DETERMINATION OF THE DOMAIN WALL ENERGY AND THE EXCHANGE CONSTANT FROM HYSTERESIS IN FERRIMAGNETIC POLYCRYSTALS

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Résumé. — La loi fondamentale, précédemment établie pour le YIG, qui relie les pertes d'hystérésis à l'état d'aimantation, est généralisée au ferrite de Ni et à des ferrites mixtes de NiZn et NiCd. L'interprétation qui en est donnée est basée sur la *théorie de la dimension de la paroi* proposée antérieurement par Globus : l'énergie dépensée pour les décrochages successifs de la paroi et l'énergie dépensée pour la formation et destruction d'une partie de la surface de la paroi seraient à l'origine des pertes traduites par le cycle d'hystérésis.

Il en résulte une méthode expérimentale de détermination de l'énergie de paroi γ et de la constante d'échange A, à partir de la simple mesure de surface des cycles d'hystérésis d'échantillons polycristallins.

Abstract. — A fundamental law previously found in YIG, which relates the hysteresis losses to the magnetization state is generalized to Ni ferrite and to mixed NiZn and NiCd ferrites. The interpretation which is given is based on *the domain wall size theory* previously proposed by Globus : the energy lost by the continuous pinning and depinning of the domain wall as it moves and the energy lost by the formation and the destruction of a part of the domain wall surface may be the origins of the losses which find their expression in the hysteresis loop.

That results in an experimental method to determine, from simple measurements of loop areas on polycrystalline samples, the domain wall energy γ and the exchange constant A.

1. **Introduction.** — In a previous paper [1], we have established a direct relation between the hysteresis losses and the domain wall energy by investigating the origin of these losses in a series of YIG samples. Such a relation resulted from an analyzis, in the light of the *domain wall size* theory [2], of the divergence between experimental and theoretical values.

As a matter of fact the pinning force (f per unit length) which controls the field dependence of the domain wall diameter was a parameter good enough to establish proportion laws as far as the effect of the grain size on the magnetization mechanisms are concerned, but using such a parameter only to compute the theoretical curves did not lead to a full quantitative agreement. In the case of the initial magnetization curve, the divergence increases with the magnetization value (Fig. 1, top), which fact we have supposed at first due to a demagnetizing effect.

However such a demagnetizing effect would be reversible : consequently the hysteresis losses might not be affected ; it can be seen on figure 1 (bottom), for a same variation of the magnetization (0.2 to 0.6 M_s), an increase of the experimental loop areas larger than that of the corresponding theoretical ones.

Since the divergence appears systematically as a law, that means that such a divergence could be due to an additional lossy mechanism related also to the first basic concept of the model — the domain wall surface variation — which originates these laws.

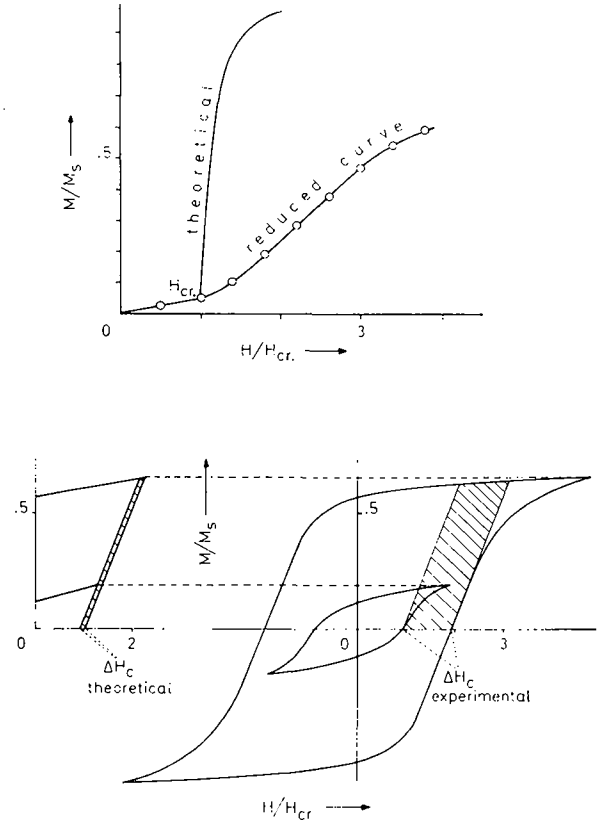

FIG. 1. — Divergence between the theoretical curves — calculated by using the pinning parameter only — and the experimental results.

