A Theory of the Uniaxial Ferromagnetic Anisotropy Induced by Magnetic Annealing in Cubic Solid Solutions

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Synopsis

According to Van Vleck, the cubic ferromagnetic anisotropy may originate from the interplay between the orbital valence and spin-orbit interaction, which results an apparent existence of the dipole-dipole coupling between spins. In a solid solution, the dipole-dipole coupling energy depends not only on the kind of an atom pair but also on the direction of spontaneous magnetization relative to the axis of the atom pair, and consequently it may yield an anisotropic equilibrium distribution of solute atom pairs at temperatures below the Curie temperature, which, in turn, may induce an additional ferromagnetic anisotropy having symmetry lower than cubic. Basing on this idea and using the same model as in Van Vleck's theory of cubic ferromagnetic anisotropy, we have calculated the uniaxial ferromagnetic anisotropy induced by magnetic annealing and obtained results, which agree well with available experimental data, especially as to its magnitude as dependent on the concentration of solute atoms and on the direction of magnetic field applied during annealing and its temperature dependence. Brief decussions in terms of the same idea are also given of the ferromagnetic behaviors of various alloys.

I. Introduction

Since Kelsall(1) first found that the presence of magnetic field during annealing resulted a large increase in maximum permeability in some nickel-iron alloys, many investigators have studied this effect of magnetic annealing on various alloys. It has been found that the hysteresis loop of permalloy, heat-treated with magnetic field and measured in the same direction as that of field applied during annealing, becomes very steep and almost rectangular, and the coercive force decreases conspicuously by magnetic annealing(2). When measured in the perpendicular direction, however, the hysteresis loop is very flat(2). It follows, then, that the material heat-treated in magnetic field is highly anisotropic. Such an anisotropic behavior may be explained by assuming that an uniaxial ferromagnetic anisotropy is induced by magnetic annealing and the direction of magnetic field present during annealing becomes the direction of easy magnetization, so that nearly all change in magnetization observed in this direction is due to the displacement of 180° domain walls. This interpretation has been evidenced by a large decrease in longitudinal magnetostriction by magnetic annealing(3) and by

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(1) G. A. Kelsall, Physics, **5** (1934), 169.

(2) J. F. Dillinger and R. M. Bozorth, Physics, **6** (1935), 279.

(3) S. Kaya, J. Fac. Sci. Hokkaidô Univ., **2** (1938), 29 ; H. J. Williams, R. M. Bozorth, and H. Christensen, Phys. Rev., **59** (1941), 1005 ; Y. Tomono, J. Phys. Soc. Japan, **4** (1948), 298.

a direct observation of the domain structure using powder pattern technique[4].

Now, what is the origin of such uniaxial ferromagnetic anisotropy induced by magnetic annealing? Bozorth *et al.*[5] concluded from their detailed investigations of this effect, especially as to the effective temperature range, with permalloys and perminvars, that the release of magnetostrictive consraint by plastic flow at high temperatures may be responsible for inducing the uniaxial anisotropy. But the objection to this explanation was offered by Becker and Döring[6] as follows :— First, a single crystal, heat-treated with magnetic field applied along a direction and high enough to saturate the crystal along this direction, is deformed magnetostrictively and homogeneously but is free from constraint, and after rotating the magnetization vectors to the perpendicular direction, it should be also free from constraint, so that the energy difference between these two directions can not be present, although actually the single crystal may show the induced uniaxial anisotropy[7]. Secondly, the observed initial permeabilities of various alloys annealed with transversal field are far smaller than those expected from the plastic deformation theory of Bozorth *et al.* This means that the induced anisotropy is far greater than that expected from the plastic flow due to the magnetostrictive constraint. Similar quantitative disagreement between the observed and theoretical magnitudes of this anisotropy has also been indicated by the experiments of Chikazumi[8].

Recently, Kaya[9] and his collaborators have proposed a quite different interpretation. They concluded, from their thorough investigation of the formation of superlattice Ni_3Fe and its influence on the magnetic properties, that the induced uniaxial anisotropy might result from the heterogenous formation of short-range order. Since the saturation magnetization and Curie temperature at ordered state of Ni_3Fe is higher than those at disordered state, the short-range order should develop in the form of prolate ellipsoid elongated in the direction of magnetic field applied during annealing, and thus an uniaxial anisotropy may be induced by the so-called "shape effect[10]", which has often been referred to the interpretation of the magnetic behavior of precipitation-type alloys. By this mechanism, they could have explained the observed order of magnitude of the induced anisotropy and its disappearence in the ordered state[11].

