The Permalloy Problem

R. M. Bozorth

Bell Telephone Laboratories, Inc., Murray Hill, New Jersey

In attempting to explain the unusual magnetic properties of the iron-nickel alloys, single crystals of alloys containing 35 to 100 percent nickel were prepared, and measurements made of the magnetic crystal anisotropy and magnetostriction as dependent on cooling rate. It is confirmed that there is a large effect of cooling rate on the anisotropy in the region near $FeNi_3$, but the experiments show also a substantial effect between 50 and 85 percent nickel. Two magnetostriction constants, λ_{100} and λ_{111}, were measured on the same crystals. The effect of cooling rate on magnetostriction was found to be substantial only in the composition range 70 to 80 percent nickel. When the specimens are quenched, λ_{111} goes through zero for a nickel content just below 80 percent nickel, a composition very close to that for highest permeability. This is understandable because the magnetostrictive strain caused by movement of the boundary between two domains, each magnetized spontaneously in a [111] direction, depends on λ_{111} alone. The same physical picture predicts that near 45 percent nickel, where [100] is the direction of easiest magnetization and λ_{100} goes through zero, the permeability *vs* composition curve should again have a maximum. Such a maximum is known to exist, and initial permeabilities as high as 15 000 have been observed.

Although simple theory suggests that domain-rotation should occur in very weak fields when the crystal anisotropy is very small (75 percent nickel in quenched alloys), nevertheless, rotation involves magnetostrictive strains which prevent μ_0 from becoming infinite. Internal poles are also likely to be formed. In slowly cooled alloys the anisotropy is zero at about 63 percent nickel; here there are random strains caused by magnetostriction and possibly also by atomic ordering.

The principal changes in magnetic properties with composition are explained in terms of the crystal anisotropy and magnetostriction, and their change with heat treatment.

INTRODUCTION

The "Permalloy problem" is to explain the characteristic magnetic properties possessed by the binary iron-nickel alloys containing 35 to 100 percent nickel. They have unusually high permeabilities which are a maximum at about 79 percent nickel, the permeabilities are influenced markedly by heat treatment and specifically are increased by cooling rapidly, and a magnetic anneal (cooling in the presence of a magnetic field) causes drastic changes in various magnetic properties, particularly the maximum permeability and the form of the hysteresis loop. The present investigation of the magnetic crystal anisotropy, and of the magnetostriction of single crystals of these alloys, is a contribution to the explanation of the behavior of these alloys as dependent on composition and heat treatment.

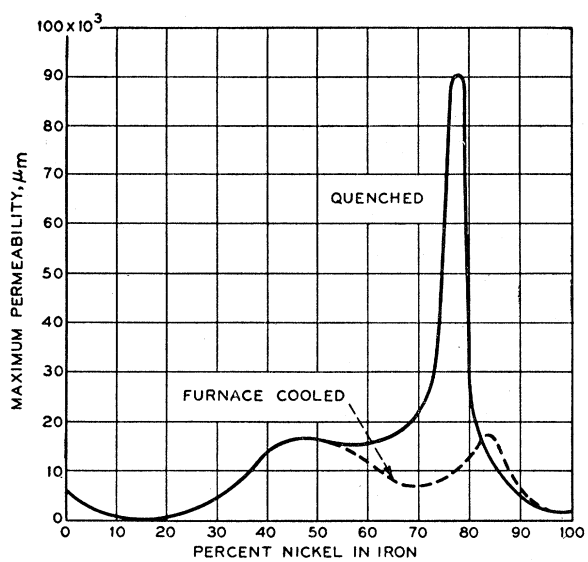

Fig. 1. Maximum permeabilities of iron-nickel alloys. Curves show mean values of many measurements taken over a period of years.

These properties are illustrated in Figs. 1–3, which were drawn some time ago and represent mean values of many data.[1] In addition to the maxima of the permeabilities at about 79 percent nickel (Figs. 1 and 2), one should note the secondary maxima at about 45 percent nickel, which we will refer to later, and the shift of the 79 percent maximum with heat treatment. The change in the form of the loop with heat treatment in a magnetic field, and the associated change in maximum permeability, are drastic; there is practically no corresponding change in initial permeability.

