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Exchange striction, the origin of polycrystalline magnetoelastic anisotropy

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It is analytically shown that the origin of the magnetoelastic anisotropy term ΔK, previously established as specific of the polycrystalline state, is related to the product of the strong isotropic virtual "exchange stresses" due to the exchange striction by the anisotropic magnetostrictive deformation constrained by the neighboring grains. A linear law of the variation of ΔK versus the exchange-striction volume anomaly $\Delta V/V$ is obtained for $NiFe_2O_4$ and yttrium iron garnet. An analytical expression of $\Delta K(T)$ as a function of ionic structure parameters is established.

This work presents the results of a confrontation between ideas and data obtained in two fields of investigation first, in the field of the total *anisotropy K* specific of polycrystals, and secondly, in the field of the *isotropic* volume anomaly due to the exchange striction. It is obviously necessary to recall results obtained in ferrimagnetic ceramics in the fields of the total anisotropy and volume anomaly.

I. HISTORICAL BACKGROUND

A. Specific polycrystalline total anisotropy K

1. History

The concept of an effective anisotropy field was introduced by several authors, in order to interpret the value of the natural spin-resonance frequency within magnetic domains, i.e., the resonance in the spontaneous internal anisotropy field found in the initial susceptibility spectra and excited in toroidal samples, without any external aligning field.

In 1950, Rado, Wright, and Emerson found two dispersions in a frequency spectrum in a magnesium ferrite sample; they attributed the first dispersion (in rf range) to the domain-wall motion and the second one at higher frequencies to the natural spin resonance in domains of the grains.[1]

For technological reasons, explained recently,[2] some authors, like Rado *et al.*, found two dispersions (or more) in frequency spectra,[3–6] others, only one.[7,8] The results of these investigations led to many calculations and intepretations, often complicated and contradictory; some authors put forward the role of the magnetocrystalline anisotropy energy K_1, others the magnetoelastic energy related to technological stresses, others the dipolar energy related to demagnetizing fields due to the porosity of the investigated samples.

Miles, Westphal, and von Hippel, found two dispersions in NiZn ferrite samples of different compositions, and analyzed the existing situation in the investigation of frequency spectra; they found that every author could speak about his "own" sample and they pointed out that from theoretical considerations, "the whole calculation rests on shaky grounds."[3]

An investigation of the frequency spectra of series of samples of the same composition, each of them sintered at a different temperature, permitted to Globus to establish the existence of a kind of anisotropy which is specific to the polycrystalline state and is related only to the intrinsic magnetic properties. Figure 1 shows schematically the results obtained for one of these compositions and the conclusions of this study.[9,10] The spectra of a series of $NiFe_2O_4$ samples sintered at temperatures from 1150 to 1400 °C (curves 1, 2, 3) present two well-separated dispersions: the first one with a variation of amplitude and frequency and the second one with the *same natural spin-resonance frequency for all samples. The spectrum "m"* corresponds to a sample with very small grains, supposed to be a "monodomain" grain sample in which each grain is a single domain.

The fact that the frequency of the natural spin resonance (NSR) is the same for all samples in spite of different granular structure parameters like grain sizes, porosity, or eventual technologic stresses led to the evident conclusion that the spontaneous anisotropy field H_K is the same in all samples and is related only to the material composition. The results, which were confirmed later for other compositions and temperatures, put an end to the previously described confusion in this field of investigation.

The spontaneous anisotropy K, specific to the polycrystalline state, was called later "total anisotropy" in order to discriminate it from the hazardous "effective anisotropy." It became one of the main parameters of magnetization process, which permitted establishment of the universal hysteresis loop.[11]

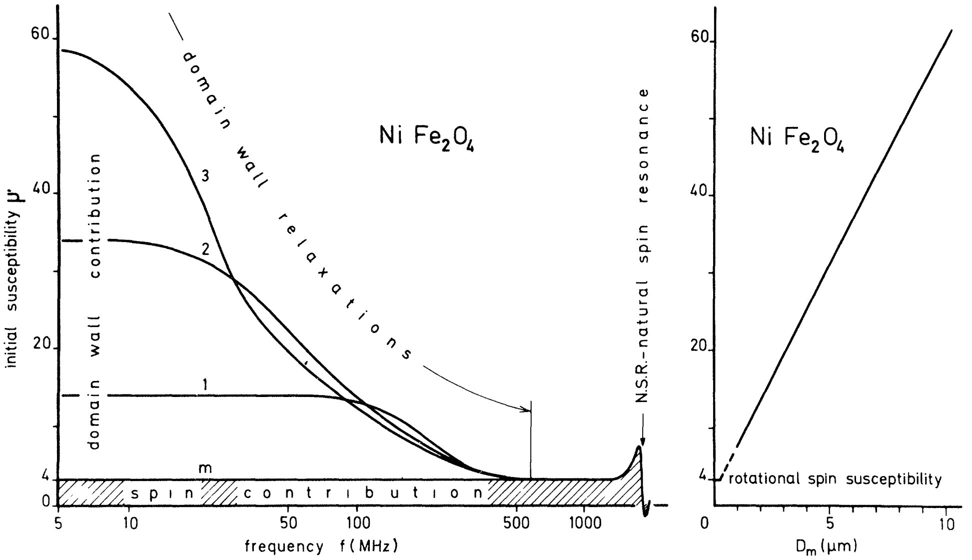


FIG. 1. Schematic representation of frequency and grain-size dependences of the initial susceptibility real part μ'. (left) Initial susceptibility μ' frequency spectra of $NiFe_2O_4$ samples, showing the same frequency of natural spin resonances (NSR) in samples of different grain diameters D_m and different porosities p. Curve 1. $D_m = 2$ μm, $p = 0.21$. Curve 2. $D_m = 5$ μm, $p = 0.05$. Curve 3. $D_m = 11$ μm, $p = 0.05$. Curve *m*. $D_m < 0.2$ μm, $p = 0.32$. (right) Extrapolation of the quasi-static initial susceptibility μ' of polydomain $NiFe_2O_4$ samples as a function of the grain diameter D_m to small values of D_m ($\simeq 0.2$ μm). The straight line shows schematically [but from numerous experimental results (Refs. 9 and 10)] the grain-size dependence of the intital susceptibility μ'.