Although a great number of experimental investigation have been done on this effect of magnetic annealing, we have few available data connected essentially to

(4) H. J. Williams and M. Goertz, J. Appl. Phys., **23** (1952), 316.
(5) R. M. Bozorth, J. F. Dillinger, and G. A. Kelsall, Phys. Rev., **45** (1934), 742; R. M. Bozorth and J. F. Dillinger, Physics, **6** (1935), 285; R. M. Bozorth, Phys. Rev., **46** (1934), 742; R. M. Bozorth, Rev. Mod. Phys., **25** (1953), 42.
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(7) S. Chikazumi, cf. reference (9).
(8) S. Chikazumi, J. Phys. Soc. Japan, **5** (1950), 327.
(9) S. Kaya, Rev. Mod. Phys., **25** (1953), 49.
(10) L. Néel, Compt. rend., **225** (1947), 109; E. C. Stoner and E. P. Wohlfarth, Trans. Roy. Soc. (London), **A240** (1948), 599; C. Kittel, E. A. Nesbitt, and W. Shockley, Phys. Rev., **77** (1950), 739.
(11) J. L. Snoek and J. Smit, cited by G.W. Rathenau, Rev. Mod. Phys., **25** (1953), 55.

the induced anisotropy concerned, since almost all of them have dealt with only magnetic properties related undefinitely to this induced anisotropy, such as the maximum permeability. The magnetic annealing has been found effective for alloys which are believed to have no phase change such as the superlattice formation. A typical example is face-centered cubic nickel-cobalt alloys[12,13]. Yamamoto and the present author[13] have recently studied the perminvar character of some nickel-cobalt alloys and reached to the conclusion that the axis of the induced anisotropy should be able to rotate within a distance far shorter than the width of domain walls and its magnitude may be maximum at 50 percent cobalt. Taking into considerations of these facts as well as the experimental results obtained recently by Chikazumi[7,14,15] on single crystals of 76 permalloy and polycrystals of some permalloys, it seems quite difficult to explain completely the uniaxial anisotropy induced by magnetic annealing by either Bozorth's or Kaya's mechanism.

In this paper, the present author starts from a new idea that the energies of atom pairs of different kinds are different and depend on the orientation of spontaneous magnetization in the same manner as the energies of pairs of magnetic dipols do, so that the equilibrium distribution of solute atoms may be anisotropic at temperatures below the Curie temperature and it may be quenched to lower temperatures where the diffusion of atoms cannot occur, giving rise to an additional uniaxial ferromagnetic anisotropy, and he develops a theory that can interpret the experimental facts regarding the uniaxial anisotropy induced by magnetic annealing in ferromagnetic cubic solid solutions, such as its dependence upon the directions of field applied during heat-treatment, composition of alloys, and temperature of heat-treatment as well as its temperature dependence. Further, it will be shown that the magnetic properties of various alloys, especially the perminvar character of body-centered cubic solid solutions may be interpreted according to the same idea.

Quite recently and after the present investigation was completed, Néel[16] proposed a theory similar to ours, basing on the same idea, which will be discussed at the end of this paper.

II. Theory of the uniaxial ferromagnetic anisotropy induced by magnetic annealing

According to Van Vleck[17], the ferromagnetic anisotropy or the dependence of the magnetization on the crystallographic direction in cubic crystals results from the interplay between the orbital valence and spin-orbit interaction, which causes

(12) H. Masumoto *et al.*, Nippon Kinzoku Gakkai-shi, **17** (1953), 607, 612; Sci. Rep. RITU, **A6** (1954), 375.
(13) M. Yamamoto, S. Taniguchi, and K. Hoshi, Nippon Kinzoku Gakkai-shi, **17** (1953), 615; Sci. Rep. RITU, **A6** (1954), 539; M. Yamamoto and S. Taniguchi, to be published.
(14) S. Chikazumi, J. Phys. Soc. Japan, **5** (1950), 333.
(15) S. Chikazumi, Unpublished.
(16) L. Néel, J. de phys., **15** (1954), 225.
(17) J. H. Van Vleck, Phys. Rev., **52** (1937), 1178.

an apparent coupling between electron spins in the form of dipole-dipole and quadrupole-quadrupole coupling. The Hamiltonian used by Van Vleck is

$$\mathbf{H} = -g\beta H\sum_i S_{zi} + \sum_{j>i} w_{ij}\,. \tag{1}$$

The first term as the Hamiltonian of the unperturbed system is the ordinary exchange term expressed by the molecular field approximation, of which the total effective field, H, is supposed to be directed along the z axis. g, β, and S_{zi} in the first term are, respectively, Lande's factor, the Bohr magneton, and the z component of spin for atom i. The second term in Eq. (1) expresses the perturbing potential, in which w_{ij} is the interaction energy between atoms i and j.

The partition function, Z, of the system can be obtained by being expanded in the power series of $(kT)^{-1}$, as

$$Z/Z_0 = 1 - <\sum_{j>i} w_{ij}>_{\mathrm{AV}}/kT + <(\sum_{j>i} w_{ij})^2>_{\mathrm{AV}}/2k^2T^2 - \cdots, \tag{2}$$

where k is Boltzman's constant, T is the temperature, Z_0 is the partition function of the unperturbed system, and $<\ >_{\mathrm{AV}}$ denotes the quantum-mechanical average. Then the free energy, F, of the system is given by the relation

$$F = -kT\log Z = -kT\log Z_0 + <\sum_{j>i} w_{ij}>_{\mathrm{AV}} - <(\sum_{j>i} w_{ij})^2>_{\mathrm{AV}}/2kT + \cdots, \tag{3}$$

which shows that the anisotropy can be found if $<\sum_{j>i} w_{ij}>_{\mathrm{AV}}$, etc. are evaluated.