A first lossy term, considered as a solid friction due to the pinning force, consists of successive pinnings and depinnings of the domain wall to the grain boundary. However this process would really be a sucession of destructions and creations of very small parts of domain wall just near the grain boundary. In the same way, much larger surface variations of the domain wall due to its motion inside the spherical grain, would also result in larger losses ; that could be the supplementary term which is looked for.

According to the theory we used, the magnetization state (which is defined by the domain wall position inside the spherical grain) gives a possible access for investigating the wall surface variation and, as it is classical, the loop area permits the hysteresis losses to be measured.

So a first investigation carried out on a series of YIG polycrystalline samples permits a law relating the energy lost to the magnetization state to be established ; from this law it was possible to deduce the value of the domain wall energy, in agreement with the theoretical value calculated by using the Landau-Lifshitz formula $\gamma = 2\sqrt{AK_1}$.

The aim of the present paper is to generalize for other compositions the results obtained on YIG samples and to attempt to evaluate the exchange constant A for such compositions.

2. **Preparation and test of the samples.** — The whole preparation process must produce samples showing not only a good ionic structure but also a good granular structure : has been achieved by using a special technology already described [3].

To characterize a composition we measure the following parameters :

— lattice constant, measured by X-rays diffraction,

— spontaneous magnetization M_s, deduced by extrapolating at zero field the M(H) curves obtained by using a vibrating sample magnetometer.

— Curie temperature T_c, defined from the downfall to zero of the initial susceptibility χ_i as the temperature increases ; near T_c the initial susceptibility value varies from several hundred (or even several thousands) to zero for a given temperature increase, the value of which being related to the quality of the ionic structure of the sample ; for a good quality material the downfall is only a few degrees wide. Then the $\chi_i = f(T)$ curve gives the values of T_c and serves as a test for the homogeneity of the ionic structure.

We also measure parameters which are specific of the investigated samples :

— actual density and porosity,

— mean grain size D_m, determined by counting the grains over a given polished area of a microphotograph.

3. **Hysteresis measurements.** — Since the materials investigated are magnetically soft, hysteresis measurements are carried out on toroïdal samples of a rectangular cross-section ; as it is classical we use two different windings, the first one for applying the magnetic field and the second one for reading the flux variations.

To characterize a given hysteresis loop and to measure the corresponding energy lost, we have chosen to record the loop by using an X-Y recorder instead of a photographic camera. Then the loop areas, the magnetization and field values can be measured with a good accuracy. Samples are magnetized with an a. c. field at 1 kHz ; the voltage induced in the secondary winding is integrated by means of an operational amplifier with the required feedback. The signals related to the applied field and to the induced voltage feed a sampling device which operates a frequency conversion, allowing us to record the loops on the X-Y recorder at an apparent frequency of about 10^{-2} Hz [4].

In the toroidal sample arrangement there is no demagnetizing affect, but the continuous applied a. c. field cannot exceed a few ten oersteds, which is however mostly large enough in our case. However it can be pointed out that the magnetic field value generated by the current in the primary winding depends on the distance to the toroïd axis which in turn results in a radius dependence of the induction. From a theoretical point of view it is mostly considered that samples with an inner to outer diameter ratio as closer to unity as possible must show the better field homogeneity.

In order to verify a possible influence of this effect upon the actual samples in the case of our investigations, we have carried out the following experiment, as described on figure 2. First of all, a given loop is recorded on a sample with a diameter ratio 4.3. After machining the sample to obtain a diameter ratio of 1.3, one records the loop corresponding to the same magnetization value at the tip of the loop. Despite some small differences around the tip, the loop areas are almost the same. In both the cases we used the notion of *mean value* for field and induction.