2. *Determination of the value of K*

As can be seen in Fig. 1 (left and right) the value of the total anisotropy K can be deduced in four different ways.

a. From NSR frequency by using the Larmor relation

$$\omega = 2\pi f = \gamma H_K \tag{1}$$

and the expression of the anisotropy field in a cubic material with $K < 0$

$$H_K = \frac{4}{3}\frac{K}{M_S} , \tag{2}$$

where H_K is the total anisotropy field (also called previously H_{eff}), f is the NSR frequency, γ is the gyromagnetic ratio, and M_S is the spontaneous magnetization.

b. From the value of the initial susceptibility in frequency spectra after the first dispersion due to the domain-wall motion, and before the NSR, by using the Snoek formula for rotational spin susceptibility[12]

$$K = \frac{2\pi M_S^2}{(\mu - 1)_c} , \tag{3}$$

where $(\mu - 1)_c$ is rf initial susceptibility.

c. From the value of the quasistatic rotational susceptibility (at low frequencies), of a "monodomain grain" sample by using the same formula (3).

d. From the extrapolated value of the quasistatic initial susceptibility of polydomain samples as a function of the mean grain diameter D_m, where for small values of D_m ($\simeq 0.2$ μm) the extrapolated value corresponds only to the rotational spin susceptibility as for the "monodomain" sample [Fig. 1 (right)].

In all cases when the two dispersions are well separated, the values obtained in these four ways are in good agreement. It is sure that the best, but most difficult way is the NSR detection.

3. *Nature of K*

The anisotropy K was larger than K_1 (the single-crystal crystalline anisotropy constant for cubic symmetry) over the whole temperature range and especially at Curie temperature where K_1 becomes negligible. It was possible to relate the supplementary term $\Delta K = K - K_1$ either to the dipolar anisotropy energy, or to the magnetoelastic anisotropy energy, or to both. It was shown that the dipolar energy, which should be related to M_s^2, is small in comparison with ΔK; some "monodomain" samples with large M_s showed a very small ΔK.[13] This fact was confirmed more recently by the value of the NSR frequency of some garnets $Y_{3-x}Gd_xFe_5O_{12}$ in which, at room temperature, $4\pi M_s$ varies with x in a very large range and where the ΔK variation is very small.[14] So it was deduced that the dipolar energy cannot be at the origin of ΔK and the nature of ΔK was attributed to a term of magnetoelastic energy.[13]

4. Hypothesis about the origin of the term ΔK

The magnetoelastic term ΔK should be related to the magnetostriction and stresses. These latter cannot be the technological stresses because ΔK is independent of the whole preparation technology of the investigated samples.

Some successive approaches[13,15,16] led Globus to elaborate an hypothesis about the origin of the term ΔK.

For polycrystals, the additional magnetoelastic energy term $\Delta K = K - K_1$ is generated by an interaction between phenomena which occur simultaneously with the spontaneous magnetization, the isotropic deformation of exchange-volume anomaly, and the magnetostrictive anisotropic deformation along the spontaneous magnetization direction. These deformations are mechanically free in the case of a single crystal, and partially constrained in the case of a polycrystal because each crystallite (grain) is surrounded by neighbors whose crystallographic orientation is different from its own. Under these conditions the term ΔK is obviously due to the intrinsic properties and becomes a specific property of the polycrystalline state.

B. Volume anomaly, exchange striction

The volume anomaly of cubic magnetic materials is the isotropic spontaneous deformation due to the isotropic exchange striction. The uniform lattice distorsion appears at the critical temperature and below, with the occurrence of the magnetic-order state associated with exchange interactions. This isotropic deformation ($\simeq 10^{-3}$) is two or three orders of magnitude higher than the anisotropic deformation corresponding to the magnetostriction ($\simeq 10^{-6}$).

For ferromagnetic materials, Néel showed that this volume anomaly results from the variation of the interaction energy (coupling forces) between magnetic atoms as a function of the distance of the magnetic layers.[17]

For insulating cubic magnetic materials, Bloch stated precisely the law for the volume dependence of the superexchange interaction[18] and related quantitatively the volume anomaly at 0 K to the variation of T_c with pressure.[19]

The theoretical expression of the volume anomaly can be found in the approximation of the molecular field theory, as follows; let us consider a magnetic material with localized moments and one kind of magnetic ion, and assume that the exchange coupling is of the Heisenberg type $-2J_{ij}S_iS_j$. Taking the formulation used by Smart,[20] the magnetic energy per unit volume is written as

$$E_m = -N\frac{3}{2}k_B T_c \frac{S}{S+1} m^2(T) \ , \tag{4}$$

where N is the number of magnetic ions per unit volume, T_c is the critical temperature (directly proportional to the exchange or superexchange interactions), S is the spin of the magnetic ion, and $m(T)$ is the reduced spontaneous magnetization, where $m(T) = M_s(T)/M_s(0)$.