We neglect here the quadrupole-quadrupole coupling term in w_{ij}, because it may be forseen that the dipole-dipole coupling has a primary importance for the induced anisotropy of low order concerned, as in the magnetostriction and in the ferromagnetic anisotropy in hexagonal crystals. Then,

$$w_{ij} = \sum_{q,q'=x,y,z} a_{ij}^{qq'} S_{qi}S_{q'j}\,, \tag{4}$$

where $a_{ij}^{qq'}(=a_{ij}^{q'q})$ is the qq' component of the coupling constant a_{ij} referred to the coordinate system (x,y,z), and S_{qi} and $S_{q'j}$ are, respectively, the q component of the spin of atom i and the q' component of the spin of atom j, and

$$<w_{ij}>_{\mathrm{AV}} = \sum_{q,q'} a_{ij}^{qq'} <S_{qi}S_{q'j}>_{\mathrm{AV}} = a_{ij}^{zz}B_1^2\,, \tag{5}$$

where

$$B_1 = SB(\theta) = \{(2S+1)/2\}\coth[\{(2S+1)/2\}\theta] - (1/2)\coth[(1/2)\theta], \tag{6}$$

$$\theta = g\beta H/kT\,, \tag{7}$$

S is the spin quantum number of atoms, which is supposed here to be the same for all atoms, and $B(\theta)$ is the Brillouin function.

Now, to obtain the anisotropy, it is necessary to express the interaction energy in terms of the coupling constants A_{ij}'s for the coordinate system fixed relative to the crystal, which is denoted by capital letters (X, Y, Z). The transformation relation between a_{ij}'s and A_{ij}'s are

$$a_{ij}^{qq'} = \sum_{p,p'=X,Y,Z} \lambda_{qp}\lambda_{q'p'} A_{ij}^{pp'}, \tag{8}$$

where λ_{qp} is the cosine of the angle between the q and p axes. Further, because of the dipole-dipole coupling, $A_{ij}^{pp'}$ may be given by

$$A_{ij}^{pp'} = C_{ij}(\delta_{pp'} - 3n_{ij}^{p}n_{ij}^{p'}), \tag{9}$$

where C_{ij} is the coupling constant, $\delta_{pp'}$ is Kronecker's delta and n_{ij}^{p} is the cosine of the angle between the direction of the atom pair (i,j) and that of the p axis. Then, from Eq. (5) it follows that

$$\left.\begin{aligned} &<\sum_{j>i} w_{ij}>_{\mathrm{AV}} = (N/2)B_1^2 \sum_j a_{ij}^{zz} \\ &= (N/2)B_1^2(\alpha_1^2 \sum_j A_{ij}^{XX} + \alpha_2^2 \sum_j A_{ij}^{YY} + \alpha_3^2 \sum_j A_{ij}^{ZZ} + 2\alpha_1\alpha_2 \sum_j A_{ij}^{XY} \\ &\quad + 2\alpha_2\alpha_3 \sum_j A_{ij}^{YZ} + 2\alpha_3\alpha_1 \sum_j A_{ij}^{ZX}). \end{aligned}\right\} \tag{10}$$

In this expression, α_1, α_2, and α_3 have been used in place of λ_{zX}, λ_{zY}, and λ_{zZ}, respectively, and we have replaced $\sum_{j<i}$ by $(N/2)\sum_j$ (N = number of atoms per unit volume), since atoms are all alike in behavior in the cubic lattice concerned here. In cubic crystals, sums like $\sum_j A_{ij}^{XY}$ in which a letter in superscript (here X or Y) appears only once vanish and

$$\sum_j A_{ij}^{XX} = \sum_j A_{ij}^{YY} = \sum_j A_{ij}^{ZZ} = (1/3)\sum_j (A_{ij}^{XX} + A_{ij}^{YY} + A_{ij}^{ZZ}) = \Omega_0. \tag{11}$$

Accordingly, Eq. (10) becomes

$$<\sum_{j<i} w_{ij}>_{\mathrm{AV}} = (N/2)B_1^2\Omega_0, \tag{12}$$

which is independent on the direction of spontaneous magnetization. Hence, the first-order effect of dipole-dipole coupling does not contribute to the ferromagnetic anisotropy in cubic crystals. The second-order effect, however, does contribute to the ferromagnetic anisotropy in the expected form, as shown by Van Vleck[17].