In previous attempts to explain these phenomena, McKeehan[2] first stated a qualitative relation between the high permeability and the low magnetostriction. Kersten[3] derived an expression for the highest initial permeability attainable in a material which has residual magnetostrictive strains. Both McKeehan and Kersten used the magnetostriction of the polycrystalline material, which passes through zero at 81 to 83 percent nickel. Lichtenberger[4] found that the composition for zero magnetostriction is different for the [100], [110], and [111] directions. Also, he noted that the permeability maximum at 79 percent lies between the composition for zero polycrystalline magnetostriction and the composition for zero crystal anisotropy.

Using the ideas of domain theory which have been developed during recent years, we expect that when the crystal anisotropy is zero and imperfections are absent,

[1] See, for example, R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., New York, 1951).
[2] L. W. McKeehan, Phys. Rev. **28**, 158 (1926).
[3] M. Kersten, Z. tech. Physik **12**, 665 (1931). See also R. Becker and Döring, *Ferromagnetismus* (Springer Verlag, Berlin, 1939).
[4] R. Lichtenberger, Ann. Physik **15**, 45 (1932).

domain rotation will occur with great ease and the permeability will be very high. Also, when the magnetostriction is zero, displacement of domain boundaries will occur with negligible resistance, and the permeability will also be high. The work summarized below was carried out in order to subject this physical picture to a quantitative test and to find out if possible the separate roles of anisotropy and magnetostriction. This necessitated determination of the crystal anisotropy constants, and of the magnetostriction at saturation in the principal crystal directions, in single crystals subjected to various heat treatments, that is, various rates of cooling.

A qualitative theory of heat treatment in a magnetic field was suggested by the author some years ago.[5] This dealt with the relief of magnetostrictive strains by diffusion, in material of low crystal anisotropy. Further consideration can now be given to this theory, using the results of the present investigation as a background.

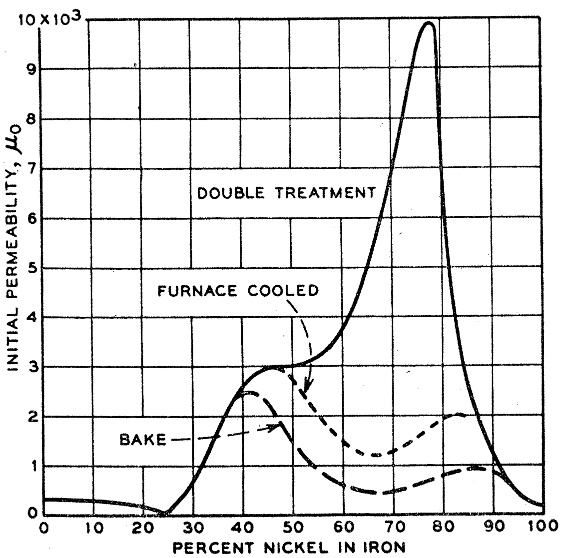

FIG. 2. Initial permeabilities of iron-nickel alloys, averaged over many measurements.

Determinations of crystal anisotropy and saturation magnetostriction in various crystal directions will now be described.

CRYSTAL ANISOTROPY

Single crystals were prepared by slow cooling of the melt, as described by Walker, Williams, and Bozorth.[6] Specimens were cut in the form of disks, usually in the (100) plane, and were normally about 1 cm in diameter and 2 mm thick. Accuracy of orientation was a few tenths of a degree. After annealing, all specimens were cooled at two standard rates: (1) they were withdrawn

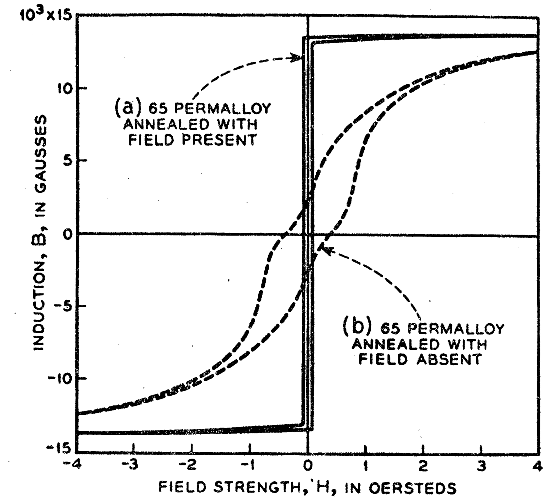

FIG. 3. Hysteresis loops of 65 Permalloy, showing effect of heat treatment in a magnetic field.

from the furnace at about 700°C and placed quickly on a cold copper plate (quenched), or (2) they were automatically cooled from above 600° to below 300°C at 2°C/hr (baked). The known order-disorder transformation of $FeNi_3$ is at 506°C. Presumably (1) gives a high degree of disorder, (2) a high degree of order, but some of our experiments indicate that a definitely higher degree of order is obtained by cooling at 1°C/hr.