Taking the assumption of Bean and Rodbell[21] in which the exchange energy (or the critical temperature) is a strong function of the atomic volume of the form V^β, and for a small-volume variation $\Delta V/V$, T_c can be written

$$T_c = T_0\left[1 + \beta\frac{\Delta V}{V}\right] \ . \tag{5}$$

T_0 would be the critical temperature if the lattice were not compressible, and β is the slope of the dependence of exchange energy (or T_c) on volume and may be either positive or negative.

The sum of the magnetic and elastic energies per unit volume is

$$E = -N\frac{3}{2}k_B T_0\left[1 + \beta\frac{\Delta V}{V}\right]\frac{S}{S+1} m^2(T) + \frac{1}{2\chi}\left[\frac{\Delta V}{V}\right]^2 \ , \tag{6}$$

where χ is the compressibility of the material.

By minimizing the total energy with respect to the volume variation we obtain directly the volume anomaly corresponding to the equilibrium state at $T < T_c$:

$$\frac{\Delta V}{V} = +N\frac{3}{2}k_B T_0 \beta\chi\frac{S}{S+1} m^2(T) \ . \tag{7}$$

ΔV is the difference between the volume of the magnetic material and the volume that a nonmagnetic material (with a similar crystallographic structure) would have at the same temperature $T < T_c$.

The volume anomaly $\Delta V/V$ can also be written as a function of the variation of T_c with pressure dT_c/dP, since it can be written [from (5)]

$$\frac{dT_c}{dP} = T_0\beta\frac{d(\Delta V/V)}{dP} = -T_0\beta\chi \ . \tag{8}$$

Hence

$$\frac{\Delta V}{V} = -N\frac{3}{2}k_B\frac{dT_c}{dP}\frac{S}{S+1} m^2(T) \ . \tag{9}$$

Let us consider a ferrimagnetic material with two sublattices i and j and with one kind of magnetic ions.

Let $m_i(T)$ and $m_j(T)$ be the reduced magnetizations of each sublattice i and j

$$m_i(T)=\frac{M_i(T)}{M_i(0)}\,,\quad m_j(T)=\frac{M_j(T)}{M_j(0)}\ .$$

Let f_i be the fraction of magnetic ions in site i, and $f_j=1-f_i$, the fraction of magnetic ions in site j, the site i being the site where the number of magnetic ions is the smallest [for example, for yttrium iron garnet (YIG) $f_i=0.4$ and $f_j=0.6$, the site i being the octahedral site a and the site j being the tetrahedral site d].

By assuming the antiferromagnetic interaction between the two sublattices i and j to be dominant, the magnetic energy can be written as, after calculations[20]

$$E_m=-N\tfrac{3}{2}k_BT_c\frac{S}{S+1}2f_im_i(T)m_j(T)\ . \qquad (10)$$

Hence the volume anomaly can be written as

$$\frac{\Delta V}{V}=+N\tfrac{3}{2}k_BT_0\beta\chi\frac{S}{S+1}2f_im_i(T)m_j(T) \qquad (11)$$

or

$$\frac{\Delta V}{V}=-N\tfrac{3}{2}k_B\frac{dT_c}{dP}\frac{S}{S+1}2f_im_i(T)m_j(T)\ , \qquad (12)$$

dT_c/dP being the variation of T_c with the pressure P.

Recently, we have experimentally determined for YIG the volume anomaly $\Delta V/V$ due to the exchange striction below T_c; these values (from 77 K to T_c) agree with those deduced from the analytical expression (12). A good agreement was also obtained in the cases of ferromagnetic and antiferromagnetic materials with localized magnetic moments.[22]

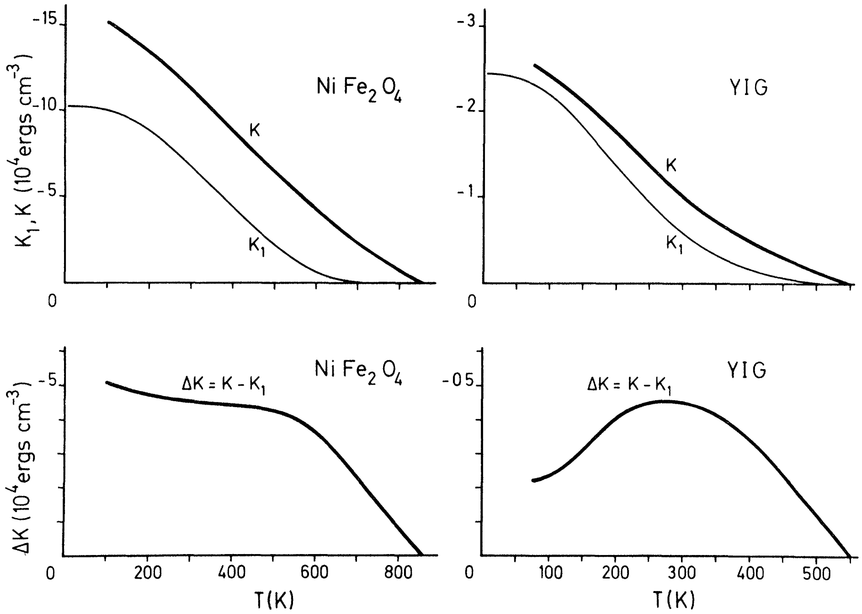
In the present work, we also observed a good agreement for $NiFe_2O_4$ between the experimental values of $\Delta V/V$ and the values deduced from the analytical expression (12), in which we take, as a first approximation, the appropriate average values for the ionic magnetic moment and for the reduced magnetizations.

