

## Domain Rotation in Nickel Ferrite

FIELDING BROWN AND CHARLES L. GRAVEL  
*Sprague Electric Company, North Adams, Massachusetts*

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An experiment has been performed which leads to the conclusion that the initial permeability and radio-frequency dispersion in sintered nickel ferrite are due principally to rotation of crystallite magnetic moments in an equivalent anisotropy field. Samples have been prepared by the partial sintering of nickel ferrite particles of size 0.5 to 1.0 micron. For a series of samples sintered to various maximum temperatures, complete permeability spectra have been obtained, and it has been found that the susceptibility at low frequencies is related to the resonant frequency of the dispersion, defined as the point where susceptibility reduces to one-half its low-frequency value, in a manner which is predictable from the theory of domain rotation. Significant contribution from domain wall motion is probably inhibited by the small grain size in the samples studied. Our results do not preclude the existence of domain wall motion as a significant phenomenon in the behavior of certain ferrites, but indicate the importance of polycrystalline structure in determining the mechanism of initial polarization.

### INTRODUCTION

THE process by which ferrites polarize in an external magnetic field has been studied in the past by a number of workers. Using single crystals of magnetite, Galt<sup>1</sup> has shown that the initial permeability is due to motion of a domain boundary and that the dispersion region at 3000 cps is due to this mechanism. In the case of polycrystalline ferrites formed by a sintering process the evidence has not been as clear.

Snoek's original work<sup>2</sup> on the permeability spectra of various Ni-Zn and Mn-Zn ferrites has shown that the resonant absorption of energy in the radio-frequency region could be explained as a gyromagnetic resonance of the magnetic moments of the crystallites in internal anisotropy fields. This conclusion was based on a comparison of the susceptibility at very low frequencies, as attributed to rotation of the magnetic moments, with the frequency at which these moments resonate. The assumption was made that the governing equations are those of Landau and Lifshitz<sup>3</sup> and that the equivalent internal fields tending to orient the moments of grains along certain equilibrium positions were the same for the static as for the dynamic case. The existence of heavy damping, distributions of resonant frequencies among crystallites, internal demagnetization, and the effective  $g$  factor for the two-sublattice ferrites<sup>4</sup> were not considered by Snoek. It was further assumed that, in the sintered ferrites studied, the motion of domain walls did not contribute to the susceptibility. Nevertheless, Snoek obtained rough agreement between theory and experiment. The view that the initial permeability of these materials is due mainly to domain rotations and that the dispersion of the permeability at high frequencies is a process of

gyromagnetic resonance has also been expressed by Wijn.<sup>5</sup>

More recently the measurements of Rado *et al.*<sup>6</sup> on Ferramic A, a commercial sintered magnesium ferrite, have been interpreted to demonstrate the existence of domain wall motions and to show that this process is the principal one evident in the initial susceptibility. In observing the permeability spectrum of Ferramic A, Rado found two dispersion regions, one at approximately 40 Mc/sec and another at 1400 Mc/sec. The former resonance disappeared when the material was divided into particles of approximately single domain size, whereas the higher-frequency resonance remained in altered form. From this fact it was concluded that the lower-frequency resonance was due to a domain wall resonance which was removed when the domain walls were eliminated by subdivision. Quantitative tests of this hypothesis gave approximate agreement between the experimental data and the theory of domain wall motion.

Before describing the present results on the initial permeability spectra of sintered nickel ferrites, it seems worthwhile to emphasize the importance of differences in composition and preparation in determining the behavior of ferrites. Thus, although a number of investigators have found domain rotation to be predominant, the possibility of wall motions in other samples is by no means excluded. For instance, in Ferramic A the wall resonance observed by Rado can be ascribed to the existence of relatively large grains in the solid material (5-50 microns) although the samples of ferrite composed of much smaller grains, as for the cases we will describe, may show a resonance understandable only on the basis of domain rotation. It is the purpose of the present work to describe experiments done on certain specially prepared materials which serve as examples of the case in which the initial polarization of the material proceeds mainly by rotation of magnetization within a domain

<sup>1</sup> J. K. Galt, Phys. Rev. **85**, 664 (1952); Revs. Modern Phys. **25**, 93 (1953).

<sup>2</sup> J. L. Snoek, Nature **160**, 90 (1947).