Measurements, already reported briefly,[7] were made by the use of a sensitive torsion magnetometer designed by Mr. H. J. Williams, with which a torque of 6 dyne-cm could be readily detected. Details of the measurements will be reported elsewhere.

The results are shown in Fig. 4. It is noted at once that the slowly cooled alloys have lower (more negative) anisotropy constants, especially in the composition range near $FeNi_3$ (76 percent nickel), at which a deep minimum occurs. This confirms measurements previously reported by Grabbe.[8] Atomic ordering was detected by x-rays and by neutron diffraction in the slowly cooled alloys containing 74 and 68 percent nickel but not in the quenched alloys or in a slowly cooled specimen containing 90 percent nickel. It seems probable that some ordering occurs in alloys containing as little as 40 percent nickel but not in those containing 90 percent or over.

The alloy containing 74 percent nickel was cooled at various rates, with the results shown in Fig. 5. This shows that the degree of ordering is still increasing as the cooling rate falls below 2°C/hr.

The composition for zero anisotropy varies from 63 to 75 percent nickel, depending on the cooling rate. It has previously been pointed out that a low anisotropy is related to other magnetic phenomena, for example, low

[5] R. M. Bozorth and J. F. Dillinger, Physics 6, 285 (1935); see also J. S. Marsh, *Alloys of Iron and Nickel* (McGraw-Hill Book Company, Inc., New York, 1938), pp. 214–219.
[6] Walker, Williams, and Bozorth, Rev. Sci. Instr. **20**, 947 (1949).
[7] R. M. Bozorth and J. G. Walker, Phys. Rev. **83**, 871 (1951).
[8] E. M. Grabbe, Phys. Rev. **57**, 728 (1940).

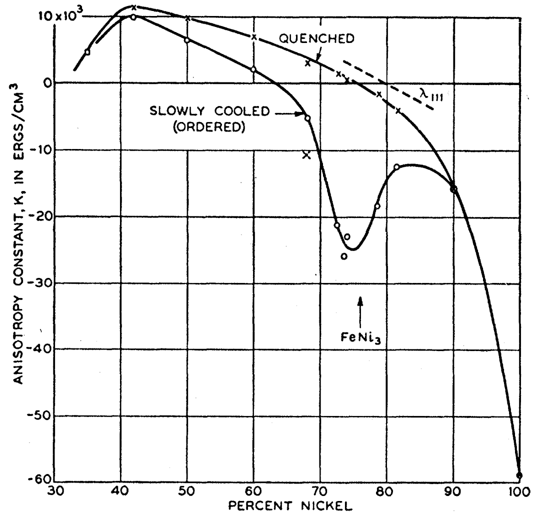

FIG. 4. Crystal anisotropy constants of quenched and of slowly cooled alloys. Broken line λ_{111} indicates the composition at which the magnetostriction in the [111] direction goes through zero.

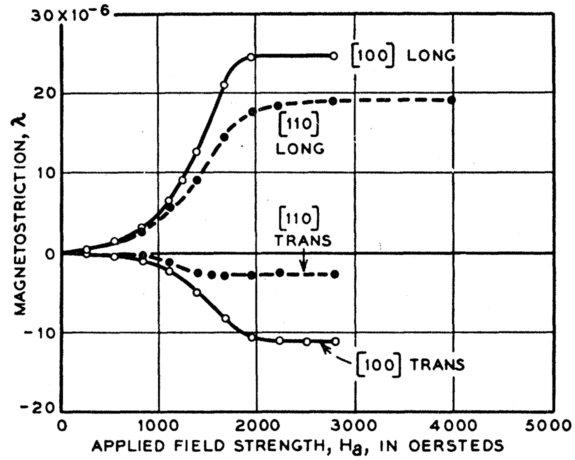

FIG. 6. Longitudinal and transverse magnetostriction of 68 percent nickel crystal, measured in [100] and [110] directions, as dependent on external applied field. Ratio of diameter to thickness of crystal, $m=4.4$.

remanence,[9] high strain sensitivity,[10] and susceptibility to magnetic anneal.[11] More quantitative tests of these relations can now be made.