II. EXPERIMENTAL RESULTS

A. Specific polycrystalline magnetoelastic anisotropy ΔK

Figure 2 (top) shows the experimental temperature dependences of the magnetocrystalline anisotropy K_1 and the total anisotropy K, both for a spinel-structure material ($NiFe_2O_4$) and a garnet-structure material (YIG).[13]

The magnetocrystalline anisotropy K_1 was measured on single crystals by ferrimagnetic resonance. The total anisotropy K was measured on polycrystals



(a) (b)

FIG. 2. (top) Experimental temperature dependences of the magnetocrystalline anisotropy K_1 (measured on single crystals) and the polycrystalline total anisotropy K (deduced from the natural spin-resonance frequency and from the rotational initial susceptibility of monodomain grain samples), for (a) $NiFe_2O_4$ and (b) YIG (Ref. 13). (bottom) Experimental temperature dependences of the specific polycrystalline anisotropy ΔK term obtained directly as the difference $\Delta K=K-K_1$ for (a) $NiFe_2O_4$ and (b) YIG .

and deduced from the natural spin-resonance frequency and from the rotational initial susceptibility on monodomain grain samples.

The specific polycrystalline magnetoelastic anisotropy ΔK was obtained directly as the difference $\Delta K = K - K_1$ [Fig. 2 (bottom)].

One can see that for YIG $\Delta K = K - K_1$ goes through a maximum at room temperature and decreases towards the low temperatures.

B. Volume anomaly $\Delta V/V$

The thermal expansion was measured by us between 77 and 1000 K by using a silica single-rod-type dilatometer. The sample length variation was detected with a linear variable-differential transformer. A precision of 0.025×10^{-3} was obtained.

Figure 3 (top) shows the thermal expansion curves for $NiFe_2O_4$ and YIG. In order to separate the magnetic thermal expansion contribution from that due to the lattice vibations, we compared the thermal expansion curves of the ferrimagnetic materials with those of nonferrimagnetic materials of similar crystallographic structure; when far from T_c the thermal expansion coefficient of the ferrimagnetic material is identical to that of the nonmagnetic material.

In the case of spinel structure [Fig. 3(a)], the nonferrimagnetic materials for comparison with $NiFe_2O_4$ are the spinels $ZnFe_2O_4$ and $CdFe_2O_4$ for $T \gtrsim 150$ K (when far from their antiferromagnetic Néel temperatures $T_N \simeq 10$ K). The spinels $ZnFe_2O_4$ and $CdFe_2O_4$ yield approximately the same relative linear expansion from $T \simeq 150$ to 900 K.[23]

In the case of garnet structure [Fig. 3(b)], the nonferrimagnetic material for comparison with YIG is the nonmagnetic garnet $Y_3Ga_5O_{12}$.[24,25]

Then one can obtain at any temperature $T \lesssim T_c$, the magnetic thermal expansion contribution from the difference

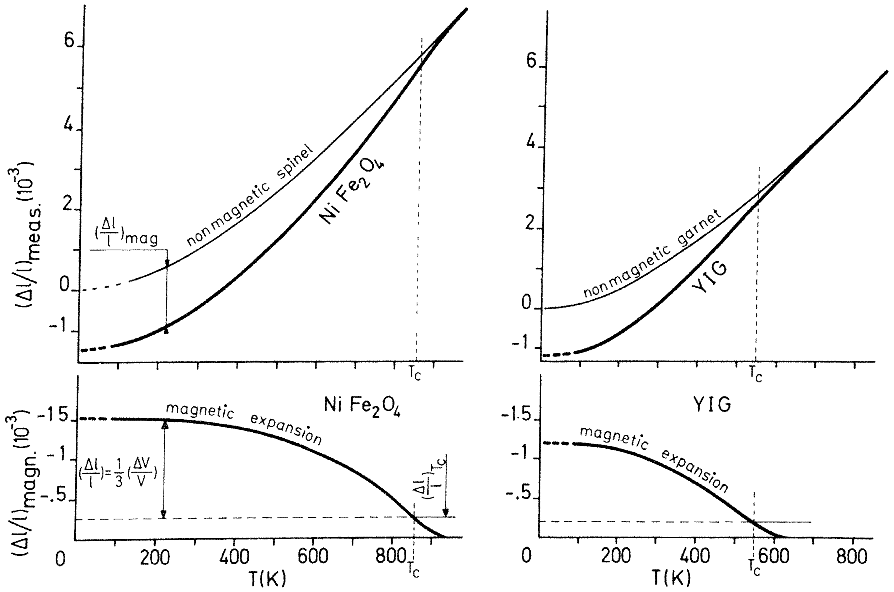
$$\left(\frac{\Delta l}{l}\right)_{\text{magn}} = \left(\frac{\Delta l}{l}\right)_{\text{magn mat}} - \left(\frac{\Delta l}{l}\right)_{\text{nonmagn mat}} . \quad (13)$$

Figure 3 (bottom) shows the magnetic thermal-expansion contribution, $(\Delta l/l)_{\text{magn}}$ for $NiFe_2O_4$ and YIG, versus temperature.

