Photoinduced Irreversible Effects on Magnetic Properties and Allied Phenomena in Magnetic Oxides V

K. Hisatake, I. Matsubara, K. Maeda, *T. Fujihara and **S. Kainuma

Department of Physics and *Chemistry,
Kanagawa Dental College,
Yokosuka, Kanagawa 238 Japan
**Department of Electrical Engineering,
Ashikaga Institute of Technology
Ashikaga, Tochigi 326 Japan

I. INTRODUCTION

The purpose of the present review is twofold ; (i) to mention the simple survey of study of disaccommodation (*DA*), with special emphasis on that of the spinel ferrites ; (ii) to discuss the photoinduced *DA* found, for the first time, and a model proposed recently by our research groups (1991) [1, 2]. It should be noticed that this effect is photoinduced phenomenon but does not indicate the coexistence of phtoinduced irreversible effect and *DA* [3]. Gamma-ray induced effect [4, 5] is also found, which will be reported in a next issue, correlated with a deep center [6].

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II. REVIEW OF STUDY OF DISACCOMMODATION (*DA*) IN SPINEL FERRITES

2. 1 General Perspectives

In the series of the reports, the simple review of *DA* has already been touched upon [1]. The recent observation of new phenomenon of *DA* and the contents of title are intimately related as shown below. Hence, this review of *DA* is again described from the renewed point of view. In general, the induced preferred directions of magnetization

parallel to the field in the region of the Bloch walls create localized potential minima. As these minima become deeper with time, the mobility of the walls becomes more restricted, resulting in a decrease in μ_i. On this decrease in μ_i with time spontaneously after demagnetization, so celled, disaccommodation (*DA*) [2, 3], much works have been studied in spinel ferrites but very few systematic work for garnet ferrites, since the application of the latter substance is limited to the high frequency or microwave region [4, 5]. *DA* is a kind of the magnetic annealing effects and the result of time dependent induced anisotropy [6]. The effects appear as the result of some directional ordering of defects or local anisotropic configurations in the crystal. In spinels, these are usually cation vacancies, substituted ions or impurities, anisotropic short range order configurations or local Jahn-Teller distortions. In order to achieve some kind of directional order of the imperfections with respect to the direction of magnetization, double requirements must be satisfied a) an anisotropic coupling exists between the magnetization vector and the local configurations (defects), and in some range of T>Tc these imperfections are allowed to move or to transform into other ones with a lower energy of their coupling to magnetization. In such a way the magnetic anneal reflects the ability of the crystal to adapt itself by an inner rearrangement in the lattice to the given magnetic state and to stabilize this state by lowering its free energy. If the magnetization is homogeneous, a macroscopic anisotropy or induced anisotropy, as already shown, it is described by

$$F_i = -F\Sigma\alpha_i^2\beta_i^2 - G\Sigma\alpha_i\alpha_j\beta_i\beta_j, \qquad (2.1)$$

where direction cosines β_i and α_i refer to the orientation of magnetization during the annealing process and anisotropy measurement respectively [2]. For polycrystals we obtain by averaging

$$F_k = K_n\cos^2\theta, \qquad (2.2)$$

here, $K_n = 2/5F + 3/5G$, and θ_i is measured from the direction of the annealing field [3]. The anisotropic coupling responsible for local directional order is of the same origin as that leading to the magnetocrystalline anisotropy ; dipole-dipole interactions, single ion anisot ropies and anisotropic exchange in pairs or clusterrs. For a given direction of magnetization the configuration with the lowest anisotropy energy is stable. When the magnetization is rotated, the energy of another configuration may become lower. Nevertheless, if the relaxation time of the spin system is sufficiently long, the initial configuration is tem- porarily retained and an induced anisotropy is observed.The relaxation of this anisotropy is realized by the rearrangement of the spin system. Spinel ferrites often show deviation from the oxygen stoichiometry (oxygen excess, contrary to the case of garnets) which leads to the presence of cation vacancies. These trigonal axis correspond to four $\langle 111\rangle$ crystal directions, namely [111], $[\bar{1}11]$, $[1\bar{1}1]$ and $[11\bar{1}]$. If their concentration on B sites with the [111] local trigonal axis differs from those in other octahedral positions, the dipole-dipole interactions contribute to an axial anisotropic term of the form [7]