MAGNETOSTRICTION

Magnetostriction was determined on the same disks that were used for anisotropy measurements, using strain gauges according to the method described by Goldman.[12] Gauges, usually of $\frac{1}{8}$-in. size, were oriented in the [100] and [110] directions in the (001) plane. A magnetic field of high uniformity and of strength up to 5000 oersteds was applied either parallel (longitudinal effect) or at right angles (transverse effect) to the gauge. Details of the method will be published elsewhere.

Figures 6 to 8 show the magnetostriction of three specimens as dependent on magnetizing field strength, as measured longitudinally and transversely in the [100] and [110] directions. Figure 6 shows that saturation is easily attained in the 68 percent nickel specimens. Figures 7 and 8 show a continually increasing length with increasing field with both longitudinal and transverse measurements. This is definitely attributed to volume magnetostriction, which is known to persist to very high fields. The volume magnetostriction, calculated from the slopes of the λ *vs* H lines in the high fields used, is in reasonably good accord with previous measurements by others. The existence of volume magnetostriction does not interfere with the desired measurement of λ_{100} and λ_{111} for reasons indicated below.

Using the 2-constant formula[1] for magnetostrictive

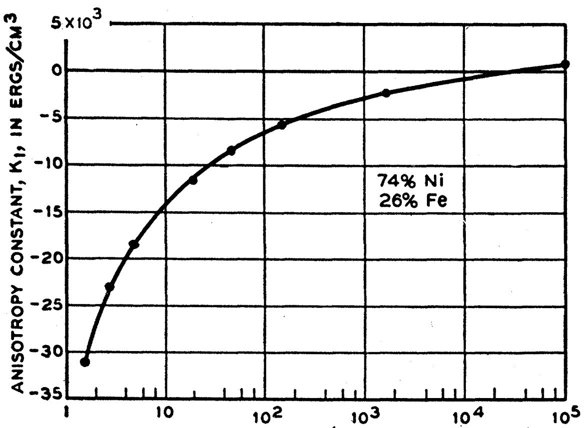

FIG. 5. Anisotropy constants of 74 percent nickel alloy as dependent on rate of cooling from 600° to 300°C. Highest cooling rate is a rough estimate only.

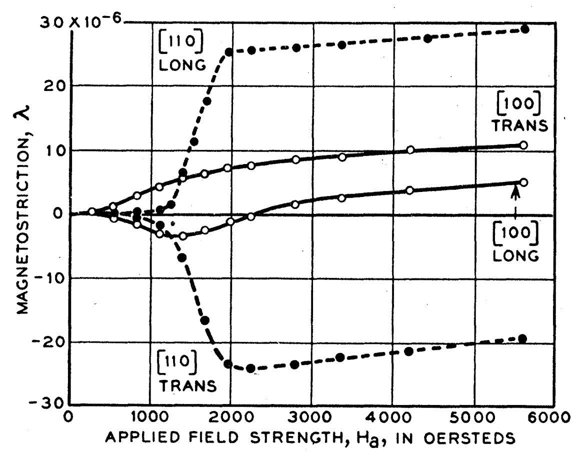

FIG. 7. Magnetostriction of 42 percent nickel crystal, $m=5.5$.

[9] R. M. Bozorth, Z. Physik **124**, 519 (1948).
[10] R. M. Bozorth and H. J. Williams, Revs. Modern Physics **17**, 72 (1945).
[11] J. F. Dillinger and R. M. Bozorth, Physics **6**, 279 (1935); see also reference 4.
[12] J. E. Goldman and R. Smoluchowski, Phys. Rev. **75**, 140 (1949).

