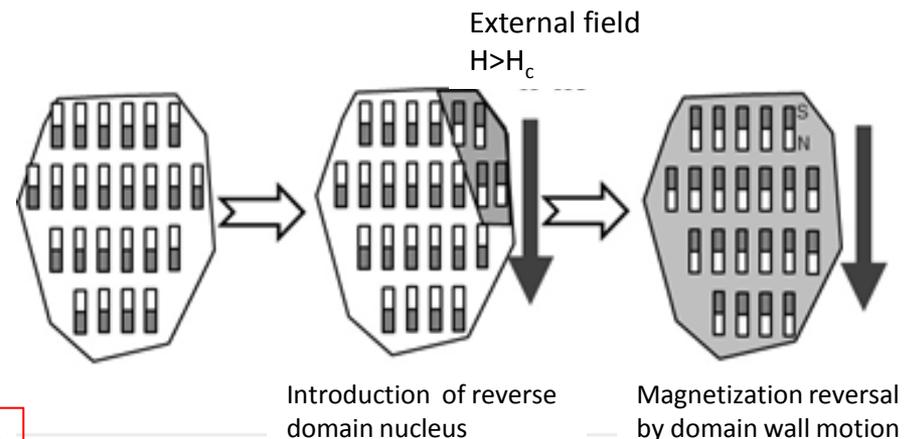


Fig.45 Magnetization reversal mechanism in the case of domain nucleation type magnet.

Coercive force in the presence of pinning sites

- Pinning site will trap a wall. Once the wall escapes from the pinning site magnetization reversal proceeds until it is trapped again by the second pinning site. Trapping is caused by a difference in the wall energy at the pinning site and the rest. The existence of pinning sites increases a coercive force. SmCo magnet is known as of this type.
- Pinning sites are introduced by grain boundaries, lattice defects and impurities, so that they depend preparation process of materials, which is the matter of engineering.

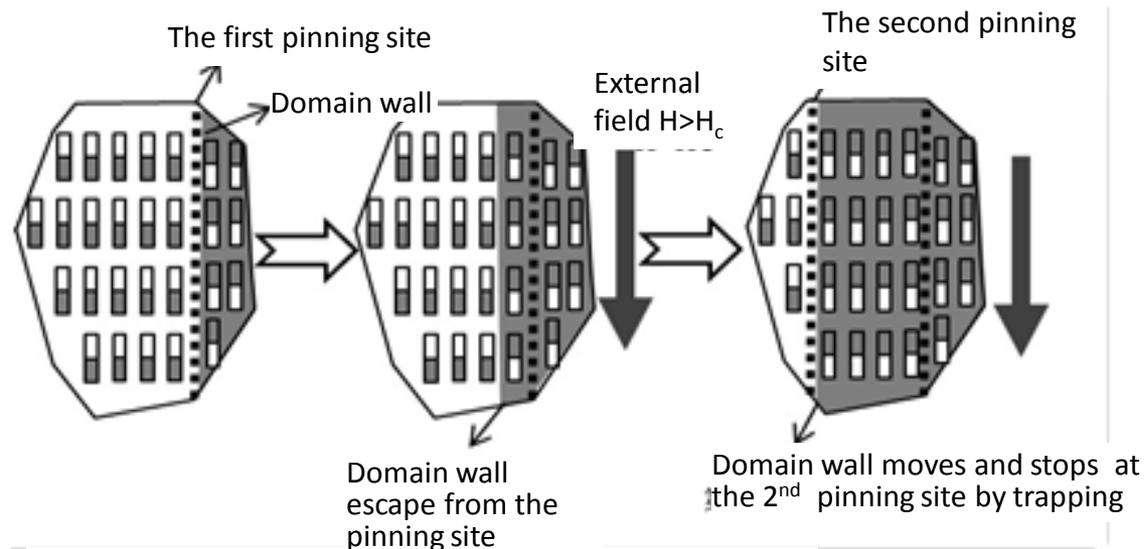


Fig.46 Magnetization reversal mechanism in the case of existence of wall-pinning sites.

Mystery of Remanence

Ratio of remanence to saturation magnetization is called “squareness”. It is an important parameter for magnetic recording.

A schematic illustration of domain structure at the reduction of magnetic field from the saturation (a) to remanence (c) is shown in Fig.47.

The single domain state of (a) is unstable but it is forced by an external field.

Therefore, reduction of the external field triggers a nucleation of domains with different magnetization directions.

Once a nucleus is generated it is subjected to wall motion and rotation as shown in (b).

Pinning sites suppress the expansion of the reverse domain and leave the total magnetization not vanished.

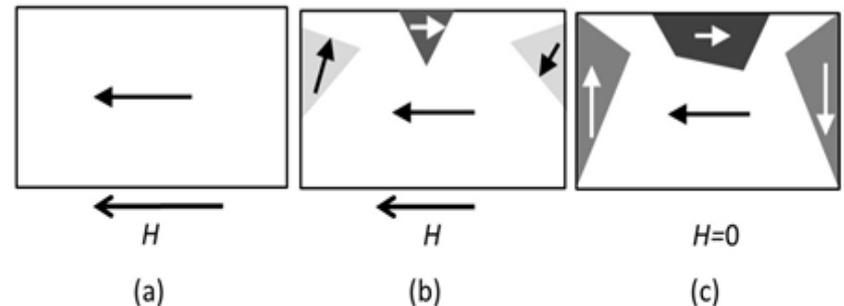


Fig.47 Schematic illustration of a mechanism for remanence magnetization

Summary

- For research and development of spintronics macroscopic phenomenon of magnetism should always be considered.
- In this lecture microscopic origin of magnetic moment in materials and macroscopic origin of hysteresis phenomena are reviewed.
- Microscopic origin of magnetic anisotropy as well as hysteresis and remanence is still unexplained.