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HEAT TREATMENT OF MAGNETIC MATERIALS IN A MAGNETIC FIELD--I

BY

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A SURVEY OF THE MAGNETIC PROPERTIES
OF A SERIES OF IRON-COBALT-NICKEL ALLOYS
WHEN ANNEALED IN A MAGNETIC FIELD

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Heat Treatment of Magnetic Materials in a Magnetic Field

I. Survey of Iron-Cobalt-Nickel Alloys

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The changes that occur in the magnetic properties of iron-cobalt-nickel alloys when they are annealed in a magnetic field, have been investigated for a series of these alloys. The maximum change for the iron-nickel alloys occurs between 65 and 70 per cent nickel and is evidenced by a large increase in maximum permeability and a hysteresis loop of rectangular shape. All of the alloys with Curie points above 500° C. and with no phase transformation have their properties similarly changed. Thorough preliminary annealing enhances the effect. With an extreme preliminary anneal of 1400° C. for 18 hours specimens of 65 permalloy have been obtained with the record value of maximum permeability of 600,000. The magnetic characteristics of materials treated in this way are relatively insensitive to stress. These magnetic characteristics are, however, highly anisotropic; the maximum permeability in one direction is as much as 150 times as large as that at right angles.

INTRODUCTION

IT has been known for many years[1] that the magnetic properties of iron and silicon steel are somewhat changed by heat treating in a magnetic field. During the early experiments with perminvar[2] it was found[3] that the constant permeability for which this material is best known could be obtained only when there was no field present during the heat treatment, and that when a field of one œrsted was maintained during annealing the maximum permeability at room temperature was 136,000 instead of the usual value of 1500. More recently, during experiments on the Barkhausen effect in straight fine wires (0.03 mm. in diameter) it was found that the hysteresis loops for these specimens had square corners and vertical sides. Investigation showed that these properties were caused by the presence of a magnetic field in the furnace in which they were heat treated. At that time the experiments of Kelsall[4] were known to us and it was decided to make a more thorough study of the effect of heat treating alloys in a magnetic field.

Accordingly we have performed the experiments described below,

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[1] H. Pender and R. L. Jones, *Phys. Rev.*, **1**, 259 (1913).

[2] G. W. Elmen, *J. Frank. Inst.*, **206**, 317 (1928).

[3] P. P. Cioffi, U. S. Patent 1708936 (1929).

[4] G. A. Kelsall, *Physics*, **5**, 169 (1934).

mainly on iron-cobalt-nickel alloys, and have made a rather detailed study [5] of two of them. We have found that the magnetic properties of many of these alloys are greatly changed by heat treatment in a magnetic field. In some specimens the maximum permeability is increased from 5000 to 300,000, in others from 20,000 to 600,000.

PROCEDURE

The samples tested were rings, 8 cm. in diameter, 0.015 cm. in radial thickness and 0.32 cm. wide, made from one turn of tape overlapped about 0.4 cm. and spotwelded. The tape was fabricated as described by Elmen.[2] These samples were annealed in hydrogen in a metal pot. Commercial hydrogen, filtered through hot palladium-asbestos to remove the free oxygen, was passed slowly through the pot during the anneal.

The samples were first annealed at 1000° C. for one hour and cooled in the furnace at a maximum rate of about 300° C./hour. Each sample was then placed in a toroidal lavite box and wound with deoxidized copper wire for magnetizing. This assembly was then given a second anneal in hydrogen at a maximum temperature of about 50° C. above the Curie point of the specimen. This temperature was maintained for one hour and then the sample cooled in the furnace as before. While the sample was cooling from this second anneal a magnetizing field of 16 œrsteds was applied.

After annealing each sample was placed in a toroidal box and two windings applied for magnetic measurements. These were made with a Haworth fluxmeter [6] and hysteresis loops were photographically recorded with the same instrument.

MAGNETIC CHARACTERISTICS

In Fig. 1, curve (a) gives the maximum permeabilities of a series of iron-nickel alloys cooled in a magnetic field as described above. Curves (b) and (c) give for comparison permeabilities obtained [7] when similar alloys are slowly cooled or rapidly cooled without an applied field. The dotted curve (d) gives the Curie points of the iron-nickel alloys here used. It is apparent that curves (a) and (d) have maxima between 65 and 70 per cent nickel, and that $\mu_{\max}$ for alloys with low Curie points is not noticeably changed by cooling in a magnetic field.

We have also found that the magnetic characteristics of many of the iron-cobalt-nickel alloys are changed when they are cooled in a mag-

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[5] See the succeeding paper, this issue.