change in length at saturation, we have

$$\lambda=(3/2)\lambda_{100}(\alpha_1{}^2\beta_1{}^2+\alpha_2{}^2\beta_2{}^2+\alpha_3{}^2\beta_3{}^2-1/3)$$
$$+3\lambda_{111}(\alpha_1\alpha_2\beta_1\beta_2+\alpha_2\alpha_3\beta_2\beta_3+\alpha_3\alpha_1\beta_3\beta_1),$$

in which the α's are the direction cosines of the magnetization with respect to the crystal ones, and the β's are the direction cosines of the measured change in length; and it is apparent that the difference between the longitudinal (λ_l) and the transverse (λ_t) changes in length in the [100] direction gives λ_{100}:

$$\lambda_l-\lambda_t=(3/2)\lambda_{100},\quad \text{for } [100].$$

Here $\beta_1=1$, $\beta_2=\beta_3=0$; $\alpha_1=1$, $\alpha_2=\alpha_3=0$ or $\alpha_2=1$, $\alpha_1=\alpha_3=0$. Not quite so obviously, the corresponding changes in length in the [110] direction give λ_{111}:

$$\lambda_l-\lambda_t=(3/2)\lambda_{111},\quad \text{for } [110].$$

Here $\beta_1=\beta_2=1/\sqrt{2}$, $\beta_3=0$; $\alpha_1=\alpha_2=1/\sqrt{2}$, $\alpha_3=0$ or $\alpha_1=-1/\sqrt{2}$, $\alpha_2=1/\sqrt{2}$, $\alpha_3=0$.

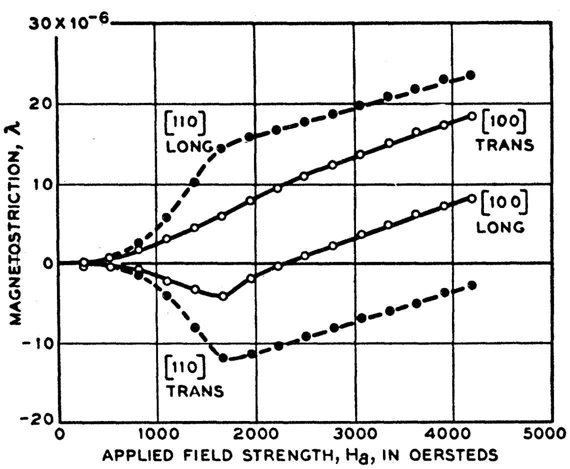

FIG. 8. Magnetostriction of 35 percent nickel crystal, showing large volume effect; $m=3.9$.

Values of λ_{100} and λ_{111} for crystals heat treated in the two ways (quenched on a cold copper plate or cooled at 2°C/hr) are shown in Fig. 9. It is apparent that the magnetostriction varies with heat treatment in a narrower composition range than was observed for the anisotropy, namely, from about 68 to 81 percent nickel. The measurements for the quenched alloys agree tolerably well with the results of Lichtenberger,[4] but the new measurements are somewhat higher in the range 40 to 50 percent nickel. The ordered alloys are more nearly isotropic than the disordered alloys, and at about 73 percent nickel λ_{100} and λ_{111} are almost equal. Data for the range 60 to 90 percent nickel are shown on a larger scale in Fig. 10.

It will be shown later that the composition at which λ_{111} equals zero is of special importance for the theory of Permalloy. According to the data just presented, this composition is slightly greater than 80 percent nickel for the quenched alloys. However, consideration

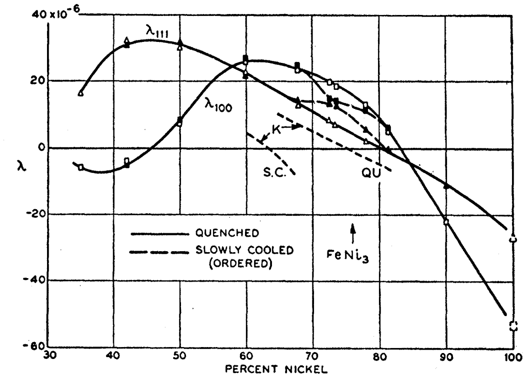

FIG. 9. Magnetostriction constants λ_{100} and λ_{111}, showing dependence on cooling rate. Broken curves show compositions at which crystal anisotropy goes through zero, for quenched (QU) and slowly cooled (S.C.) alloys.