<sup>3</sup> L. Landau and E. Lifshitz, Physik Z. Sowjetunion **8**, 153 (1935).

<sup>4</sup> R. K. Wangsness, Phys. Rev. **93**, 68 (1954); F. Brown and D. Park, Phys. Rev. **93**, 381 (1954).

<sup>5</sup> H. P. J. Wijn, Leiden University, thesis, 1953 (unpublished).

<sup>6</sup> G. T. Rado, Revs. Modern Phys. **25**, 81 (1953).

under the joint influence of the applied signal and an equivalent anisotropy field.

### EXPERIMENTAL

In initiating the present work it was our purpose to attempt a stepwise reversal of the experimental procedure used by Rado<sup>6</sup> in which the permeability spectrum of solid Ferramic A was compared with the subsequently obtained spectrum of the material in finely powdered form. Our procedure was, therefore, to start with a fine ferrite powder whose spectrum had been determined and to sinter pressed samples of this powder at various maximum temperatures in order to trace the appearance of the lower-frequency absorption peak described by Rado.

#### A. Sample Preparation

The toroidal samples used for measurements of the permeability spectrum were prepared in a manner similar to that used in standard ferrite manufacture except that extra precaution was taken to assure a powder of known properties as the starting material. To do this, stoichiometric proportions of NiO and Fe<sub>2</sub>O<sub>3</sub> were thoroughly mixed with KCl in powdered form in the ratio of approximately twenty parts by weight of KCl to NiO and Fe<sub>2</sub>O<sub>3</sub>. This mixture was then heated to 960°C in an "Alundum" crucible, a temperature sufficient to melt the salt. Upon cooling the contents and dissolving away the KCl, a fine brown ferromagnetic powder was obtained which was shown by x-ray analysis to consist of more than 95 percent nickel ferrite. Chemical tests showed that the ratio of nickel to iron was stoichiometric nor could we detect by chemical analysis the presence of unreacted NiO or Fe<sub>2</sub>O<sub>3</sub>. The particle sizes of this powder were determined by electron microscope observation and were found to be principally within the range of 0.5 to 1.0 micron.

After washing and drying the powder, the toroidal samples were pressed using a pressure of about 30 000 lb/in.<sup>2</sup> The nominal dimensions of the samples were  $\frac{1}{2}$  in. o.d.  $\times$   $\frac{1}{4}$  in. i.d.  $\times$   $\frac{1}{16}$  in., but the subsequent firing caused shrinkage so that permeability measurements were based on final dimensions. Individual samples were fired to maximum sintering temperatures in the range 960°C–1327°C in such a way that a distribution of permeability values was obtained. Results and conclusions have been based on comparison of the spectra obtained for the various samples, no quantitative reference being made to the firing conditions with the exception that the packing factor (relative density) of each sample was determined and used in applying the theory of domain rotation.

We have noted a distinct color change in the material as sintering was carried to higher temperatures. Before sintering, the powder was reddish brown changing gradually to black at the highest firing temperatures.

We attribute this change to an increase in particle size as sintering progresses, and have found by microscopic observation that the most thoroughly sintered samples were made up of grains as large as 2 to 3 microns in diameter. Color changes were not attended by detectable changes in chemical composition.

#### B. Measurements

In determining the magnetic spectra of the various partially sintered toroids, Rado's method<sup>6</sup> was used in the frequency region above 140 megacycles. This is essentially a standing wave technique in which the standing wave pattern of a short-circuited coaxial line is measured with and without a specimen placed next to the short circuit. The shift in position of the voltage minimum gives the real part of the permeability,  $\mu'$ , whereas the standing wave ratios give the imaginary part,  $\mu''$ . Rather than grind the numerous samples to an exact fit with the coaxial line, we have adjusted the measured values of  $\mu'$  and  $\mu''$  according to relations given previously.<sup>7</sup> When corrected for misprints, these relations are

$$\mu'/\mu_0 = [(\mu_m'/\mu_0) \log(D_4/D_1) - \log(D_2/D_1) - \log(D_4/D_3)] [\log(D_3/D_2)]^{-1}, \quad (1a)$$

$$\mu''/\mu' = (\mu_m''/\mu_m') [\log(D_2/D_1) + \log(D_4/D_3) + (\mu'/\mu_0) \log(D_3/D_2)] [(\mu'/\mu_0) \log(D_3/D_2)]^{-1}, \quad (1b)$$

where  $\mu'$  and  $\mu''$  are the corrected values,  $\mu_m'$  and  $\mu_m''$  are the corresponding measured values,  $D_4$  is the inside diameter of the outer conductor of the coaxial line,  $D_3$  is the outside diameter of the sample toroid,  $D_2$  the inside diameter of the sample, and  $D_1$  is the diameter of the inner conductor of the coaxial line.  $\mu_0$  is the permeability of free space.