One can see at T_c a nonzero deformation $(\Delta l/l)_{T_c}$ which is due to short-range order effects (or spin-correlation effects).

For analyzing the magnetic thermal expansion contribution, we assume that the ferrimagnetic contribution $(\Delta l/l)$ due to the ferrimagnetic-order energy below T_c (with $\Delta l/l \neq 0$ for $T < T_c$ and $\Delta l/l = 0$ for $T > T_c$) can be separated from the short-range contribution, taking as a first approximation

$$\left(\frac{\Delta l}{l}\right) = \left(\frac{\Delta l}{l}\right)_{\text{magn}} - \left(\frac{\Delta l}{l}\right)_{T_c} . \quad (14)$$



(a) (b)

FIG. 3. (top) Thermal expansion curves. (a) For $NiFe_2O_4$ and for a nonmagnetic spinel material. (b) For YIG and for a nonmagnetic garnet material. (bottom) Magnetic thermal expansion curves for (a) $NiFe_2O_4$ and (b) YIG, obtained from the differences between the thermal expansions of the ferrimagnetic materials and those of the nonmagnetic materials of similar crystallographic structure at the same temperature.

Moreover, considering that the magnetic thermal-expansion contribution is isotropic (within the limit of the experimental precision $\pm 0.025\times10^{-3}$),[22] we define the volume anomaly as

$$\frac{\Delta V}{V}=3\frac{\Delta \mathrm{l}}{\mathrm{l}}\ . \qquad (15)$$

For the analysis of results, we introduce a plausible work concept in which the deformation $\Delta V/V$ is replaced by the notion of an associated virtual "exchange stress" σ_n defined by

$$\sigma_n(T)=\frac{\Delta V}{V}(T)/\chi\ , \qquad (16)$$

for $T\leq T_c$, where χ is the compressibility of the material.

This virtual exchange stress would correspond to the deformation $\Delta V/V$ of the magnetic material with respect to that of a nonmagnetic material of similar crystallographic structure at the same temperature.

One can see (Fig. 3) that for $NiFe_2O_4$ and YIG, the volume anomaly is a contraction of the lattice; therefore the "exchange stress" would be a compressive stress. Taking the compressibility values of $\chi\simeq0.5\times10^{-12}$ dyn^{-1}cm^2 for $NiFe_2O_4$ (Ref. 26) and $\chi\simeq0.6\times10^{-12}$ dyn^{-1}cm^2 for YIG (Ref. 27), the exchange stress would be (for example, at low temperatures), for $NiFe_2O_4$ since

$$\left(\frac{\Delta V}{V}\right)_0\simeq-3.75\times10^{-3}\ \text{[Fig. 3(a)]}$$

then:

$$\sigma_n(0)\simeq-7.5\times10^{9}\ \text{dyn/cm}^2\ ,$$

for YIG since

$$\left(\frac{\Delta V}{V}\right)_0\simeq-3.0\times10^{-3}\ \text{[Fig. 3(b)]}$$

then:

$$\sigma_n(0)\simeq-5.0\times10^{9}\ \text{dyn/cm}^2\ .$$

So the order of magnitude of σ_n is the same for $NiFe_2O_4$ and YIG and it is higher than that classically found for the technological stresses in ceramics [$\simeq3\times10^{8}$ dyn/cm^2 (Ref. 28)].

C. Relation between $\Delta K(T)$ and $(\Delta V/V)(T)$

Now, let us compare the experimental results with the hypothesis concerning the origin of ΔK. In that hypothesis ΔK was a magnetoelastic anisotropy term produced by a mechanism based on the interaction, in the case of polycrystalline state, between the isotropic volume anomaly $\Delta V/V$ and the anisotropic magnetostrictive deformation along the spontaneous magnetization direction (Sec. I A 4).

We assume here that, for the analysis of the relation between $\Delta K(T)$ and $(\Delta V/V)(T)$ from T_c to low temperatures, one can take the compressibility χ to be temperature independent. This hypothesis seems justified since:

(1) In the case of YIG, Young's modulus decreases from 2.06×10^{12} dyn/cm^2 at 0 K to 2.00×10^{12} dyn/cm^2 at 320 K.[29] Moreover, from measurements of the acoustic-resonance frequency (Forster apparatus) on our YIG samples, the Young's modulus decreases from 2.00×10^{12} dyn/cm^2 at 320 K to 1.95×10^{12} dyn/cm^2 at T_c, and a very weak anomaly is observed near T_c ($\simeq0.01\times10^{12}$ dyn/cm^2).

(2) In the case of $NiFe_2O_4$, Young's modulus measured on one porous sample decreases from 1.63×10^{12} dyn/cm^2 at 300 K to $1,57\times10^{12}$ dyn/cm^2 at T_c and an anomaly is observed near T_c ($\simeq0.05\times10^{12}$ dyn/cm^2).[30]

By comparing $\Delta K(T)$ and $\sigma_n(T)=(\Delta V/V)(T)/\chi$ for $NiFe_2O_4$ and YIG from 77 K to T_c, and taking the compressibility χ to be temperature independent [$\chi\simeq0.5\times10^{-12}$ dyn^{-1}cm^2 for $NiFe_2O_4$ (Ref. 26) and $\chi\simeq0.6\times10^{-12}$ dyn^{-1}cm^2 for YIG[27]] we find