$$E_o^{dip} = A_{dip}N(\bar{m} - \bar{m}')(\alpha_1\alpha_2 + \alpha_2\alpha_3 + \alpha_3\alpha_1) \qquad (2.3)$$

with $\bar{m}'$, $\bar{m}$ denoting the average magnetic moments in the [111] sites and the other sites,

respectively and N is the number of B sites per unit volume and $A_{dip} \simeq 4 \times 10^{-18}$ erg/cm^3. Entropy of this system is expressed by the equation,

$$S \simeq 3nk \log n + n' \log n' \quad (n, n' \ll N) \qquad (2.4)$$

where N is the total number of B-sites in the whole crystal, n the number in [111]-sites and n' the number in the other sites. If the direction of magnetic moment is [111], then the free energy is

$$F = E^a_{dip} - TS$$
$$\simeq 4A\bar{m}(n-n') - kT(3n \log n + n' \log n'). \qquad (2.5)$$

From the minimizing condition of the free energy,

$$n - n' \simeq -N_v/4 \cdot 16A/3kT \cdot m^2 \quad (A \ll kT) \qquad (2.6)$$

and so,

$$F^a_{dip} = -3kT(\alpha_1\alpha_2\alpha_1'\alpha_2' + \alpha_2\alpha_3\alpha_2'\alpha_3' + \alpha_3\alpha_1\alpha_3'\alpha_1') \qquad (2.7)$$

This leads to the induced anisotropy of a G type (*i. e.* $F=0$) as follows,

$$K_T = G\ (T_a, T) = N_v 16A^2/3kT(g\bar{S})^4 (M(T)/M_0)^2 (M(T')/M_0)^2 \qquad (2.8)$$

where T_a and T are the annealing and mesuring temperatures respectively. $(\alpha_1, \alpha_2, \alpha_3)$ and $(\alpha_1', \alpha_2', \alpha_3')$ are the annealing and measuring direction of $g\bar{S}$; average magnetic moment in μ_B. Note that such an anisotropic distribution of vacancies and/or nonmagnetic ions also causes the distribution of magnetic ions to the induced anisotropy via the mechanism of single ion anisotropy or anisotropic exchange [8].

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2. 2 Cation Vacancy in Spinal Ferrites

The rate at which cations at B sites of the spinel lattice can migrate to the prefferred sites depends on the cation vacancy concentration since the cation must have a cation vacancy as a nearest neighbor in order to move. For ferrites not containing Co, the ionic rearrangement model has been generalized by Iida and Inoue [1]. In their model the induced anisotropy is due to an energetically favorable arrangement of the constituent ions on the various sites of the spinel lattice. The cation vacancy concentration determines only the relaxation time of the ionic rearrangement. On the other hand, Yanase [2] and Ohta [3] showed that the anisotropy is induced by a prefferential location of cation vacancies in the lattice. Cation vacancies were found to have direct effect on DA. There are two possible mechanisms by which the cation

vacancy contributes to it. One is the case which the vacancy accelerates the diffusion of metal ions in the spinel lattice without any direct contribution to the anisotropy. In the other case, the vacancy itself makes a direct contribution to the anisotropy through the displacement of its position in spinel B-sites. As suggested by Yanase [2], Fe^{2+} facilitates the diffusion of cation and makes DA rapid. Moreover, it was theoretically concluded that the preferential occupation of cation vacancies is large enough to DA at low frequencies, as already described in the Chap. 2. 1.