In the frequency range between 30 Mc/sec and 140 Mc/sec we have used a measuring technique adapted from a method of van der Burgt *et al.*<sup>8</sup> This consists of determining the capacity required to resonate a small section of short-circuited coaxial line ( $L \ll \frac{1}{4}\lambda$ ) with and without a sample toroid at the shorted end. If the  $Q$  of this resonant circuit is also found in both cases,  $\mu'$  and  $\mu''$  can be calculated. The relations are

$$\mu' = (\omega^2 L_B)^{-1} (C_F^{-1} - C_A^{-1}), \quad (2a)$$

$$\mu'' = (\omega^2 L_B)^{-1} [(C_F Q_F)^{-1} - (C_A Q_A)^{-1}], \quad (2b)$$

where  $L_B$ , the inductance of the space occupied by the toroid, is given by  $L_B = 0.4606 h \log(b/a) \times 10^{-6}$  henry, in which  $b/a$  is the ratio of outer to inner diameter of the toroidal specimen and  $h$  is its height in meters. Further,  $C_A$  and  $Q_A$  are the resonant capacity and  $Q$  of the circuit with the specimen absent, and  $C_F$  and  $Q_F$

<sup>7</sup> Massachusetts Institute of Technology Technical Report XXXVI, Laboratory for Insulation Research, July, 1950 (unpublished).

<sup>8</sup> Van der Burgt, Gevers, and Wijn, Philips Tech. Rev. 14, 245 (1953).

are the corresponding values with the toroid in place. In practice, the  $Q$ 's and capacities were measured using a  $Q$  meter with a 3-inch coaxial line suitably mounted at its terminals to receive specimens. For each spectrum an additional measurement was made at 2 Mc/sec using a separate permeameter<sup>9</sup> designed for use at this frequency in conjunction with a  $Q$  meter. Since it is important in all magnetic measurements on ferrites at radio-frequencies to eliminate the possibility of dimensional resonance, we have checked our permeability spectra with samples of thickness from 3 millimeters to 0.5 millimeter with no significant changes in our results.

### RESULTS

In Fig. 1 are shown the spectra of five of the sample toroids sintered at progressively higher temperatures and consequently exhibiting progressively higher packing factors (density of samples relative to that of a single crystal of nickel ferrite). All of the curves show a marked dispersion region occurring around 2000 Mc/sec for the lightly sintered sample and moving to lower frequencies as the sintering temperature is increased. The toroids with packing factor 0.746 and 0.770 have spectra in qualitative agreement with that measured by Rado.<sup>6</sup> However, we have been unable to identify, within the significance of our measurements, the slight absorption peak which occurs in Rado's measurement at around 3000 Mc/sec. The fact that others have observed this peak whereas we do not strongly suggests that the high-frequency peak in this material is dependent upon the nature of the specific sample used and is not an inherent property of nickel ferrite. On the other hand, the broad resonance occurring at some hundreds of Mc/sec is certainly a general characteristic of the material. We have therefore concentrated our attention on the nature of the processes underlying this lower frequency peak.

As proposed by Snoek,<sup>2</sup> the dispersion may be due to the precession of the magnetic moments of domains in an equivalent anisotropy field. This anisotropy field may have contributions from magnetocrystalline anisotropy such as has been found to affect the resonance conditions in single crystals at microwave frequencies, from internal strains in the sintered material, from the internal magnetic fields created by a distribution in direction of crystallites, and from the shape anisotropy of grains. Polder has pointed out<sup>10</sup> further that the relative orientation and interaction of adjacent grains in the material may cause the dynamic demagnetizing factor to differ from the static one, thus shifting the position of the resonance frequency away from a value predicted taking only the other above mentioned anisotropy sources into account. It is very difficult to estimate the magnitudes of these effects in a sintered ferrite, so that it is sufficient to note that the resonances we observed between 200 and 2000 Mc/sec occur at roughly

<sup>9</sup> Made by National Electronics Laboratories, Inc.