(1) A linear law for the variation of $\Delta K(T)$ vs $\sigma_n(T)$ (Fig. 4) from low temperatures to T_c for $NiFe_2O_4$ [Fig. 4(a)] and from room temperature to T_c for YIG [Fig. 4(b)]. So, for these temperature ranges

$$\Delta K(T)=\lambda\sigma_n(T)=\lambda\frac{\Delta V}{V}(T)/\chi\ . \qquad (17)$$

(2) A proportionality factor, called λ because it appears to be of magnetostrictive nature from the expression (17). λ is temperature independent in these temperature ranges and specific to the material. λ appears to be directly related to the magnetostriction coefficient λ_{111} at 0 K because

$$\lambda\simeq\tfrac{1}{3}(\lambda_{111})_{0\,\mathrm{K}}\ , \qquad (18)$$

since for $NiFe_2O_4$

$$\lambda\simeq-6.7\times10^{-6}\ \text{[Fig. 4(a)]}$$

and

$$(\lambda_{111})_{0\,\mathrm{K}}\simeq-21.5\times10^{-6}\ \text{(Ref. 31)}$$
$$\simeq-23.8\times10^{-6}\ \text{(Ref. 32)}\ .$$

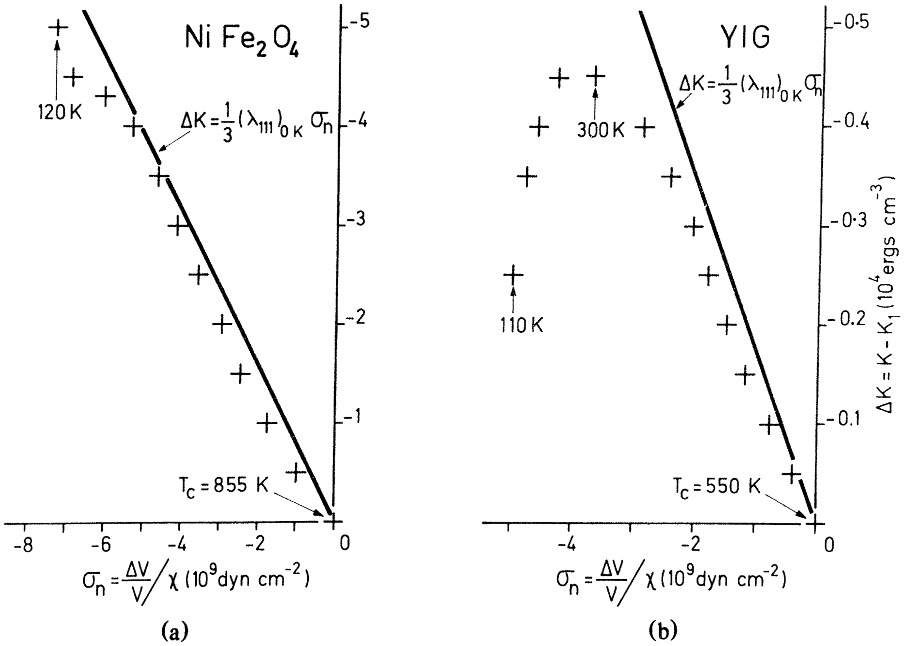
for YIG

$$\lambda\simeq-1.5\times10^{-6}\ \text{[Fig. 4(b)]}$$

and

$$(\lambda_{111})_{0\,\mathrm{K}}\simeq-5.4\times10^{-6}\ \text{(Ref. 33)}$$
$$\simeq-4.8\times10^{-6}\ \text{(Ref. 34)}\ .$$

(3) A deviation from the linear law appears for YIG at low temperatures.



(a) (b)

FIG. 4. Experimental relation between the specific polycrystalline anisotropy $\Delta K(T)$ term and the virtual "exchange stresses" $\sigma_n(T) = (\Delta V/V)(T)/\chi$ [$(\Delta V/V)(T)$—volume anomaly, χ—compressibility] from T_c to low temperatures, for (a) $NiFe_2O_4$ and (b) YIG. The experimental variations (points +) are compared with the calculated linear laws $\Delta K(T) = \frac{1}{3}(\lambda_{111})_{0\,K}\sigma_n(T)$ (thick line).

III. DISCUSSION AND INTERPRETATION

All the results including the deviation from the linear law for YIG at low temperatures become understandable if we consider the variation of the influence of the different magnetostrictive deformations λ_{111} and λ_{100} as a function of temperature.

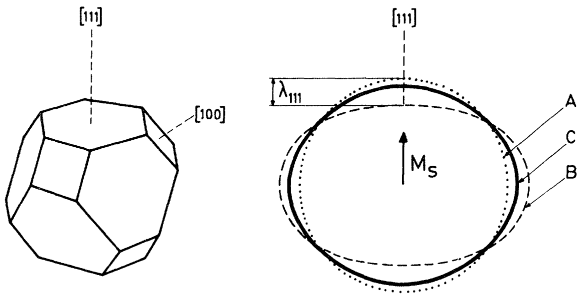
First, let us consider a mechanically free *single-crystal* spherical grain above T_c [sphere A in Fig. 5(b)]. At T_c and below a spontaneous magnetostrictive deformation λ_{111} occurs along one of the four [111] axes, which corresponds to the spontaneous magnetization direction (for $NiFe_2O_4$ and YIG) in the spontaneous magnetic state [ellipsoid B in Fig. 5(b)]. For $NiFe_2O_4$ and YIG, this deformation is a contraction ($\lambda_{111} < 0$). Therefore, simultaneous with the contraction along this [111] axis, a lattice expansion normally occurs in other planes including the perpendicular equatorial plane [ellipsoid B in Fig. 5(b)] in agreement with the classical assumption that magnetostrictive deformations take place at approximately *constant volume.*

Now, let us consider a *polycrystal.* From the proposed hypothesis, the anisotropic magnetostrictive deformations of each "single-crystal" grain are partially constrained by the neighboring grains whose crystallographic orientation is different from its own. So the expansion in the equatorial plane is partially constrained by the neighboring grains and, accordingly, the contraction along the spontaneous-magnetization [111] direction will be smaller than in the single crystal [ellipsoid C in Fig. 5(b)].