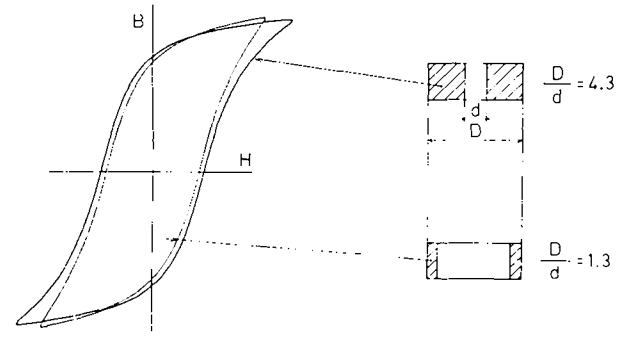

FIG. 2. — Independence of the hysteresis loop areas despite the large diameter ratio variation for a toroïdal sample.

Then in the case of soft ferrimagnets, any field inho-

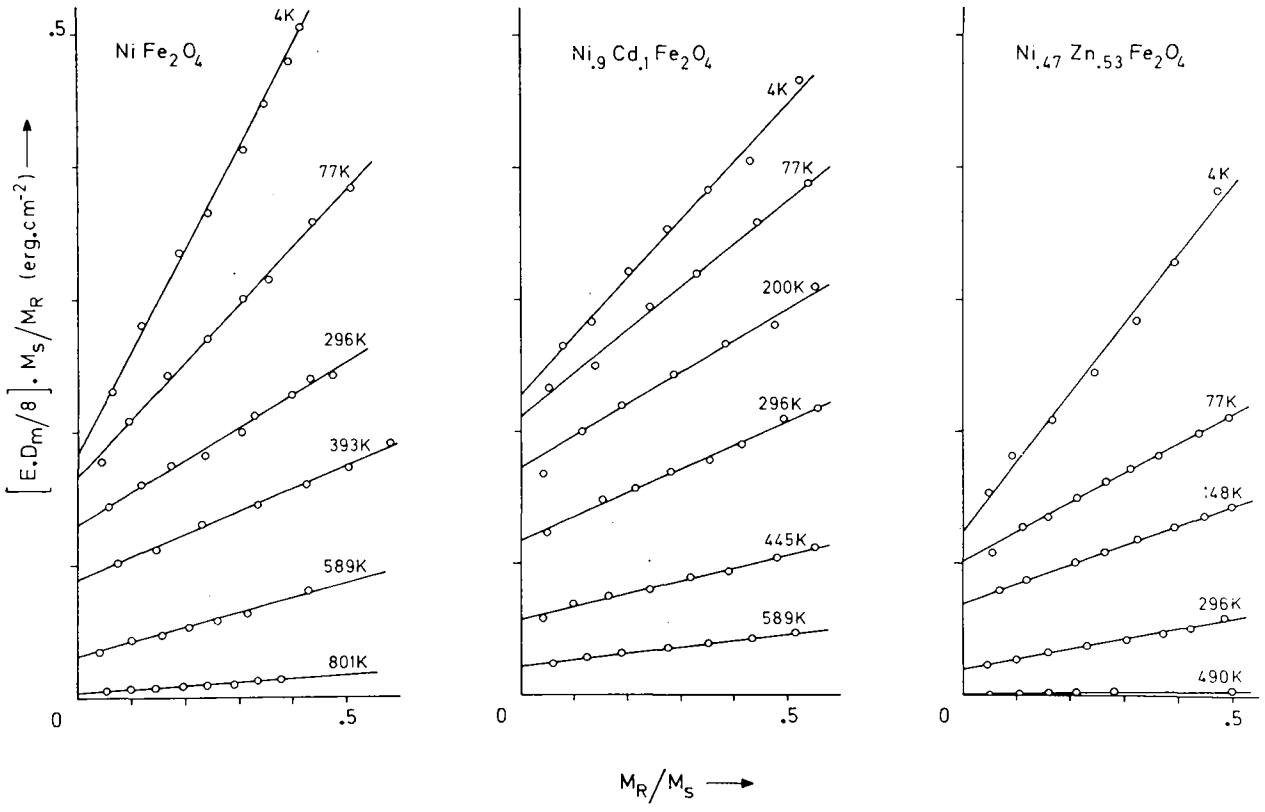

FIG. 3. — The fundamental law relating the hysteresis losses E of each loop to its relative remanent magnetization $m_R = M_R/M_S$.