[6] F. E. Haworth, *Bell Sys. Tech. J.*, **10**, 20 (1931).

[7] G. W. Elmen, *J. Frank. Inst.*, **207**, 583 (1929), and more recent data by G. A. Kelsall.

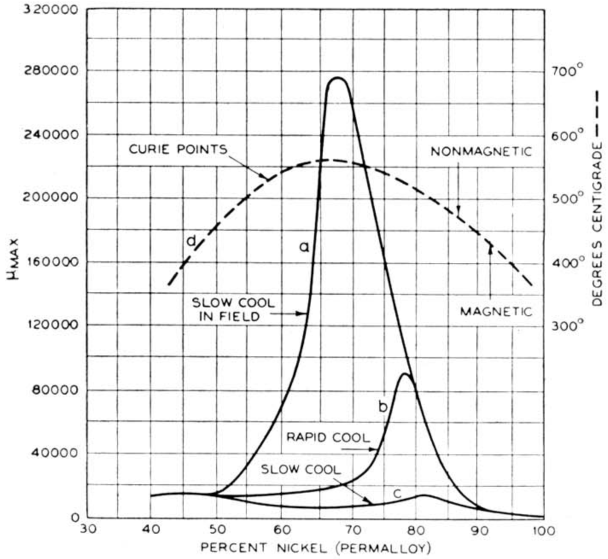

Fig. 1—The maximum permeabilities of iron-nickel alloys cooled in different ways after annealing at 1000° C. Curie points of the same alloys are shown by the dashed line.

netic field. All of the alloys tested which have compositions lying within the outlined area shown in Fig. 2 are affected by such treatment. Some compositions outside of this area near the iron-cobalt line are still under investigation. Of these iron, and the alloy with 48 per cent Fe and 52 per cent Co, show some changes. The points on the diagram indicate the alloys tested.

Here again it was found that alloys with high Curie points are affected by cooling in a magnetic field. Curves (a) and (b) in Fig. 2 indicate alloys with Curie points of 500° C. Curves (c) and (d) indicate alloys with phase transformations near room temperature.

Table I gives the values of coercivity, H_c, and maximum permea-

TABLE I

MAGNETIC CHARACTERISTICS OF IRON-COBALT-NICKEL ALLOYS

Composition	Annealed Without a Field		Annealed With a Field	
Fe-Co-Ni	H_c	μ_{max}	H_c	μ_{max}
10–20–70.......	1.16	1,490	0.28	11,600
10–39–51.......	2.54	2,440	0.69	7,100
11–59–30.......	3.86	1,110	2.28	5,300
11–78–11.......	3.56	1,730	2.54	4,100
30–25–45.......	0.90	2,000	0.09	115,000
30–44–26.......	2.90	1,150	0.74	12,500
45–10–45.......	0.69	7,550	0.44	18,300
48–52..........	0.70	11,000	0.55	22,400

bility, $\mu_{\max}$, of representative alloys. The values for the identical tape annealed without an applied field were kindly furnished by G. A. Kelsall. The conventional magnetic characteristics given in Table I do not, however, give a complete picture of the peculiar cyclic magnetic properties of these alloys, which are shown by their hysteresis loops.