So far, the ferromagnetic anisotropy of cubic crystals composed of like atoms have been considered. We now consider cubic solid solutions of the substitutional type, composed of two kinds of atoms A and B, restricting our attention only within the nearest neighbor atom pairs, since the energy of the dipole-dipole coupling as a consequence of the interplay between the orbital valence and spin-orbit interaction decreases more rapidly than the inverse three power with an increase of the distance between atoms. Furthermore, in order to see an approximate behavior of the induced anisotropy, we may assume that the spin quantum numbers are alike for all the constituent atoms as in the case of cubic metal lattice, but that only the coupling constants C_{ij}'s in Eq. (9) differ for different kinds of nearest-neighbor atom pairs, so that the coupling constans of nearest-neighbor atom pairs A–A, B–B, and A–B are C_{AA}, C_{BB}, and C_{AB}, respectively. In the absence of an isotropic interaction among atoms (such as that associated with the superlattice formation or precipitation) other than that of magnetic

origin, we may suppose that, if the concentration of B atoms is $n(\ll 1)$ and the total number of nearest-neighbor atoms is z, the nearest-neighbor atoms around a given atom are nz of B atoms and $(1-n)z$ of A atoms. Since, in the nearest-neighbor assumption, only the distribution of B atoms in the neighbors of a given B atom may be responsible for the induced anisotropy, we may replace $(N/2)\sum_j$ in the previous equations by $(N/2)n\sum_j'$, where $\sum_j'$ denotes the summation over all nearest neighbors of a given B atom.

The diffusion of atoms takes place at high temperatures below the Curie temperature, and an increase by unity in the number of B–B atom pairs in one of the nearest-neighbor directions through any interchange between A and B atoms results an increase by unity in the number of A–A atom pairs and a decrease by two in the number of A–B atom pairs in the same direction. Then the change in the energy of the dipole-dipole coupling caused by an interchange between an A atom and a B atom occuring at the temperature T' may be expressed, from Eqs. (9), (8) and (5), as $CB_1^2(T')(1-3\cos^2\phi)$, where $C=C_{AA}+C_{BB}-2C_{AB}$ and ϕ is the angle between the direction of spontaneous magnetization and the direction of the B–B atom pair considered, and thus the probability that an B–B atom pair takes the direction specified by the angle ϕ in the equilibrium state, $\omega(\phi)$, is given approximately by

$$\omega(\phi) = (1/z)\exp\{-CB_1^2(T')(1-3\cos^2\phi)/kT'\}, \tag{13}$$

because the energy of the dipole-dipole coupling is far smaller than the thermal energy so that the deviation from isotropic distribution is very small. This means that the equilibrium distribution of solute atom pairs becomes anisotropic below the Curie temperature. Then, the results obtained above for pure metals, which may be applicable for solid solutions having the isotropic distribution of solute atom pairs, does not hold for this case, and the first-order effect of dipole-dipole coupling gives rise to an ferromagnetic anisotropy in the form as;

$$F = F_0+\alpha_1^2\Omega^{XX}+\alpha_2^2\Omega^{YY}+\alpha_3^2\Omega^{ZZ}+2\alpha_1\alpha_2\Omega^{XY}+2\alpha_2\alpha_3\Omega^{YZ}+2\alpha_3\alpha_1\Omega^{ZX}, \tag{14}$$

where $$F_0 = -kT\log Z_0$$

and $$\Omega^{pp'}=(N/2)nB_1^2(T)\sum_j' A_{ij}^{pp'}. \tag{15}$$

Since $A_{ij}^{pp'}$ in this case is given by

$$A_{ij}^{pp'}=zn\omega(\phi)C_{ij}(\delta_{pp'}-3n_{ij}^p n_{ij}^{p'}), \tag{16}$$

$\Omega^{pp'}$ can be calculated by expanding the exponential in $\omega(\phi)$ (Eq. (13)) with respect to $(kT')^{-1}$ since $|C|/kT'\ll 1$ and by retaining only the first-order term, and further by using C instead of C_{ij} in Eq. (16) for the same reason as for Eq. (13), and it is expressed as

$$\Omega^{pp'}=(N/2)n^2CB_1^2(T)\sum_j'[1-CB_1^2(T')\{1-3(n_{ij}^X)^2\}/kT'](\delta_{pp'}-3n_{ij}^p n_{ij}^{p'}),$$

where the direction of field applied during annealing is taken as the X axis[18].

(18) Taking directly the crystal axes as the coordinate axes, we can express the induced anisotropy in a simpler form, which will be shown in a later paper.

Further, as the terms such as $\sum_j'(\delta_{pp'}-3n_{ij}^p n_{ij}^{p'})$ vanish, it becomes that

$$\Omega^{pp'} = -\{(9/2)Nn^2C^2B_1^2(T)B_1^2(T')/kT'\}\sum_j'\{(n_{ij}^X)^2 n_{ij}^p n_{ij}^{p'}-(1/3)n_{ij}^p n_{ij}^{p'}\}. \quad (17)$$

Since the nearest-neighbor directions are twelve <110> directions for face-centered cubic lattice, eight <111> directions for body-centered cubic lattice, and six <100> directions for simple cubic lattice, values of $\sum_j'(n_{ij}^X)^2 n_{ij}^p n_{ij}^{p'}$ and of $\sum_j' n_{ij}^p n_{ij}^{p'}$ are as given in Table 1, where $(\beta_1, \beta_2, \beta_3)$, $(\beta_1', \beta_2', \beta_3')$ and $(\beta_1'', \beta_2'', \beta_3'')$ are the

Table 1. Values of $\sum_j'(n_{ij}^X)^2 n_{ij}^p n_{ij}^{p'}$ and $\sum_j' n_{ij}^p n_{ij}^{p'}$ for various cubic lattices.