mogeneity is smoothed by the magnetization flux, because of the small value of the magnetizing field (a few oersted) as a comparison with the flux due to the spontaneous magnetization (several thousands gauss) [3]. So, in the present case, the influence of the sample diameter ratio upon the hysteresis losses can be neglected.

4. **Experimental results.** — The results presented here have been obtained on series of spinel structure samples of three compositions for which we know the magnetocrystalline anistropy values :

$NiFe_2O_4$, $Ni_{0.9}Cd_{0.1}Fe_2O_4$ and $Ni_{0.47}Zn_{0.53}Fe_2O_4$.

To make the analyzis easier, we have chosen to characterize a given hysteresis loop by its remanent magnetization value M_R (or its relative value $m_R = M_R/M_s$; for all the loops we have measured the corresponding energy lost at several temperatures from 4.2 K to T_c.

Most of the results are shown in figure 3, where the quantity $(ED_m/8)\ (1/m_R)$ is plotted as a function of m_R (multiplying E by D_m removes the grain size effect previously shown [2, 5]).

Such a presentation shows clearly the existence of a fundamental law (i.e. a law valid as a function of temperature) relating the losses and the magnetization state, which confirms the results already obtained on YIG[1] : up to $m_R \simeq 0.5$ one observes a straight line which intersects the Y-axis at a non zero point.

As it will be shown later the slope of each line is proportional to the domain wall energy value per surface unit, which values being represented as a function of temperature by the circles on figure 4 (bottom) for $NiFe_2O_4$, on figure 4 (top) for $Ni_{0.9}Cd_{0.1}Fe_2O_4$ and

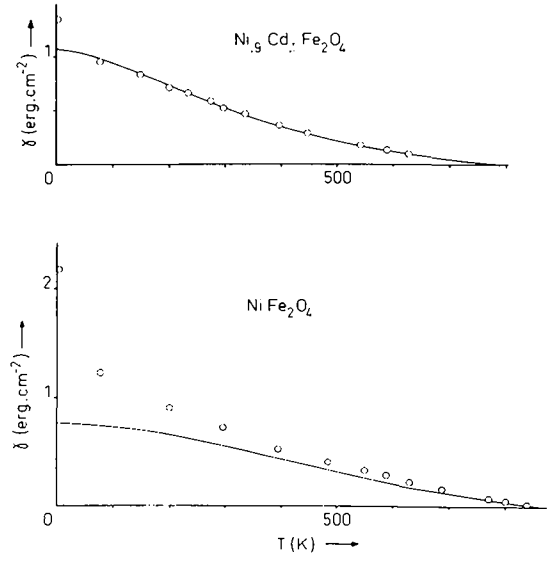

FIG. 4. — Experimental (circles) and theoretical (continuous line) value of the domain wall energy γ of $Ni_{0.9}Cd_{0.1}Fe_2O_4$ (top) and of $NiFe_2O_4$ (bottom).