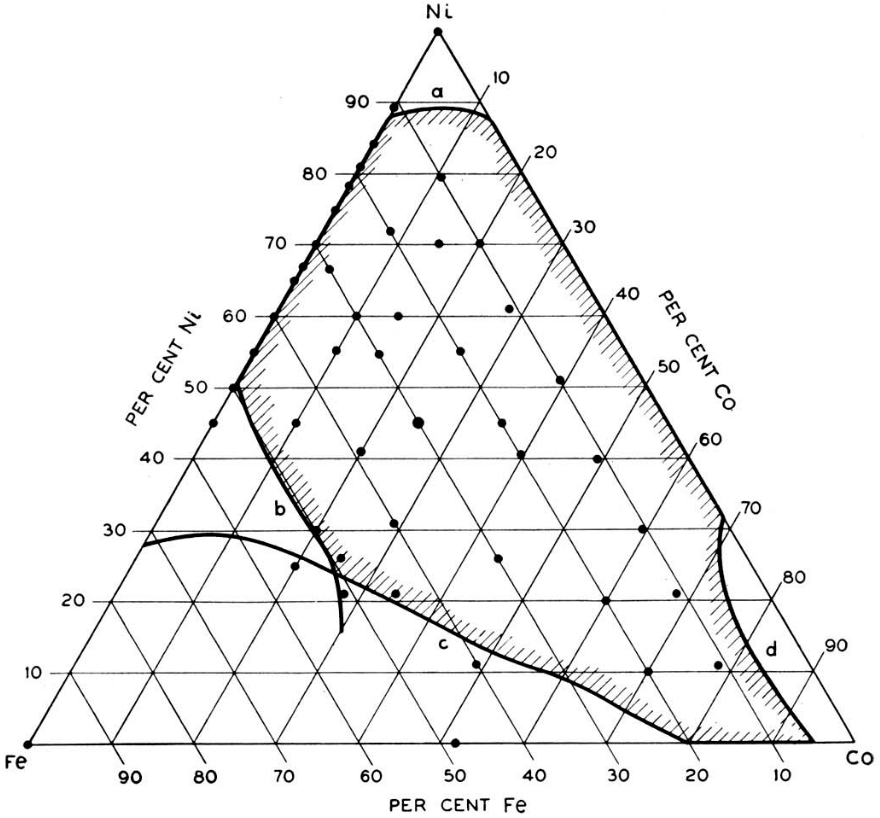

Fig. 2—The alloys investigated. The magnetic properties of the compositions within the shaded area are changed by annealing in a magnetic field. Curves (a) and (b) represent compositions with Curie points of 500° C. Curves (c) and (d) represent compositions with phase transformations near room temperature.

HYSTERESIS LOOPS

The shape of the hysteresis loop is often very radically changed by treatment in a field. The two loops in Fig. 3 show this for specimens of 65 per cent Ni, 35 per cent Fe (65 permalloy). Loop (a) was taken for a specimen annealed without the application of a field and loop (b) for one annealed in a field. The square corners and vertical sides that are present in exaggerated form in the latter condition are characteristic of the loops of materials affected by this treatment. The vertical sides represent changes in magnetic flux which are not instantaneous.

The time necessary for these flux changes depends upon the dimensions of the sample and upon the excess of the applied field over the coercivity. In Fig. 3(b) the time required for the flux to change along each of the vertical sides was about one minute when the applied field was held constant at H_c.

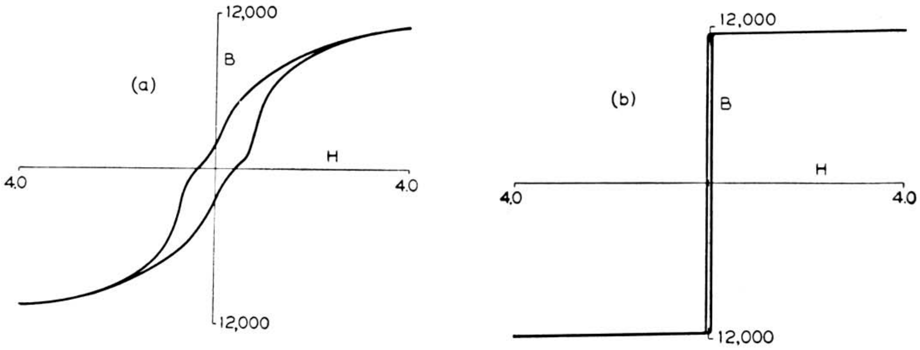

Fig. 3—Hysteresis loops of 65 permalloy cooled in different ways from 1000° C. For loop (a) no field was applied during cooling; for loop (b) a field of 10 œrsteds was maintained during cooling.

Other alloys annealed in a field give hysteresis loops that are very different from those usually found. Some loops of this type are shown in Fig. 4. Loop (a) is for a specimen containing 25 per cent Fe, 15 per cent Co and 60 per cent Ni. An alloy containing 20 per cent Fe, 70 per cent Co, 10 per cent Ni was used for loop (b). Loop (c) is for a 0.051 cm. wire of 65 permalloy, and loop (d) is for a specimen containing 15 per cent Fe, 15 per cent Co and 70 per cent Ni.

It is impossible to demagnetize completely many of the samples that have loops with vertical sides. The sample of Fig. 3(b) could be demagnetized only partially and others could not be demagnetized much below their remanence even by reducing the reversed field in steps as small as 0.003 œrsted, or by an alternating field continuously decreasing in amplitude.

The maximum value of intensity of magnetization obtainable for alloys annealed in a field is no larger than for similar alloys annealed without the application of a field. Approximate saturation is obtained, however, for the former with very much lower applied fields than for the latter.