(i) $\sum_j'(n_{ij}^X)^2 n_{ij}^p n_{ij}^{p'}$

p	p'	f. c. c.	b. c. c.	s. c.
X	X	$2\{1+(\beta_1^2\beta_2^2+\beta_2^2\beta_3^2+\beta_3^2\beta_1^2)\}$	$(32/9)\{(1/4)+(\beta_1^2\beta_2^2+\beta_2^2\beta_3^2+\beta_3^2\beta_1^2)\}$	$4\{(1/2)-(\beta_1^2\beta_2^2+\beta_2^2\beta_3^2+\beta_3^2\beta_1^2)\}$
Y	Y	$1-(\beta_1^2\beta_1'^2+\beta_2^2\beta_2'^2+\beta_3^2\beta_3'^2)$	$(16/9)\{(1/2)-(\beta_1^2\beta_1'^2+\beta_2^2\beta_2'^2+\beta_3^2\beta_3'^2)\}$	$2(\beta_1^2\beta_1'^2+\beta_2^2\beta_2'^2+\beta_3^2\beta_3'^2)$
Z	Z	$1-(\beta_1^2\beta_1''^2+\beta_2^2\beta_2''^2+\beta_3^2\beta_3''^2)$	$(16/9)\{(1/2)-(\beta_1^2\beta_1''^2+\beta_2^2\beta_2''^2+\beta_3^2\beta_3''^2)\}$	$2(\beta_1^2\beta_1''^2+\beta_2^2\beta_2''^2+\beta_3^2\beta_3''^2)$
X	Y	$-(\beta_1^3\beta_1'+\beta_2^3\beta_2'+\beta_3^3\beta_3')$	$-(16/9)(\beta_1^3\beta_1'+\beta_2^3\beta_2'+\beta_3^3\beta_3')$	$2(\beta_1^3\beta_1'+\beta_2^3\beta_2'+\beta_3^3\beta_3')$
Y	Z	$-(\beta_1^2\beta_1'\beta_1''+\beta_2^2\beta_2'\beta_2''+\beta_3^2\beta_3'\beta_3'')$	$-(16/9)(\beta_1^2\beta_1'\beta_1''+\beta_2^2\beta_2'\beta_2''+\beta_3^2\beta_3'\beta_3'')$	$2(\beta_1^2\beta_1'\beta_1''+\beta_2^2\beta_2'\beta_2''+\beta_3^2\beta_3'\beta_3'')$
Z	X	$-(\beta_1^3\beta_1''+\beta_2^3\beta_2''+\beta_3^3\beta_3'')$	$-(16/9)(\beta_1^3\beta_1''+\beta_2^3\beta_2''+\beta_3^3\beta_3'')$	$2(\beta_1^3\beta_1''+\beta_2^3\beta_2''+\beta_3^3\beta_3'')$

(ii) $\sum_j' n_{ij}^p n_{ij}^{p'}$

	f. c. c.	b. c. c.	s. c.
$p=p'$	4	8/3	2
$p\neq p'$	0	0	0

direction cosines of X, Y, and Z axes, respectively, referred to the crystal axes. Then, we have

$$\left.\begin{aligned}
\Omega^{XX} &= -ANn^2C^2B_1^2(T)B_1^2(T')\{(\beta_1^2\beta_2^2+\beta_2^2\beta_3^2+\beta_3^2\beta_1^2)+B\}/kT',\\
\Omega^{YY} &= (A/2)Nn^2C^2B_1^2(T)B_1^2(T')\{(\beta_1^2\beta_1'^2+\beta_2^2\beta_2'^2+\beta_3^2\beta_3'^2)+B\}/kT',\\
\Omega^{ZZ} &= (A/2)Nn^2C^2B_1^2(T)B_1^2(T')\{(\beta_1^2\beta_1''^2+\beta_2^2\beta_2''^2+\beta_3^2\beta_3''^2)+B\}/kT',\\
\Omega^{XY} &= (A/2)Nn^2C^2B_1^2(T)B_1^2(T')(\beta_1^3\beta_1'+\beta_2^3\beta_2'+\beta_3^3\beta_3')/kT',\\
\Omega^{YZ} &= (A/2)Nn^2C^2B_1^2(T)B_1^2(T')(\beta_1^2\beta_1'\beta_1''+\beta_2^2\beta_2'\beta_2''+\beta_3^2\beta_3'\beta_3'')/kT',\\
\text{and}\\
\Omega^{ZX} &= (A/2)Nn^2C^2B_1^2(T)B_1^2(T')(\beta_1^3\beta_1''+\beta_2^3\beta_2''+\beta_3^3\beta_3'')/kT',
\end{aligned}\right\} \quad (18)$$

where $A=9$, 16, and -8 and $B=1/3$, 0, and $-1/3$ for face-centered, body-centered, and simple cubic lattices, respectively. It is to be noted that the simple cubic lattice is equivalent to the interstitial solid solution of body-centered cubic lattice.