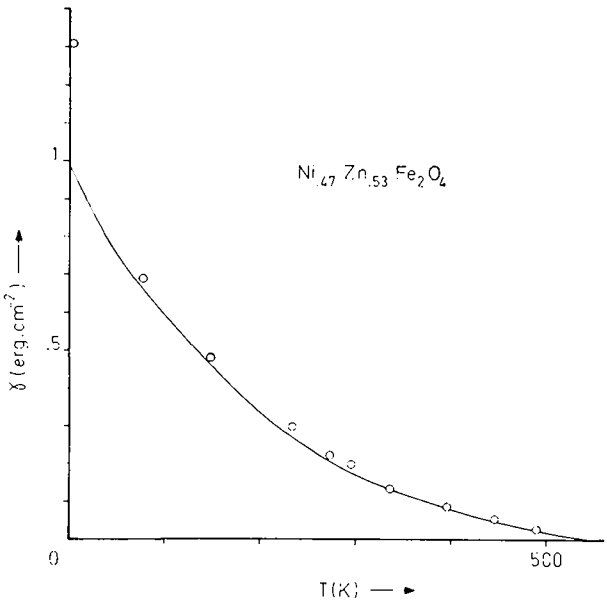

FIG. 5. — Experimental (circles) and theoretical (continuous line) values of the domain wall energy γ of $Ni_{0.47}Zn_{0.53}Fe_2O_4$.

on figure 5 for $Ni_{0.47}Zn_{0.53}Fe_2O_4$. Continuous lines represent the theoretical values.

Looking at the shapes of the $\gamma(T)$ curves, it can be seen that, if we start from the Curie temperature value T_c, the γ value increases smoothly in all the cases when the temperature decreases down to 77 K ; the increase is relatively strong between 77 and 4.2 K, particularly for the Ni ferrite ; in this case the value at 4.2 K is twice as large as the value at 77 K. Such a rapid increase, observed in all the compositions we have investigated, is not yet explained.

5. **Discussion and interpretations.** — 5.1 MECHANISM OF THE LOSSES AND LOSS VARIATION LAW. — Since in the present paper we are only ocncerned with dissipative phenomena in the quasi static state, we do not consider the reversible bulging of the domain wall but only the displacement of a flat domain wall.

According to the theory proposed by Globus [2-3], in a remanent state the domain wall of a radius r_0 is flat and located at a distance a_0 from the diametrical position (refer to Figure 6).

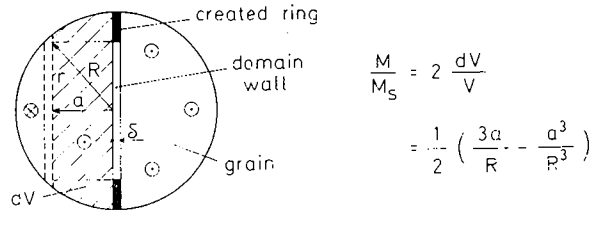

FIG. 6. — The unique domain wall in a spherical grain used in the *domain wall size theory* [2-3]. An energy lost proportional to the domain wall energy which is included in the peripherical ring would be responsible for the divergence shown on figure 1.

The relative remanent magnetization is

$$m_R = \frac{M_R}{M_s} = 2\frac{dV}{V}$$

where dV is the volume swept by the domain wall when moving from the diametrical position to a_0 and V is the volume of the grain.

Let the reduced quantities :

$$\alpha_0 = \frac{a_0}{R}, \qquad \rho_0 = \frac{r_0}{R}$$

$$m_R = \frac{1}{2}(3\alpha_0 - \alpha_0^3).$$

Now we consider the dissipative terms related to the domain wall displacement (as described in paragraphe 1) :

• first of all is the energy lost due the continuous pinning and depinning of the wall at the boundary when it moves, which can be calculated by considering the peripheric pinning force $F = 2\pi rf$ (f by length unit) as a friction force giving rise to the energy lost

$$W_1 = 8\pi R^2 f \int_0^{\alpha_0} \rho \, d\alpha ;$$

• second is a term proportional to the energy included in the peripheric ring. A full loop includes [4]

$$W_2 = n\gamma 4\pi R^2 \alpha_0^2 ;$$

where n is a proportion coefficient.