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2. 3 Spinel Ferrites Containing Co^{2+} Ions

In the case of spinel ferrites containing the Co^{2+} ions they may be thought of as isolated. In the presence of their low lying levels corresponding to the spin Hamiltonian are anisotropic and hence their equilibrium distribution among the four types of the B positions will depend on the direction of magnetization. The resulting induced single ion anisotropy is again of a G type with [1]

$$G=nN[(\sqrt{3}|\alpha\lambda|/2)\tanh(\sqrt{3}|\alpha\lambda|/2kT)-(\alpha\lambda)^2/4\cdot\mu_B H_{ex}]$$
$$n=(N_{1\bar{1}1}+N_{111}-N_{11\bar{1}}-N_{\bar{1}11})/N, \qquad (2.9)$$

where T is the temperature of measurement, N the total number of Co^{2+} in the B sites, N [1$\bar{1}$1] their equilibrium number in positions with local axis in [1$\bar{1}$1] etc., λ the parameter of spin-orbit coupling and α a numerical factor, $1<\alpha<3/2$. In the special case of Co substituted magnetite $|\alpha\lambda| \simeq 132cm^{-1}$ and $\mu_B H_{ex} \simeq 320cm^{-1}$ were estimated [1]. Single ion contributions to the induced anisotropy arising from the preferential distribution in the B sites are also expected from ions with a singlet orbital ground state, via the D term in the spin Hamiltonian. These anisotropies are usually weak but in special cases (Fe^{2+}) they may achieve a considerable strength [3, 4]. If the concentration of Co^{2+} (or other active ions) is increased these ions cannot be considered as isolated and contributions of more complicated local configurations including pairs and eventually larger groups of active ions become important [5, 6]. As an example a cluster containing an isolated $Co^{2+}-Co^{2+}$ pair is shown in Fig. 2. 1. It should be noted that there are six inequivalent orientaions of this pair corresponding to six face diagonals ⟨110⟩. If the occurence of differently oriented pairs is unequal, an induced anisotropy appears depending quadratically on the Co^{2+} concentration. Experimentally this was found to be almost a pure F-type anisotropy ($G \simeq 0$). The origin of the anisotropy may be sought in both the anisotropic exchange and a local modification of the ligand field changing the single ion contributions of each ion within the pair [7]. In the case of a small part of octahed ral positions is occupied by Mn^{3+} ions the overall symmetry of the crystal remains cubic, but local Jahn-Teller distortions may appear. Assuming these to be static the ligand field of Mn^{3+} lowers its symmetry which usually becomes tetragonal. The so called D term in the equation of spin Hamiltonian [7]

$$H=\mu_B H_{ex} gS+D[S_\zeta^2-1/3S(S+1)]+1/6a\{S^4_x+S^4_y+S^4_z-1/5S(S+1)(3S^2+3S-1)]$$
$$+1/36F[7S_\zeta^4-6S(S+1)S+5S_\zeta^2-6/5S(S+1)+3/5S^2(S+1)^2]. \qquad (2.10)$$