Figure 5(a) shows, for a cubic-structure ceramic material, a theoretical grain shape obtained from a truncated octahedron (tetrakaidecahedron).[35] This grain shape shows that the plane perpendicular to the [111] axis is fairly near the [100] axis and we can analyze the material behavior in this direction. The temperature dependences of λ_{111} and λ_{100} for $NiFe_2O_4$ (Ref. 31) and YIG (Ref. 33) are shown in Fig. 6; one can see that they are monotonic except that one of λ_{100} for YIG, which goes through a maximum at room temperature and decreases at low temperatures.

We assume that the constraint phenomenon is related to the magnetoelastic behavior in the plane perpendicular to the [111] axis. In the case of YIG, we observe a decrease of $\Delta K(T)$ towards the low temperatures [Fig. 2(b)] and simultaneously a similar decrease of λ_{100} in the same temperature range [Fig. 6(b)]; since the plane perpendicular to the [111] axis is fairly near the [100] axis, we can think that the two phenomena are related. So the deviation found for YIG towards the low temperatures [Fig. 4(b)] seems to strengthen the plausibility of the proposed mechanism.

It is also striking that in both cases, in spite of the differences between the values of λ_{111} and λ_{100} (the different order of magnitude and the different respec-



(a) (b)

FIG. 5. (a) Theoretical grain shape for a cubic-structure ceramic material, obtained from a truncated octahedron (tetrakaidecahedron) (Ref. 35). (b) Idealized *spherical* grain shape. Sphere A (dotted line): single-crystal spherical grain, at $T > T_c$. Ellipsoid B (dashed line): mechanically free single-crystal grain at $T < T_c$ showing the magnetostrictive contraction λ_{111} along the spontaneous magnetization [111] direction and the magnetrostrictive expansion in the perpendicular equatorial plane. Ellipsoid C (thick line): the same crystal grain in a polycrystalline state at $T < T_c$ showing the anisotropic magnetostrictive deformations partially constrained by the neighboring grains whose crystallographic orientations are different.

tive values of λ_{111} and λ_{100} in both cases are, $\lambda_{111} < \lambda_{100}$ for $NiFe_2O_4$, $\lambda_{111} > \lambda_{100}$ for YIG), the proportionality factor λ between $\Delta K(T)$ and $\sigma_n(T)$ is found to be directly related to an *anisotropic* property, the magnetostrictive deformation along the magnetization [111] direction

$$\lambda \simeq \tfrac{1}{3}(\lambda_{111})_{0\,\mathrm{K}} \ .$$

The coefficient of about $\frac{1}{3}$ is the same for $NiFe_2O_4$ and YIG; it would represent the "averaged constraint" for the magnetostrictive deformation along the magnetization [111] direction for each grain within the polycrystal. This result seems plausible when a random orientation of the grains is considered.

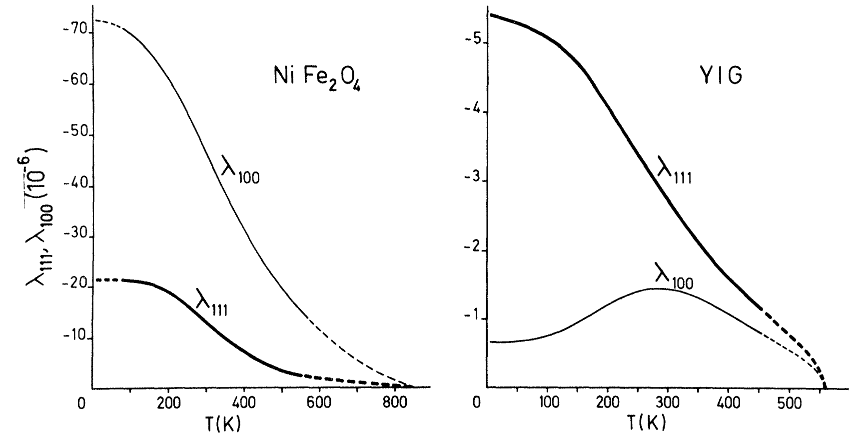
We can now express the magnetoelastic anisotropy term ΔK in terms of ionic parameters by using the single-ion theory of magnetostriction. E. R. Callen *et al.* applied the theory of magnetostriction in cubic insulators to the Néel model of a ferrimagnet, and evaluated for each symmetry of strain mode the individual magnetoelastic coupling coefficients of each type of site (or sublattice). They found an excellent agreement between the theoretical predictions of the temperature dependence of magnetostriction constants and the observed values for YIG.[33]

By using the same single-ion theory of magnetostriction, E. M. Smokotin *et al.* evaluated the magnetoelastic coupling coefficients of each type of site for $NiFe_2O_4$ and obtained, for the temperature dependence of λ_{111} and λ_{100}, a good agreement between theory and experiment.[31]