So, due to these two lossy terms, the total hysteresis loss E per volume unit is

$$E = \frac{3}{4\pi R^3} 8\pi R^2 f \int_0^{\alpha_0} \rho \, d\alpha + n\gamma 4\pi R^2 \alpha_0^2$$

or

$$\frac{ED}{6} = 2f \int_0^{\alpha_0} \rho \, d\alpha + n\gamma\alpha_0^2 .$$

In the case of moderate magnetization values ($m_R \lesssim 0.5$, i.e. $\alpha \lesssim 0.35$ and $\rho \gtrsim 0.94$) we can simplify the frictional term by assuming $\rho \simeq 1$; then we obtain :

$$\frac{ED}{6} \simeq 2f\alpha_0 + n\gamma\alpha_0^2 .$$

In these conditions, the expression of the relative remanent magnetization is also simplified :

$$m_R \simeq \frac{3\alpha_0}{2}$$

which permits the energy lost to be related directly to the magnetization, state as follows :

$$\frac{ED}{8}\frac{1}{m_R} \simeq f + \frac{n\gamma}{3} m_R .$$

This simplified calculation gives a variation similar

to the observed experimental law. The most important fact is that the second order term (with respect to m_R) appears to be directly proportional to the domain wall energy value γ, which can be esaily deduced, according to such a way, from the slope of the experimental straight lines.

5.2 DOMAIN WALL ENERGY. — Figure 7 whows our previous experimental results [1] plotted for a

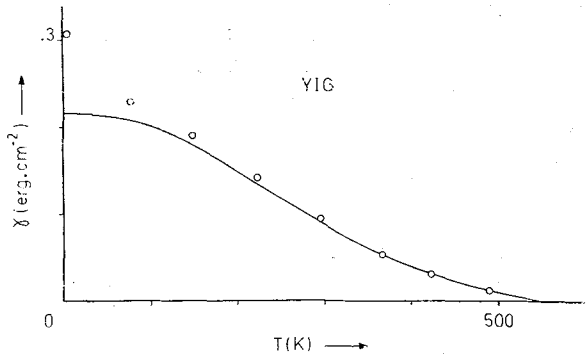

FIG. 7. — Experimental (circles) and theoretical (continuous line) values of the domain wall energy γ of YIG.

series of YIG samples wich are completed with some recent results at 4.2 K. Solid lines are the theoretical values of the domain wall energy γ_0 calculated by using the Landau-Lifshitz formula [6] $\gamma_0 = 2\sqrt{AK_1}$ where A is the exchange constant deduced from the results published by Le Craw and Walker [7] and K_1 is the magnetocrystalline anisotropy.

The better agreement between calculated and measured values is obtained over a wide temperature range by using the proportion coefficient value $n = 1$.

Such a n value means that, in a representation using a spherical grain, it would be necessary to give energy not only to create a part of domain wall but also to destroy it.

It is surprising that such a simple mathematical representation of a process occuring in a single grain permits a quantitative absolute picture of the mechanism acting at the whole material scale to be obtained : even indeed if it is assumed that energy must be lost only to create the domain wall, the corresponding value that we deduce is only twice as large as the theoretical value.

At the actual material scale it is plausible, due to continuity reasons, that the decrease of a domain wall in a grain is possibly followed by an increase of the domain wall size in the neighbouring grains : then the observed losses may correspond to the energy needed to displace the domain wall in the actual material.

Unfortunately the results we have obtained on YIG cannot be directly checked on other materials, because of a lack of data about the exchange constant value.

Nevertheless, considering that, in the one hand n shows in the case of YIG a constant value over a wide variation range of the intrinsic properties and that, in the other hand we always observe in the case of Ni ferrites and mixed Ni ferrites the same law despite the large variations of the parameters, we will use in all the cases presently investigated the value $n = 1$ that we have found.

In such a way we obtain the experimental values of the domain wall energy which are represented by the circles on figure 4 (top) for $Ni_{0.9}Cd_{0.1}Fe_2O_4$, figure 4 (bottom) for $NiFe_2O_4$ and figure 5 for $Ni_{0.47}Zn_{0.53}Fe_2O_4$.

5.3 EXCHANGE CONSTANT. — Now, using the values of γ so determined, we will make an attempt to get some informations about the value of the exchange constant A.