then yield uniaxial anisotropy of F-type ($G\simeq 0$) provided a preferential distribution of the distortion axes may be created by magnetic anneal. It should be noted that due to local stresses and other defects often only a small part of the distortions is expected to be free to move and to take part in the process [8]. The microscopic mechanism underlying the relaxation process mentioned above usually have the character of a diffusion process (e. g. migration of ions and or vacancies, electron hopping) [9]. In order to achieve thermodynamic equilibrium in the magnetic annealing process some rearrangements in lattice should be fulfiled. The relevant relaxation times depend exponentially on temperature according to the Arrhenius relation, $\tau=\tau_0 \exp E/kT$ [10]. Except for a few simple cases [see Appendix I] when isolated ions or vacancies rearrange, two or more relaxation times usually exist. In practice, the so-called logarithmic distribution of relaxation times is considered as a good approximation. If the diffusion in question concerns the ions, the activation energies E are typically between 0.7-2.5 eV depending on the type of ions and that of the process [11]. Generally the presence of cation vacancies makes the diffusion process faster, the factor E is lowered and the preexponential factor τ_0 in Arrehnius equation becomes inversely proportional to the vacancy concentration within certain limits [12]. A detailed analysis of the vacancy assisted diffusion of Co^{2+} ions in magnetite was made by Iida *et al* [13]. When the local anisotropic configurations refer to anisotropic distribution of electronic charge (*e. g.* the distribution of Fe^{2+} and Fe^{3+}), the rearrangement process is attained easily by a migration of electrons. The activation energy is then less than 0.7 eV and often 0.1 eV [14]. A special case is represented by the local Jahn-Teller distortions [15]. They are connected with the ordering of partly orbitals and their reorientation does not need any diffusion even though some thermal activation is necessary in order to overcome a potential barrier. It is possible that the distortion is usually stabilized by some defects. Previously one of the authors reported the posibility that they reorient due to movement of these defects. However, rigid evidence to support the assumption has not been obtained. So far most of the information on induced anisotropy effects is described to be obtained. But a sensitive and convenient measurement of DA is used in our research. As the anisotropic effect of Co^{2+} is strong, it gives usually much larger contributions to the induced anisotropy than other possible sources. For low Co^{2+} concentration x the results may be inter preted in terms of local ordering of both isolated Co^{2+} ions and $Co^{2+}-Co^{2+}$ pairs in octahedral sublattice. Iida *et al.* [5] found that the presence of vacancies does not change the megnitude of the induced anisotropy but the rate of ordering τ was found to be inversely proportional to the vacancy concentration if this is larger than 10^{-5}. For lower concentrations of cation vacancies, the activation energy is increased from the usual value $\simeq$1 eV to 2 eV indicating a more difficult diffusion mechanism. The induced anisotropy is usually lowered due to fluctuations in the local symmetry which primarily concerns the G term [15]. In spinel ferrites containing both Ca^{2+} and Co^{3+} ions in octahedral sites a preferential occupation of certain positions by Co^{2+} may be achieved by the electron transfer between Co^{2+} and Co^{3+} ions. If the concentration of Co ions on equivalent sites is sufficient for a direct electron exchange $Co^{2+} \rightleftarrows Co^{3+}$, the activation

energy is low ≃0.1 eV. The Co ions are then separated by many other ions, *e. g.*, Fe^{3+}, so that the electron transfer involves also these ions as intermediat. This results in rather high activation energy up to ≃0.6 eV [16], the picture of which may be thought of as large energy gap [17].

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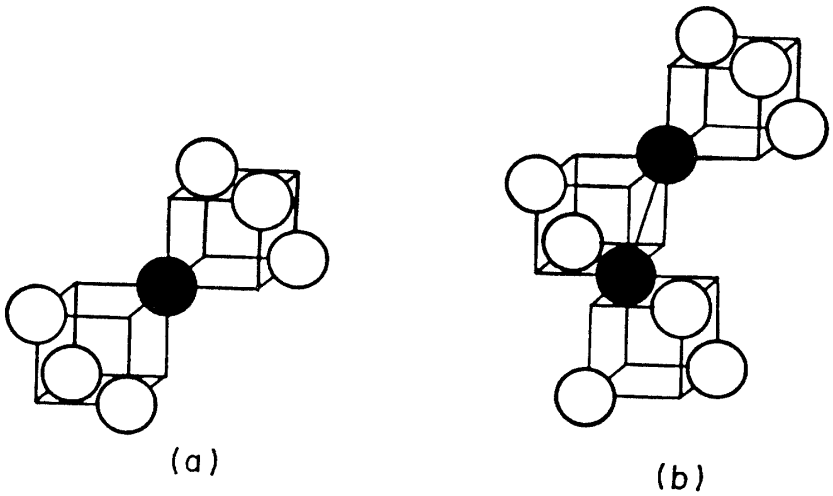
Total number or various configuration in iron-cobalt ferrites.



288
(4 × 2 ×36)

6144
(6 × 4 ×256)

Fig. 2. 1. Two fundamental units to the anisotropy [13] :
(a) single ion in various crystalline fields,
(b) paired ions in various crystalline fields.

2. 4 Spinel Ferrites Containing Fe^{2+} Ions

Induced anisotropy and various after-effect were studied in several systems containing Fe^{2+} ions usually simultaneously with cation vacancies. The after effect involving the two processes may be attributed to different distributions of Fe^{2+} and Fe^{3+} around the vacancy. Besides valency redistribution these ions may contribute to the magnetic annealing process by reorientation of local Jahn-Teller distortions as described in the chap. 2. 3. The most detailed study was performed on the $Mn_x\ Fe_{3-x}O_4$ [1]. Zaveta *et al.* [2] found that the double peaks of tan δ in the sample between 4 K and 400 K. They attributed these peaks to $Fe^{2+}-Fe^{3+}$ valency redistribution and to reorientation free Mn^{3+} distortions.