In this single-ion theory of magnetostriction, the magnetostrictive deformation $(\lambda_{111})_{0\,\mathrm{K}}$ is directly related to the magnetoelastic coupling coefficients $B^{\epsilon}_{0.2}$

$$(\lambda_{111})_{0\,\mathrm{K}} = \frac{1}{3c_{44}}\left(\frac{15}{4\pi}\right)^{1/2}[B^{\epsilon}_{0.2}(i)+B^{\epsilon}_{0.2}(j)] \ , \qquad (19)$$

where c_{44} is the elastic coefficient, which is considered, like the compressibility χ, to be temperature independent, and $B^{\epsilon}_{0.2}(i)$ and $B^{\epsilon}_{0.2}(j)$ are the magnetoelastic coupling coefficients of site i and site j for



(a) (b)

FIG. 6. Magnetostriction constants λ_{100} and λ_{111} as a function of temperature. (a) Single-crystal $NiFe_2O_4$, from E. M. Smokotin *et al.* (Ref. 31). (b) Single-crystal YIG, from E. R. Callen *et al.* (Ref. 33).

the strain along the [111] direction. So for $NiFe_2O_4$ (Ref. 31)

$$B^{\epsilon}_{0.2}\ (\text{tetra}) = +137 \times 10^6\ \text{ergs/cm}^3\ ,$$

$$B^{\epsilon}_{0.2}\ (\text{octa}) = -178 \times 10^6\ \text{ergs/cm}^3\ ,$$

and for YIG (Ref. 33)

$$B^{\epsilon}_{0.2}\ (\text{tetra}) = -17.2 \times 10^6\ \text{ergs/cm}^3\ ,$$

$$B^{\epsilon}_{0.2}\ (\text{octa}) = +5.67 \times 10^6\ \text{ergs/cm}^3\ .$$

Now the experimental linear law

$$\Delta K(T) \simeq \tfrac{1}{3}(\lambda_{111})_{0\,\mathrm{K}}\sigma_n(T) \simeq \tfrac{1}{3}(\lambda_{111})_{0\,\mathrm{K}}\frac{\Delta V}{V}(T)/\chi \tag{20}$$

becomes, using relations (11) and (19),

$$\Delta K(T) \simeq \frac{1}{3}\left[\frac{1}{3c_{44}}\left(\frac{15}{4\pi}\right)^{1/2}[B^{\epsilon}_{0.2}(i) + B^{\epsilon}_{0.2}(j)]\right] \times N\frac{3}{2}k_B T_0 \beta \frac{S}{S+1} 2 f_i m_i(T) m_j(T) \tag{21}$$

for one ferrimagnetic polycrystalline material with two sublattices *i* and *j*.

This form of the analytical expression (21) shows that the specific polycrystalline magnetoelastic anisotropy ΔK is an intrinsic physical property.

(1) Its temperature dependence, as for magnetocrystalline anisotropy K_1 and magnetostrictive coefficents λ_{111} and λ_{100}, is given by a function of the magnetization; this function is simply the *product* of the temperature dependences of the reduced magnetization of *each sublattice*.

(2) Its magnitude is proportional to (a) the *sum* of the magnetoelastic coupling coefficients for *each sublattice* for the strain along the spontaneous magnetization direction; (b) the inverse of the elastic constant c_{44}; (c) the magnetic parameters related to the exchange interactions (the magnetic order critical temperature T_0 and the slope β of the dependence of superexchange energy on volume); and (d) the magnetic parameters related to the magnetic structure are N, the number of magnetic ions per unit volume, S, the ion-magnetic moment, and f_i, the fraction of magnetic ions in site *i*.

In addition, the present results permit the assumption that the atomic origin of the specific polycrystalline magnetoelastic anisotropy ΔK could be a *coupling mechanism between the superexchange interaction and the spin-orbit interaction* via the electronic-charge clouds of magnetic ions in the crystal field, a mechanism which would be specific of the polycrystalline state.

In this work we used the classical notion of a magnetoelastic anisotropy energy of the form $\lambda\sigma$ and we attributed the "virtual stress" σ to the apparent parameter of the large isotropic-exchange striction. However, the actual mechanism of the atomic structure variation which is the origin of the phenomenon is not known exactly and it is possible that another approach could lead to the same results.

IV. CONCLUSIONS

(1) The hypothesis about the origin of the magnetoelastic anisotropy ΔK specific of the polycrystalline state is analytically verified; ΔK is related to the *product* of the strong isotropic "exchange stresses" due to the exchange striction by the anisotropic magnetostrictive deformation constrained by the neighboring grains.

(2) The experimental results obtained on $NiFe_2O_4$ and YIG. (a) the linear law of variation of $\Delta K(T)$ as a function of $\sigma_n(T) = (\Delta V/V)(T)/\chi$, and (b) the apparent deviation from this linear law at low temperatures for YIG, becomes understandable when the proposed magnetoelastic mechanism is considered.

(3) An analytical expression of $\Delta K(T)$ is established as a function of ionic structure properties, namely, exchange striction, magnetostrictive coupling, and sublattice magnetizations.

The plausibility of these conclusions is strengthened by other partial results obtained in the field of polycrystalline magnetoelastic-anisotropy investigation on other compositions such as $Ni_{1-x}Zn_xFe_2O_4$, $Ni_{1-x}Cd_xFe_2O_4$, and $Y_{3-x}Gd_xFe_5O_{12}$. The final results for these compositions are planned to be published later.

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