<sup>10</sup> D. Polder and J. Smit, *Revs. Modern Phys.* 25, 89 (1953).

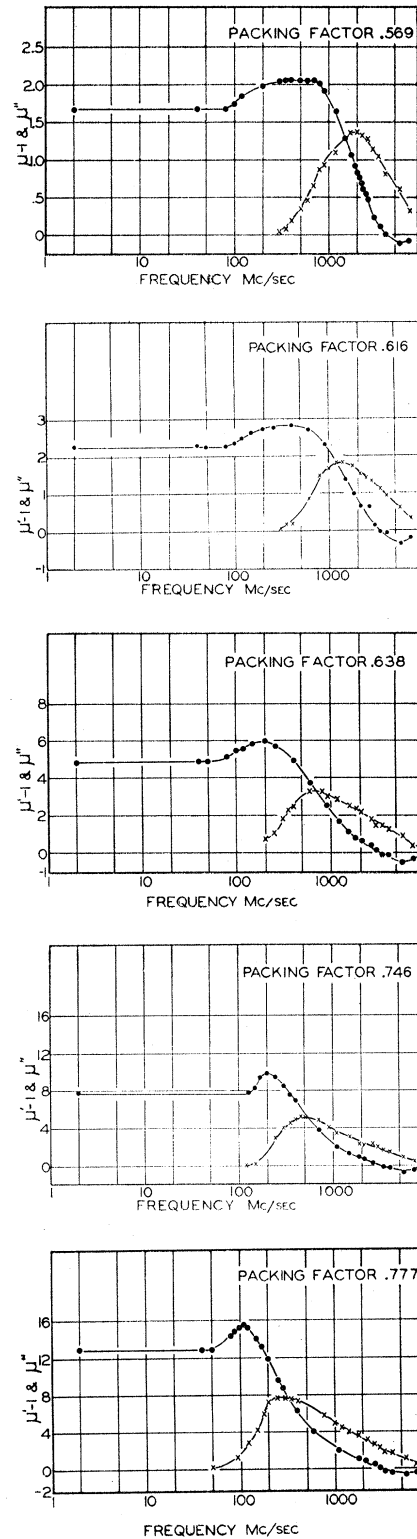


FIG. 1. Permeability spectra of partially sintered nickel ferrite. From top to bottom the five samples represented were sintered at progressively higher maximum temperatures in the range 960-1327°C. The circles represent  $\mu' - 1$  and the crosses represent  $\mu''$ .

TABLE I. Comparison of observed susceptibilities with those calculated from the theory of domain rotation.

$p$	$\omega_r \times 10^9$	$\omega_2 \times 10^9$	$\omega_1 \times 10^9$	$\chi_0/p$ (expt)	$\chi_0/p$ [Eq. (3a)]	$\chi_0/p$ [Eq. (3b)]	Bracket [Eq. (3a)]	Bracket [Eq. (3b)]
0.569	12.6	3.8	0.31	0.23	0.28	0.28	1.0	1.0
0.616	10.7	3.8	0.25	0.29	0.32	0.32	0.99	1.0
0.638	6.0	3.8	0.13	0.60	0.50	0.47	0.87	0.82
0.565	4.4	...	1.0	0.76	...	0.74	...	0.95
0.746	4.4	3.8	0.13	0.83	0.74	0.80	0.95	1.0
0.662	3.5	...	0.75	0.96	...	0.91	...	0.93
0.777	2.5	3.8	1.1	1.39	1.2	1.4	0.85	1.0
0.805	1.9	...	0.75	1.45	...	1.8	...	1.0

the same frequencies as that predicted by an extrapolation of microwave resonance conditions to the case of zero field. On the above basis one would expect a zero-field intercept at around 900 Mc/sec for the case of single crystals of nickel ferrite, or 1400 Mc/sec extrapolating the data of Okamura<sup>11</sup> on sintered samples.