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2. 5 Negative Disaccommodation or Accommodation

Apart from so far mentioned after effect, a new phenomenon which was characterized by an increase of the initial permeability μ_i with time after demagnetization, negative *DA* or accommodation (*A*) was observed for the first time in Co-doped Fe-ferrites, by Marais *et al.* [1]. Though this phenomenon may be explained as the process of destruction of the uniaxial anisotropy induced by the demagnetization process. Then Kainuma *et al.* observed the phenomenon also in Mg-Cu-ferrits system and reported the details of the observation [2]. It should be noted that both of *DA* and *A* coexist in the same sample. They proposed a model that this phenomenon is due to domain itself but not domain wall. The mechanism has, however, not been completely established as yet. Most recently, P. Allia *et al.* observed both of *DA* and *A* in amorphous magnetic metals [3].

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III. PHOTOINDUCED DA IN YIG

3. 1 Introduction

This section mainly concerns with the recently results and continuing study in our laboratory [1, 2]. On warming, the sample after illuminating with white light at 77 K, *DA* occurs around 125 K. Unilluminated samples, however, show no *DA* over all the measuring temperatures. An origin of this phenomena is discussed in a relation with a role of oxygen vacancy and the photoexcited electron. It has been found two phenomena as the photoinduced magnetic effects on YIG ; the decrease of the real part of the permeability at low temperatures and the appearance of the peak of the imaginary part of permeability on warming after illumination. In this paper, third one, photoinduced *DA* is reported. And a model is proposed to explain these phenomena, taking account of oxygen vacancy.

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3. 2 Experimental Procedure and Results

Single crystals of YIG were grown by a floating zone method, described elsewhere. The sizes of the sample are similar : 3.6mm in outer diameter, 1.8mm in inner diameter and 1.0mm in height. Chemical analysis using a highly sensitive x-ray microanalyzer and ICP shows that Si or Ca impurities were not detected within an accuray of 0.01mol%. An annealing of these samples at 700°C for 6h in an oxygen atmosphere was carried out before the measurement in order to remove mechanical stresses. A two-phase lock-in amplifier was used for measuring both μ' and μ''. The sample was demagnetized by decreasing the amplitude of 1 kHz magnetic field from 3 Oe. As a source of illumination, white light of a halogen lamp was used, the intensity of which was approximately 0.01 W/cm^2 at the surface of the samples, neglecting the other factors such as, the absorption loss of the glass tubes and of liquid nitrogen. The sample was demagnetized at each temperature, and the permeability was measured after 5 or 50 seconds. The circles in Fig. 1 shows the real part of the permeability of YIG on warming with the ratio of 0.5K/min. after illuminated at 77K for 5 minutes. It is seen that *DA* occurs around 125 K and its maximum value reaches 25%. Unilluminated samples, however, show no *DA* over all the measuring temperatures. The temperature dependence of μ' of unilluminated sample is shown as the solid line in Fig. 3. 1. The figure shows also photoinduced permeability effect (*PE*) ; the permeability is decreased by the white light at 77 K. This reduction remains after illumination as far as the sample is kept at low temperatures, even after demagnetization. After the sample is heated above about 250 K, it shows no influence of illumination, even cooled again. To estimate the vanishing temperature of the photoinduced *DA* effect, the illuminated sample was warmed up from 77 K to a certain temperature before measurement. Figure 3. 2 shows the permeability of the sample cooled again from 180 K. There is no *DA* and around this temperature is vanishing point of photoinduced *DA*. But the value of μ' at 77 K is unchanged and the peak of μ'' remains by this procedure. So it indicates that YIG takes the middle state in the process of relaxation from the photoexcited state to the ground state.