Now, we can calculate the anisotropy induced by annealing in magnetic field applied along any direction using Eqs. (14) and (18). For a particular case of the induced anisotropy in a plane (X, Y) containing the direction of magnetic field applied during annealing, F may be written as:

$$F = F_0 - \Omega_1 \cos^2\theta + \Omega_2 \cos\theta \sin\theta , \tag{19}$$

where $$\Omega_1 = \Omega^{YY} - \Omega^{XX} \text{ and } \Omega_2 = 2\Omega^{XY}, \tag{20}$$

and θ is the angle between the magnetization vector and the direction of magnetic annealing in this plane. Further, for the induced anisotropy in the $(1\bar{1}0)$ or (001) plane as the (X, Y) plane in which the X axis coincides with the [111], [110], or [001] direction, F may take commonly the following form;

$$F = F_0 - \Omega_1 \cos^2\theta , \tag{21}$$

where $$\Omega_1 = A' N n^2 C^2 B^2{}_1(T) B_1{}^2(T')/kT', \tag{22}$$

and A' is the numerical constant, of which values for various cubic lattices are as given in Table 2.

Table 2. Values of A' for various cubic lattices.

(X, Y) plane	X-axis	f. c. c.	b. c. c.	s. c.
$(1\bar{1}0)$	[111]	9	8	0
	[110]	27/4	4	9/2
	[001]	9/2	0	9
(001)	[110]	9	8	0
	[100]	9/2	0	9

III. Comparison with available experimental data

In the following, we compare our theory with available experimental results. In the first place, we estimate the order of the induced anisotropy. The anisotropy constant measured in the $(1\bar{1}0)$ plane of a face-centered cubic solid solution as heat-treated in magnetic field directed along the [111] direction is given, from Eqs. (21) and (22) and Table 2, by

$$\Omega_1 = 9 N n^2 C^2 B_1{}^2(T) B_1{}^2(T')/kT'. \tag{23}$$

If we put $T' = 8 \times 10^2$°K, $n = 1/4$, $B_1{}^2(T) = 1$, $B_1{}^2(T') = 1/10$, and $|C| = |C_{AA} + C_{BB} - 2C_{AB}|$ $= 10^{-15} \sim 10^{-16}$ ergs[17] in this equation, Ω_1 becomes $10^3 \sim 10^5$ergs/cm^3. Chikazumi's[7] measured data of the induced anisotropy in the $(1\bar{1}0)$ plane of a 76 permalloy single crystal (Fig. 1) shows that the induced anisotropy is uniaxial and its magnitude is about 10^3ergs/cm^3, in accordance with the theory. It is to be noted, however, that Chikazumi's experimental results can not directly be compared with the theory, since the corresponding equilibrium temperature is uncertain because of the finite cooling velocity of about 22°C/min employed by Chikazumi. The adoption of such a relatively high cooling velocity may be inevitable for preventing the formation of superlattice Ni_3Fe, of which the ordering energy is so far greater than the dipole-dipole coupling energy that the decrease in the number

of solute atom pairs due to ordering may predominate over their anisotropic distribution and correspondingly the induced anisotropy may decrease with ordering.

Eq. (21) shows that the induced anisotropy in the $(1\bar{1}0)$ plane has the form $-\Omega_1\cos^2\theta$, when the direction of field applied during annealing is in the [111], [110], or [001] direction, which means that the direction of easy magnetization of the induced anisotropy should be always the direction of field applied during annealing in these cases. This is in a complete agreement with the above-cited experimental results obtained by Chikazumi[7] (c.f. Fig. 1). Further, Eq. (22) and Table 2 indicate that the ratio of Ω_1 values (equal to the ratio of A' values) in these cases should be 1 : 0.75 : 0.5 for face-centered cubic solid solutions, and the experimental ratio is 1 : 0.56 : 0.36 according to the observation of Chikazumi[7]. It may be said, taking into considerations of the difficulty in the experiment and of the approximation in the theory, that both ratios are in a good agreement with each other.

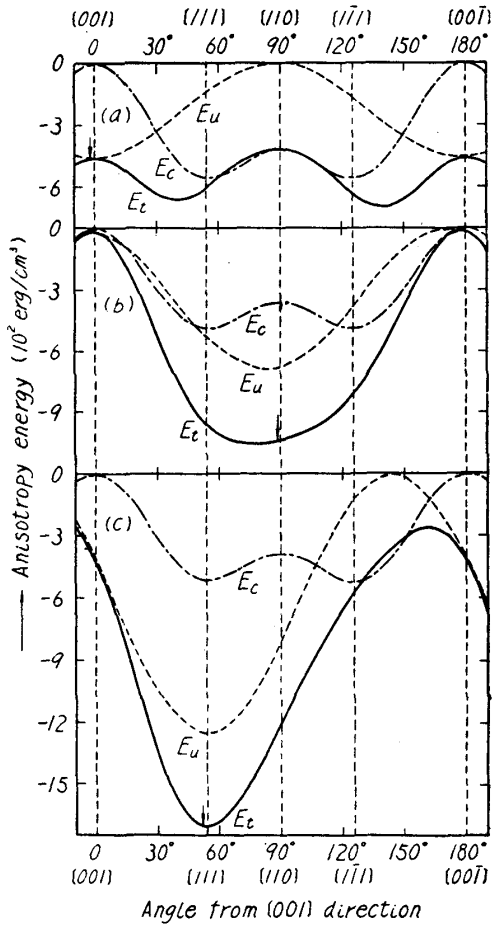

Fig. 1. Dependence of the anisotropy energy of a 76 permalloy crystal on the orientation in the $(1\bar{1}0)$ plane. Arrows indicate the orientations along which the external magnetic field was applied during cooling (Cooling velosity = 22°C/min). The total energy, E_t, is analyzed into E_c and E_u; the former has the cubic symmetry, and the latter is the uniaxial anisotropy. (Reproduced from Fig. 8 of Kaya's[9] paper.)