In our analysis we have therefore assumed that the resonance field is determined in some unknown manner by the above factors, but that in every case the position of the resonance frequency is related to the initial susceptibility by a relation, similar to that of Snoek,<sup>2</sup> which has recently been derived by Park.<sup>12</sup> Park's theoretical expression is

$$\omega_r(\chi_0/p) = \frac{2}{3}\gamma_E M_s [(2\omega_1/\omega_2)^{\frac{1}{2}} \ln(\omega_2/\omega_1)], \quad (3a)$$

where  $\omega_r$  = angular frequency at which  $\chi$  reduces to one-half its low-frequency value,  $\chi_0$ ;  $\chi_0 = (\mu' - 1)/4\pi$  at low frequencies;  $p$  = packing factor of the sintered ferrite, i.e., the density of the material relative to that of a single crystal;  $\gamma_E$  = effective value<sup>4</sup> of  $ge/2mc = 19.4 \times 10^6$  (oersted)<sup>-1</sup>;  $M_s$  = saturation moment of nickel ferrite = 265 emu;  $\omega_2$  = angular frequency at which  $\mu' - 1$  is a minimum, and  $\omega_1$  = angular frequency at which  $\mu' - 1$  is a maximum. It can be seen that Eq. (3a) is similar to that given by Snoek, except that porosity of the material is accounted for through  $p$ , and that an additional factor appears on the right-hand side of the equation.

However, in applying Eq. (3a) it is necessary to determine the spectrum up to the frequency at which  $\mu' - 1$  is a minimum (about 6000 Mc/sec) so that it is often more convenient to use a slightly modified form

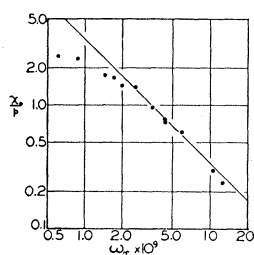


FIG. 2. Comparison of magnetic spectral data on partially sintered nickel ferrites with theory of domain rotation.  $\chi_0 = (\mu' - 1)/4\pi$  is the low-frequency susceptibility,  $p$  is the packing factor, and  $\omega_r$  is the angular frequency where  $\chi = \frac{1}{2}\chi_0$ . The circles give the experimental points, and the solid line is a plot of Eq. (3a) in which the term in brackets has been taken as unity.

<sup>11</sup> Okamura, Torizuka, and Kojima, Phys. Rev. **88**, 1425 (1952).

<sup>12</sup> D. Park, Phys. Rev. **95**, 652(A) (1954). See also the succeeding article [Phys. Rev. **97**, 60 (1954)].

of the expression. In most cases encountered experimentally, it can be shown that  $\omega_r \approx (\omega_1\omega_2)^{\frac{1}{2}}$ , in which case Eq. (3a) becomes

$$\omega_r(\chi_0/p) = \frac{2}{3}\gamma_E M_s \left[ \frac{2\sqrt{2}\omega_1}{\omega_r} \ln(\omega_r/\omega_1) \right]. \quad (3b)$$

Under these conditions it is only necessary to extend the measurement up to  $\omega_r$ , which in the present case would never exceed 2000 Mc/sec.

In Table I are shown experimental values of the various quantities which enter Eqs. (3a) and (3b), as determined from the curves of Fig. 1, along with data from other spectra not shown. In column 5 of the table, the experimental values of  $\chi_0/p$  are to be compared with the calculated values in columns 6 and 7 which have been determined from the two forms of the theory mentioned. It can be seen that the agreement is quite good in both cases, being well within experimental uncertainties. In columns 8 and 9, it is also seen that the approximation used to get Eq. (3b) is quite valid in those cases where our data have permitted use of the more exact expression. It is further interesting to note that the inclusion of a distribution of resonant frequencies among crystallites in the derivation of Eqs. (3), giving rise to the term in brackets, does not strongly modify Snoek's original equation in which such a distribution was not considered, since the bracket never departs from values close to unity.

The inverse proportionality indicated by Eqs. (3), and exhibited by the data, is shown in Fig. 2, where the solid line represents the theoretical expression plotted under the assumption that the term in brackets is in fact unity. For values of  $\chi_0/p$  less than 1.5 the agreement is adequate but is not as satisfactory for larger ordinates, a possible explanation being that the bracketed term of Eqs. (3) could depart strongly from unity in these cases. However, since the spectra associated with the points of highest  $\chi_0/p$  contained no resonant rise of  $\mu' - 1$ , the bracketed term could not be determined.