3. 3 Discussion

We have modified the "two-center model" for YIG : Si proposed by Enz [1, 2] to explain *PE* in YIG : The electron trapped in the vacancy is excited by photon and transfer it to "normal site" far away from the vacancy, which decrease μ' by its large unisotropy. And the migration of the photoexcited Fe^{2+} or electron enhances μ''. On the other hand, something must move to stabilize the magnetic wall in order for *DA* to occur. Yanase [3] has explained *DA* in ferrite by the diffusion of cation vacancy, which is facilitated by Fe^{2+}. In our sample, however, cation vacancy is not expected. The candidates of the source of *DA* are Fe^{2+}, photoexcited electron from oxygen vacancy, and the oxygen vacancy itself. The activation energy of *DA* has not been obtained, because this *DA* comes from the exited state, and the relaxation of the state make the

temperature dependence of the relaxation time of *DA* complicated. The activation energy of μ'' peak is 0.1 eV, which is consistent with electron migration and rather large compare to the value of vacancies, for example 1 eV derived from *DA* of ferrite. So it is concluded that the migration of Fe^{2+} contributes to μ'', and oxygen vacancy is the origin of *DA*. Oxygen vacancy is taken as electrically positive and attracts O^{2-} ion. In unilluminated sample, however, it traps electron to be neutral. So it is natural to consider that photo-illumination increase mobility of oxygen vacancy to induce *DA* by removing the electron from oxygen vacancy. As *DA* and *PE* appear even in dark after illumination, excited electron is found to be unable to return to oxygen vacancies at low temperatures. Above about 250 K, the excited electrons can return to oxygen vacancies to show no trace of illumination. On this relaxation process, around 180 K, the sample takes the middle state, in which only *PE* appears. This suggests that the vacancy has a potential barrier and that electron stays at the neighbor of the vacancy. In this state, where the electron isn't trapped by the vacancy, the cancellation of the charge around the vacancy seems to depress the migration of vacancy. It is supported by the fact that the relaxation time of *DA* is increased by heating.

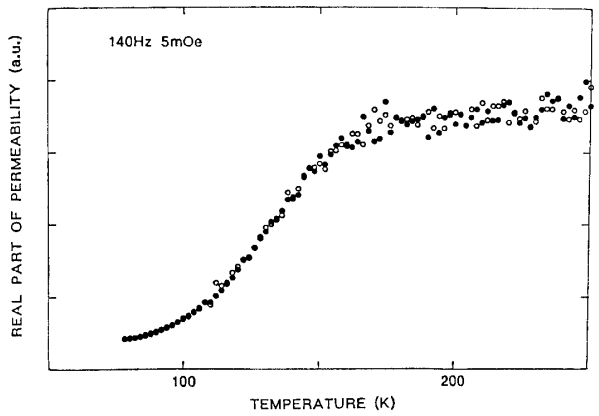

Fig. 3.1. Temperature dependence of μ' of YIG after illumination at 77K. The sample is demagnetized at each temperature ; open and filled circles indicate the permeability after 5 and 50 seconds, respectively. Solid line indicates μ' of the unilluminated sample.

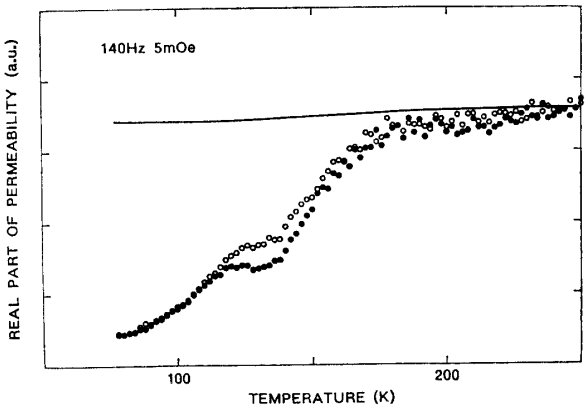

Fig. 3.2. Temperature dependence of μ' of YIG after cooled from 180K, which is illuminated at 77K previously. The meanings of open and filled circles are same as Fig. 3.1.