According to Eqs. (19) and (20), the direction of easy magnetization of the induced anisotropy does not coincide with the direction of field applied during annealing even in the $(1\bar{1}0)$ plane when the field direction is not along one of the principal crystal directions. For example,

Table 3. The extent of deviation, $\Delta\phi$, of the direction of easy magnetization of the induced uniaxial anisotropy from the direction of magnetic field applied along an arbitrary direction, ϕ, in the $(1\bar{1}0)$ plane during annealing towards the [111] direction in face-centered and body-centered cubic solid solutions.

ϕ measured from [001] direction	$\Delta\phi$	
	f. c. c.	b. c. c.
30°0′	7°55′	19°6′
70°0′	3°24′	7°45′

when the field direction lies along an arbitrary direction in the $(1\bar{1}0)$ plane, the direction of easy magnetization deviates from that direction towards the [111] direction, and the extent of this deviation, $\Delta\phi$, for face-centered and body-centered cubic solid solutions is as shown in Table 3. Chikazumi[15] found experimentally for a 76 permalloy single crystal that this deviation was 13°36′ when the field during annealing was applied along an direction inclined by 30° to the [001] direction. This experimental value agrees, in the degree of order and in the sign, with the theoretical prediction.

Eq. (17) or (18) indicates that the induced anisotropy varies with temperature as $B_1^2(T)$. If the saturation magnetization at temperatures T and 0°K are denoted respectively by I_T and I_0, then $B_1(T) \fallingdotseq SI_T/I_0$ for the approximation adopted in our theory. Then, the indueed anisotropy should be proportional to I_T^2. Fig. 2 shows the experimentally found and the theoretically expected temperature dependence of the induced anisotropy. The experimental curve is that obtained by Chikazumi[14] from the magnetization curves of a 78.5 permalloy polycrystal heat-treated in circular magnetic field, while the theoretical curve which expresses the variation with temperature of the square of the saturation magnetization in an arbitrary scale, is that plotted by the present author from Chikazumi's data.

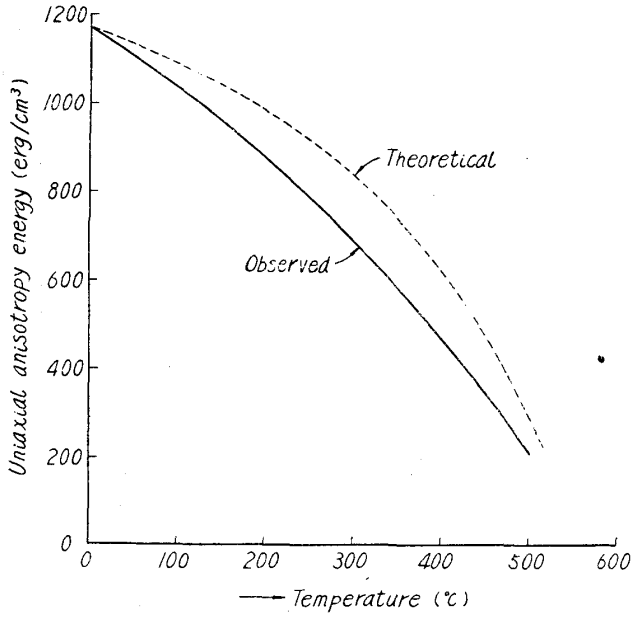

Fig. 2. Temperature dependence of the induced uniaxial anisotropy of 78.5 permalloy as compared with the theoretical one.

As may also be seen from Eq. (17) or (18), the induced anisotropy may increase with concentration of solute atoms as n^2 at low concentrations, and it may be expected to reach to a maximum at 50 percent of solute concentration. This concentration dependence agree well with the experimental evidence obtained by us with annealed nickel-cobalt alloys[13] and obtained by Chikazumi[15] with permaloy polycrystals. Further, the induced anisotropy should vary with annealing temperature as $B_1^2(T')/kT'$, but the experimental evidence supporting this conclusion is not available.

Thus, we have shown that our theory can interpret well the available experimental data, in spite of simplifying assumptions and approximations used, and it may be said that the induced anisotropy is of the magnetocrystalline origin and it occurs for any anisotropic distribution of solute atom pairs, superposed on the proper cubic anisotropy. In view of few experimental results now available, it is desirable to check the theory for various lattice structures particulary in equilibrium

conditions.