of the 5-constant formula[1] for magnetostriction:

$$\lambda_s=h_1(\alpha_1{}^2\beta_1{}^2+\alpha_2{}^2\beta_2{}^2+\alpha_3{}^2\beta_3{}^2-1/3)$$
$$+2h_2(\alpha_1\alpha_2\beta_1\beta_2+\alpha_2\alpha_3\beta_2\beta_3+\alpha_3\alpha_1\beta_3\beta_1)$$
$$+h_4(\alpha_1{}^4\beta_1{}^2+\alpha_2{}^4\beta_2{}^2+\alpha_3{}^4\beta_3{}^2+2s/3-1/3)$$
$$+2h_5(\alpha_1\alpha_2\alpha_3{}^2\beta_1\beta_2+\alpha_2\alpha_3\alpha_1{}^2\beta_2\beta_3+\alpha_3\alpha_1\alpha_2{}^2\beta_3\beta_1$$
$$+h_3(s-1/3)\text{ for nickel, }+h_3s\text{ for iron,}$$
$$s=\alpha_1{}^2\alpha_2{}^2+\alpha_2{}^2\alpha_3{}^2+\alpha_3{}^2\alpha_1{}^2,$$

shows that

$$\lambda_{111}=2h_2/3+2h_5/9,$$

whereas the last term is neglected in the 2-constant formula. Obviously the constant h_5 might possibly make an important contribution. (If we take the five constants determined[1] by Döring from the data of Masiyama for nickel, we have

$$2h_2/3=-31\times10^{-6},$$
$$2h_2/3+2h_5/9=-20\times10^{-6},$$

a large discrepancy between the results of the 2-constant

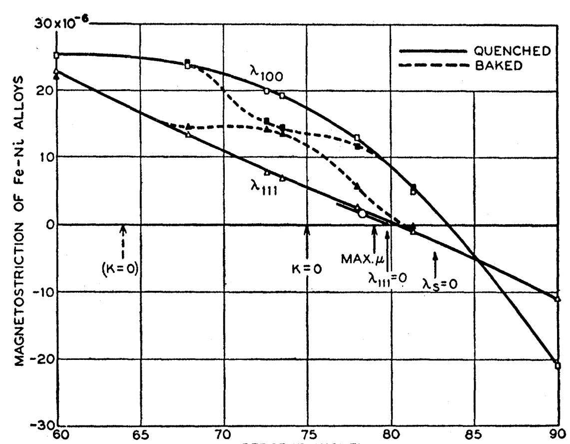

FIG. 10 Magnetostriction constants for range 60 to 90 percent nickel. Single point shows λ_{111} as determined with greater accuracy for a specimen containing 0.5 percent manganese.

and 5-constant formulas.) Therefore, it seemed necessary to determine both h_2 and h_5 in order to be sure of λ_{111}. Moreover, since most of the data on the magnetic properties of the Permalloys were taken for specimens which contained 0.5 percent of manganese, a single crystal containing 78 percent nickel and 0.5 manganese was prepared and a disk cut from it oriented accurately parallel to a (011) plane.

The method used for evaluating the five magnetostriction constants has been reported briefly by Bozorth and Hamming,[13] and will be described in detail elsewhere. As a matter of fact, h_5 in 78 percent nickel is found to be negligibly small. Results for the quenched specimens are $h_1=13$, $h_2=2.6$, $h_3=-0.3$, $h_4=-0.1$, all $\times 10^{-6}$. Therefore,

$$\lambda_{111}=1.7\times 10^{-6}.$$

This point is plotted with a large circle in Fig. 10. It lies close to the λ_{111} curve previously drawn, and a new line drawn parallel to the old crosses the axis at

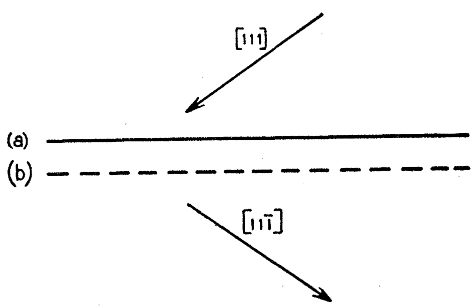

FIG. 11. Ideal domain boundary separating [111] and [11$\bar{1}$] domains. No poles are formed at boundary since the normal component is constant across the boundary. The strain produced by boundary movement depends only on λ_{111}.

about 79.5 percent nickel. This is very close to the composition for highest permeability.

Incidentally, it may be pointed out that h_3 is a measure of the volume change associated with domain rotation. This is the first time that such a volume change has been measured. However, its value is small and may not exceed the probable error.

INTERPRETATION OF RESULTS

High permeability in material free from imperfections may be expected to result either from domain rotation against weak crystal anisotropy forces or from boundary displacement when the opposing force is small, as it may be expected to be when the magnetostriction is small. It should be pointed out that the magnetostriction which opposes boundary movement is that in the *easy crystallographic direction*, namely, λ_{111} when [111] is the direction of easy magnetization, as it is in the 79 percent nickel alloy. It was for this reason that care was taken in the determination of λ_{111}, as mentioned above.