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Appendix I

The origin of *DA* is the pinning of a magnetic domain wall by the local anisotropy around a defect, i. e., a vacency or an impurity. If the defect migrate to change the local anisotropy, the magnetic moment in the domain wall might be stabilized. When the number of the pinning point increase, the permeability is decreased.

It is necessary, however, to change the anisotropy before the wall move away from the defect for the occurrence of the pinning. This condition is described as,

$$u < u_\tau = B/\tau \quad (B: \text{const.}) \qquad \text{(A 1)}$$

where u and τ are the velocity of the domain wall and the time constant of the change of the local anisotropy, respectively.

In the alternating field, the domains wall vibrate with a amplitude a, proportional to the permeability. So the displacement of the wall y is given as,

$$y=a \sin(\omega t) \qquad \text{(A 2)}$$

where ω is the angular frequency. And the velocity of the wall n is obtained as follows.

$$u=a\omega\cos(\omega t)=\omega(a^2-y^2)^{1/2} \qquad \text{(A 3)}$$

Figure A 1 shows the y dependence of u for several value of a, The region of y which satisfies the pinning condition (A 1) has the maximum at a_τ $(=u_\tau/\omega)$. On the assumption thet the region is proportional to the rate of the increasing of the number of the pinning point, u (or a) dependence of dn/dt is given as fig. A 2. This relation may be described approximately as,

$$\frac{dn}{dt}=\frac{A^2\mu}{A^2+\mu^2} \qquad \text{(A 4)}$$

where A is the constant proportional to $1/\tau$. And the relation,

$$n=k/\mu \quad (K\text{: const.}) \qquad \text{(A 5)}$$

is assumed: (A 4) becomes,

$$\frac{d\mu}{dt}=-\frac{kA^2\mu^3}{A^2+\mu^2} \qquad \text{(A 6)}$$

Solving (A 6),

$$\frac{1}{2}\mu^{-2}-A^2\log(\mu)=kt+C \quad (C\text{: const.}) \qquad \text{(A 7)}$$

is obtained.

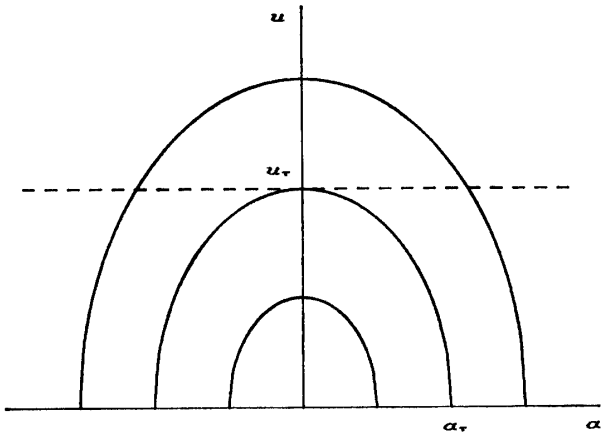

Fig. A1 The velocity of the magnetie domain wall as a function of the displacement for several amplitudes. The region below the dashed line satisfies the pinning condition.

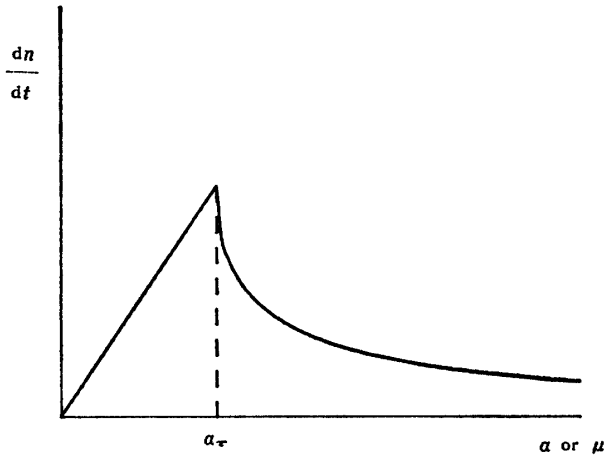

Fig. A2 Permeability or amplitude dependence of the rate of the increase of pinning points.