Inherent in the derivation of Eqs. (3) has been the assumption that the equivalent anisotropy field for resonance is the same as that experienced at very low frequencies. The absence of any appreciable effect of the type proposed by Polder<sup>10</sup> has thus been assumed.

In view of the present agreement between theory and experiment, an agreement obtained without recourse to the Polder effect, it can be concluded that the initial susceptibility in the nickel ferrites we have studied is due principally to domain rotation in an equivalent anisotropy field with little contribution from the dynamic interaction of crystallite magnetic moments.

For comparison we may estimate the resonance conditions to be expected if the initial susceptibility in our material were principally due to the motion of domain walls. Rado *et al.*<sup>13</sup> give an approximate expression for the relation between the susceptibility at low frequencies due to wall motion, which we will call  $\chi_{01}$ , and the resonance frequency of walls, designated here as  $\omega_{01}$ . Their expression can be written as

$$\omega_{01}(\chi_{01}/p)^{\frac{1}{2}} = \gamma_E M_s (8\pi\delta/d)^{\frac{1}{2}}, \quad (4)$$

where  $\delta \sim (kT_c/Ka)^{\frac{1}{2}}$  and  $d$  is an average domain size. Clearly, if we assume that  $\delta$  and  $d$  do not change sufficiently during sintering to alter the right-hand side of (4), the predicted relationship between  $\omega_{01}$  and  $\chi_{01}$  is quite different from the one we have observed experimentally in which  $\omega_0$  and  $\chi_0$  are inversely proportional. We have further observed that  $d$  does not increase in our material to values larger than 2 to 3 microns (assuming that each grain contains one or more domains) in those samples sintered to highest packing factor. For the lightly sintered samples  $d$  was 0.5 to 1.0 micron so that the right side of (4) could not decrease by a factor of more than 2.5 in the sintering process. The anisotropy constant,  $K$ , is the only other quantity in (4) which could change in sintering in such a way as to cause ambiguity in applying (4). In order that wall resonance give an inverse proportion between these two quantities, we find that  $K$  would have to increase by a factor of  $10^4$  which seems quite unlikely.

It is also worthwhile to inquire to what extent our dispersion phenomena may be explained by domain wall relaxation, since the approximate governing equation<sup>13</sup> predicts an inverse proportion between  $\chi_{01}$ ,

and the relaxation frequency,  $\omega_{02}$ . We write Rado's expression as

$$\omega_{02}(\chi_{01}/p) = M_s^2 R/\eta, \quad (5)$$

where  $R = \delta/d$ ,  $\eta = \beta\gamma\delta$ , and  $\beta$  is the wall velocity parameter. Inserting  $\beta = 0.026$ ,<sup>14</sup> we obtain  $M_s^2 R/\eta \sim 2 \times 10^{10}$ . Inasmuch as our experimental results indicate a  $\omega_0\chi_0$  product of around  $2 \times 10^9$ , it seems unlikely that domain wall relaxation can contribute strongly. Further qualitative evidence in agreement with this conclusion is obtained by noting the distinct rise in  $\mu' - 1$  which occurs in most of our samples just below the region of maximum absorption; this aspect of our curves suggests a resonance process rather than a pure relaxation.

### CONCLUSIONS

The experiment described above has led to the conclusion that the initial permeability in our samples is due principally to domain rotation. This result is probably due to the fact that the crystallite size in the materials studied was restricted to values small enough to inhibit the formation and motion of domain boundaries in the usual sense. In this respect our samples exhibit pronounced single domain behavior.

It seems hardly necessary to emphasize that the results of our experiment do not preclude the existence of domain wall motion as an important model in understanding the initial permeability in certain ferrites. Nor can contributions from wall motion be completely excluded in the present case. Our experiment presents, however, an example of a situation in which, because of the relatively small size of crystallites in the sintered material, the principal contribution to initial permeability comes from domain rotation.

We wish to acknowledge our debt to David Park and J. Kenneth Moore for continued discussions and suggestions on all phases of the work and to V. R. Abate for help in making the measurements.

<sup>13</sup> Rado, Wright, and Emerson, Phys. Rev. **80**, 273 (1950).

<sup>14</sup> J. K. Galt and H. G. Hopper, Revs. Modern Phys. **25**, 93 (1953).