[13] R. M. Bozorth and R. W. Hamming, Phys. Rev. **87**, 209 (1952).

According to domain theory,[1] as developed by Becker and others, the rotation of magnetization by the field, against the forces of crystal anisotropy, gives the following formula for the initial permeability:

$$\mu_0-1=2\pi\,\overline{\sin^2\theta}(I_s^2/K),$$

θ being the smallest angle between the magnetization and a crystal axis, or

$$\mu_0\approx 2I_s^2/K.$$

A formula for μ_0 for the moving boundary process has been given by Becker and Döring[3] in terms of internal strains σ_i. Putting $\sigma_i=\lambda E$, as Kersten has proposed, we have

$$\mu_0\approx 5I_s^2/(\lambda^2 E),$$

E being Young's modulus and λ being the saturation magnetostriction in the direction of easy magnetization.

According to simple theory one might expect the initial permeability to become infinite at the two compositions at which K and λ are zero. This is obviously not the case. In the quenched alloys the lack of a maximum at $K=0$ (75 percent nickel) can be accounted for by the presence of residual internal strains caused by magnetostriction, the existence of which was pointed out by Kersten. These will restrict rotation in the same way that crystal anisotropy will, and the resulting initial permeability may be estimated by replacing K by $\lambda^2 E$. If we use the λ for the polycrystalline material λ_s, then for 75 percent nickel we have

$$\mu_0\approx 2I_s^2/\lambda_s^2 E\approx 9000$$

as the highest μ_0 we should expect, provided the full magnetostrictive strains remain in the material at room temperature. If they are relieved somewhat by diffusion during the cooling of the material, μ_0 can be larger, and if imperfections are present μ_0 is less.

In a similar way the calculated limit of the initial permeability of the slowly cooled alloy having $K=0$ (63 percent nickel) is 2000 or more. This fits rather well with a measured value[11] of 3000 in 65 percent nickel that had been carefully purified in hydrogen, a treatment that increases the permeability of 45 percent nickel from 3000 to 15 000, according to measurements of Williams.[14] It is not necessary to invoke any strains of atomic ordering in order to account for the permeability observed.

Therefore, reduction of the anisotropy to zero does not lead to high permeabilities (rotation process) unless the magnetostriction also approaches zero. However, disappearance of the magnetostriction in the direction of easy magnetization can cause high permeability (moving boundary) even though the anisotropy is not zero. The importance of λ_{111} in a material for which [111] is the direction of easy magnetization is illustrated in Fig. 11, which is particularly applicable to

[14] H. J. Williams, reference 1, pp. 122–123.

the understanding of maximum permeability (see below). The magnetostriction strain in the material caused by movement of the boundary from (a) to (b) is found by calculation, using either the 2-constant or the 5-constant formula, to be a shear strain determined entirely by λ_{111}. A similar calculation shows that only λ_{100} is involved when [100] is the direction of easy magnetization. Becker and Döring[3] have already proposed that the initial permeability is controlled by the magnetostriction in the direction of easy magnetization (λ_{100} or λ_{111}).

Some suggestions regarding the effect of annealing in a magnetic field were proposed by the author at an earlier date,[5] and it appears at the present time[15] that the proposed explanation, of the "freezing in" of magnetostrictive strains, is basically correct. However, one of the features of the phenomenon that has not been explained is that the initial permeability remains unchanged by the treatment while the maximum permeability is increased by a large factor. This may be understood by reference to Fig. 12. Let the solid curve represent the local strain (σ) from point to point (x) in the material, and let P represent the position of a wall before application of a field, according to the representation of Becker and Döring.[3] The initial permeability is determined by the form of the σ *vs* x curve near P. When the field is increased toward that corresponding to maximum permeability, so that the wall moves over the top of the strain "hill," it will move freely until a higher hill is encountered, as shown by the horizontal solid line. This occurs when the strain is not increased by boundary movement, a situation that takes place with a 180° wall, or when the magnetostriction in the direction of easy magnetization is zero. Ordinarily materials contain walls of both kinds, but in materials that have responded to heat treatment in a magnetic field only 180° walls are present.[16] However, when walls of both kinds occur, movement of the 90° walls (or 70° or 110° when $K<0$) builds up a strain, as shown in Fig. 11 and as represented in Fig. 12 by the line S. When this strain is added to the local strain we obtain the dotted curve. In this case the wall movement is limited by strains of random amplitude more severely than it is when only 180° walls exist, and in the latter case corresponding to the material heat-treated in a field, the maximum permeability is therefore higher.