In the case of YIG, by analyzing the value of A deduced from the data of Le Craw and Walker, it can be seen the following :

1) thermal variations of $A(T)$ are well represented by the function $(1 - T/T_c)^{1/2}$ except for the very low temperature range ;

2) the extrapolated experimental value

$$A(0) \simeq 5 \times 10^{-6} \text{ erg.cm}^{-1}$$

is in good agreement with the evaluation of the classical ferromagnetism

$$A(0) \simeq \frac{kT_c}{a} \simeq 0.61 \times 10^{-6} \text{ erg.cm}^{-1}$$

with $T_c = 550$ K and $a = 12.376 \times 10^{-8}$ cm.

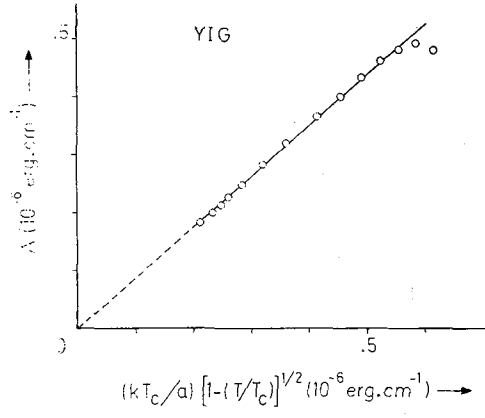

FIG. 8. — Agreement between the values of the exchange parameter $A(T)$ of YIG deduced from the results of Le Craw and Walker [7], and the new proposed formula

$$A(T) = \frac{kT_c}{a}(1 - T/T_c)^{1/2}.$$

Figure 8 shows that $A(T)$ deduced from Le Craw-Walker results is always proportional to

$$(kT_c/a)(1 - T/T_c)^{1/2}$$

and is only smaller by 20 percents.

If we assume that such a determination may be used for the other compositions investigated, then we can calculate the theoretical values of the domain wall energy γ_0, represented by the solid lines on figures 4 and 5.

To calculate $A(T)$ the following values have been used :

$T_c = 858$ K $\quad a = 8.34 \times 10^{-8}$ cm for $NiFe_2O_4$

$T_c = 781$ K $\quad a = 8.38 \times 10^{-8}$ cm for $Ni_{0.9}Cd_{0.1}Fe_2O_4$

$T_c = 540$ K $\quad a = 8.40 \times 10^{-8}$ cm for $Ni_{0.47}Zn_{0.53}Fe_2O_4$.

As mentionned above we find at low temperatures, like in the case of YIG, experimental values of γ higher than those calculated for the compositions investigated. But apart from these exception in this range of temperatures, a good agreement is obtained in the case of NiCd and NiZn ferrites (divergence less than a few percent) ; the experimental value obtained in the worse case, i. e. in the case of Ni ferrite, is only higher by less than 20 percent than the calculated value, over a wide temperature range (77 K to 858 K).

Even if it remains to know why values measured at low temperatures are, like for YIG, higher than those calculated by this way, it can be assumed that the value of A is not so far from the actual value, because of the agreement found over a wide range of temperatures and compositions.

6. **Conclusion.** — By investigating three spinel ferrites, the present work confirms the fundamental law, previously found in YIG, which relates the hysteresis losses to the magnetization state.

Using the « domain wall size » theory proposed by Globus, it has been possible to show that two lossy mechanisms act simultaneously : the pinning-depinning of the domain wall and the variation of the domain wall surface ; both of them are related to the domain wall energy through a process of creation and destruction of parts of the domain wall.

It is possible to determine the domain wall energy γ from a simple measurement of the hysteresis losses on polycrystalline samples.

The good agreement found between the experimental values of the domain wall energy and the theoretical values calculated from the Landau-Lifshitz formula (by using an empirical determination of the exchange parameter A) permits the following exchange law to be proposed, in the absence of a better one :

$$A(T) = \frac{kT_c}{a}(1 - T/T_c)^{1/2} .$$

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