IV. Comparison with Néel's theory

As noted before, Néel[16] has recently and independently derived nearly the same results for the induced anisotropy, basing on the same idea as ours. The main differences between Néel's and our theories are as follows.

In Néel's theory, the coupling energy of an atom pair is expanded by Legendre's polynomial, the first term of which corresponds to the dipole-dipole coupling. Referring to that the purely magnetic dipole-dipole coupling is too small to lead the observed magnetostriction, a dipole-dipole coupling caused by another origin, probably by the spin-orbit coupling, is introduced as parameters. This rather phenomenological introduction of the dipole-dipole coupling responsible for the induced anisotropy has made its temperature dependence obscure. Therefore, the temperature dependence of the induced anisotropy and its magnitude as dependent on the annealing temperature is not included in Néel's theory. However, Néel has been able to estimate the coefficients of dipole-dipole coupling for various kinds of atom pairs in any solid solution by calculating the magnetostriction constants and by comparing them with the experimental data, and has shown that C in our Eq. (17) or (18) certainly has the value of about $10^{-15}\sim10^{-16}$ ergs for iron-nickel and iron-cobalt alloys in accordance with our assumption. Furthermore, he has calculated the induced anisotropy without the restrictions of diluteness and ideality of solutions[19].

With the exception of these two main differences, Néel's results are in agreement with ours, although the different coordinate system has been adopted by him for expressing the induced anisotropy.

V. Applications of the theory

We note here some conclusions derived from our theory. When the ferromagnetic solid solutions are cooled down from high temperatures beyond the Curie temperature without appling a magnetic field, the induced anisotropy is produced along the directions of magnetization vectors even in domain walls as well as in the interior of domains. Thus domain walls are stabilized by the induced anisotropy, and then the wall displacements becomes far more difficult than when the stabilization is absent, namely, in quenched state. Hence, the permeability of solid solution generally increases by quenching.

Further, the stabilization of domain walls by the induced anisotropy leads to the perminvar character. As seen from Eqs. (21) and (22) and Table 2, body-centered cubic solid solutions show no induced anisotropy along the [100] direction. Hence, when the cubic anisotropy is positive, they can not be expected to show the perminvar character. This is the reason why iron-cobalt alloys containing less than about 50 percent cobalt is not included in the perminvar region, while the

(19) Recently, Iwata of our laboratory has studied the statistical theory in Bethe's approximation of anisotropic distribution of atom pairs in solid solutions having both isotropic and anisotropic interactions using Takagi's method and has obtained the results different from Néel's.

alloys containing more than 50 percent cobalt is included.

When the magnetic field is applied at high temperatures below the Curie temperature and where the diffusion of atoms can take place, the redistribution of atoms appropriate to the new domain structure occurs. This induces the magnetic after-effect. The observations by Kühlwein[20] and Fahlenbrach[21] on iron-nickel and iron-cobalt alloys may be explained partly by this mechanism.

The detailed consideration of this section will be shortly given in a subsequent paper.

Summary

A new idea about the origin of the uniaxial anisotropy induced by magnetic annealing in ferromagnetic cubic solid solutions has been proposed based on the anisotropic coupling between atom pairs, which results from the interplay between the spin-orbit coupling and orbital valence as in the theory of cubic anisotropy by Van Vleck. This anisotropic coupling may be described approximately in the form of dipole-dipole and quadrupole-quadrupole coupling. Of these, the dipole-dipole coupling cannot yield any anisotropy for cubic metals and solid solutions having isotropic distribution of atom pairs. The presence of dipole-dipole coupling, however, makes ferromagnetic solid solution to have an anisotropic equilibrium distribution of atom pairs below the Curie temperature, and the anisotropy having lower symmetry than cubic is induced.

The results of calculation made basing on the new idea are as follows:

(i) The anisotropy induced in this way is uniaxial and the direction of easy magnetization does not always coincide with the direction of magnetic field, high enough to saturation, applied during annealing.

(ii) Its magnitude depends on the direction of magnetic field applied during annealing, and on the annealing temperature, T, as I_T^2/T, where I_T is the spontaneous magnetization at T.

(iii) It increases in proportion to the square of the concentration of solute atoms in low concentration range, if the solid solution is ideal.

(iv) It depends on the temperature as the square of spontaneous magnetization does.

(v) Its maximum value has been estimated to be about $10^3 \sim 10^5 \mathrm{ergs/cm^3}$.

It has been shown that these theoretical results agree well with the available experimental data, and thus the uniaxial ferromagnetic anisotropy induced by magnetic annealing in ferromagnetic solid solutions may be considered as the magnetocrystalline anisotropy appeared in consequence of any anisotropic distribution of atom pairs in solid solutions.

As applications of the theory, brief discussions have been given on the stabilization of domain walls by this uniaxial anisotropy in ferromagnetic cubic solid solutions annealed without magnetic field, which may result the so-called perminvar-

(20) H. Kühlwein, Phys. Zeits., **32** (1933), 860.
(21) H. Fahlenbrach, Ann. Phys., [6] **2** (1948), 355.

type magnetic properties and on the magnetic after-effect in ferromagnetic cubic solid solutions at high temperatures.

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