A different explanation of the increase in maximum permeability of material heat treated in a magnetic field can be based on the existence of Néel "spikes."[17] Williams and Shockley[18] have shown that in silicon-iron, where there are six directions of easy magnetization, these spikes exert a pull on neighboring domain walls and so contribute to the coercive force. However, in magnetically annealed material in which there are only two directions of easy magnetization[16] the geometry does not permit the existence of spikes of this kind, and so the retarding action is absent and the coercive force is smaller and the maximum permeability larger.

Let us now compare the experimental results with the predictions of theory:

(1) In the quenched alloys the permeability is a maximum very near the composition at which $\lambda_{111}=0$. The theory also requires that near 45 percent nickel, when [100] is the direction of easy magnetization and λ_{100} is zero, we should expect another maximum in the μ_0 *vs* composition curve. Such maxima are apparent in Figs. 1 and 2, though they are not as pronounced as they are near 79 percent nickel. This may be because K is larger at 45 than at 79 percent nickel, and consequently, domain rotation will not contribute as much toward the permeability. In alloys purified by treatment in hydrogen, however, Williams[14] has observed an initial permeability of 15 000.

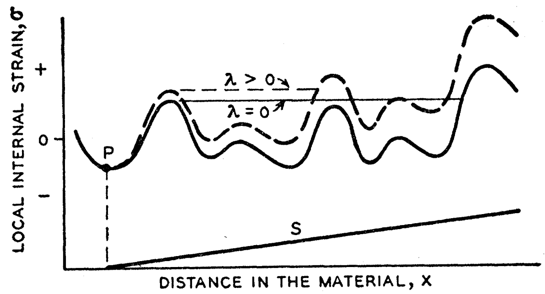

FIG. 12. Diagram of internal strains in material having zero magentostriction or having only 180° boundaries (solid curve), or in material in which stress is developed by boundary movement (dotted curve), showing different magnitudes of maximum permeability expected.

(2) In slowly cooled alloys as compared with quenched alloys, the optimum composition is shifted toward higher nickel content, in agreement with the shift in composition for $\lambda_{111}=0$.

(3) The higher permeabilities of the quenched alloys appear to result from the smaller anisotropy of the latter (about 10 times smaller) at the composition at which $\lambda_{111}=0$. The same reason appears to exist for the lower permeability at 45 percent nickel than at 79 percent nickel, as mentioned above under (1). In slowly cooled alloys the maximum (and initial) permeabilities of the 45 and 80 percent nickel alloys are comparable. Also, the magnitudes of the anisotropy constants are about the same (positive in the one case, negative in the other).

(4) Two mechanisms have been suggested, both of which are probably applicable, for the increase of the maximum permeability of iron-nickel alloys by heat treating in a magnetic field.

[15] E. C. Stoner, Repts. Prog. Phys. **13**, 83–183 (1950), especially pp. 167–8.
[16] H. J. Williams and M. Goertz, J. Appl. Phys. **23**, 316 (1952).
[17] Williams, Bozorth, and Shockley, Phys. Rev. **75**, 155 (1949).
[18] H. J. Williams and W. Shockley, Phys. Rev. **75**, 178 (1949).

The general course of the permeability *vs* composition curves of the Permalloys, and their changes with heat treatment, can be understood in a qualitative way in terms of the magnetostriction and crystal anisotropy, when we note the importance of the magnetostriction in the direction of easy magnetization, and of the internal fields associated with the rotation process.

This work was carried out with the able assistance of J. G. Walker in the preparation of the crystals. The author is also indebted to Mrs. M. H. Read for the detection of atomic ordering by x-rays, and more recently to Dr. L. Corliss, Dr. J. M. Hastings, and Dr. R. J. Weiss of the Brookhaven National Laboratory for observation of atomic ordering in one of the crystals by neutron diffraction. Finally, the benefit of discussions with H. J. Williams, J. K. Galt, and E. A. Nesbitt is gratefully acknowledged.