STRESS SENSITIVITY

It is well known that the application of stress to many of these alloys will produce hysteresis loops that are nearly rectangular.[8] In

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[8] F. Preisach, *Ann. d. Physik*, **3**, 737 (1929). See also references 9 and 10, as well as the earlier paper by R. Forrer, *Comptes rendus*, **180**, 1394 (1925).

the case of 65 permalloy in tension a loop very similar in shape to that given in Fig. 3(b) is obtained. From this it was thought that the hysteresis loop of this alloy annealed in a field would not change with the application of tension. To test this, two straight samples of 65 permalloy tape 60 cm. long were annealed. One was annealed in a field of 10 œrsteds and the other was annealed in zero field (the earth's field was compensated). Measurements were made in a magnetic yoke [9] in such a way that tension could be applied during measurement. The maximum permeabilities as a function of the stress applied are given in Fig. 5. Here it is seen that, as predicted, the specimen annealed in a field was affected relatively little by tension.

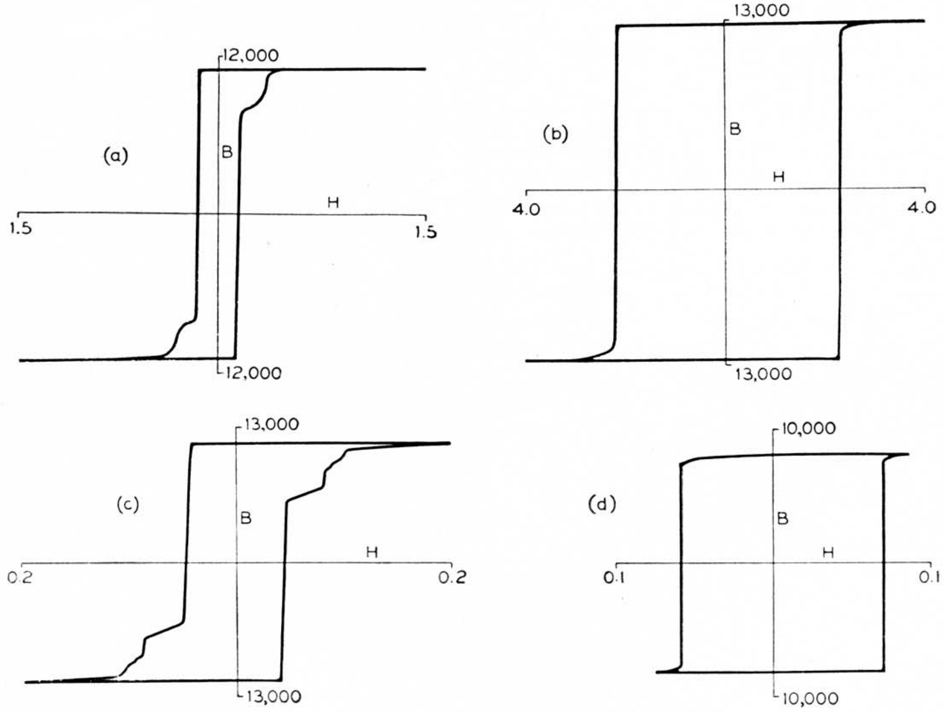

Fig. 4—Hysteresis loops of different materials annealed in a magnetic field: (a) 25 per cent Fe, 15 per cent Co, 60 per cent Ni; (b) 20 per cent Fe, 70 per cent Co, 10 per cent Ni; (c) 65 permalloy fine wire; (d) 15 per cent Fe, 15 per cent Co, 70 per cent Ni.

In straight pieces of thin tape and of fine wire annealed in a field the sudden magnetic changes (corresponding to the vertical sides of loops) were found to travel along the sample at a definite velocity in a manner very similar to that observed by Sixtus and Tonks [10] and others in strained wires.

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[9] O. E. Buckley and L. W. McKeehan, *Phys. Rev.*, **26**, 261 (1925).

[10] K. J. Sixtus and L. Tonks, *Phys. Rev.*, **39**, 357 (1932), and earlier papers.

TEMPERATURE OF ANNEAL

The temperature at which the material is annealed has much to do with the maximum permeability and the shape of the hysteresis loop. This is well illustrated by the following results. Similar ring samples of hard rolled 65 permalloy tape were annealed for one hour at given temperatures and then cooled to room temperature in an applied magnetic field. The values of maximum permeability obtained in this way are plotted in Fig. 6(a) against the maximum temperature of the anneal. Curve (b) gives the maximum permeabilities on similar samples annealed without the application of a field. These results and others indicate that thorough annealing is necessary for maximum effects.

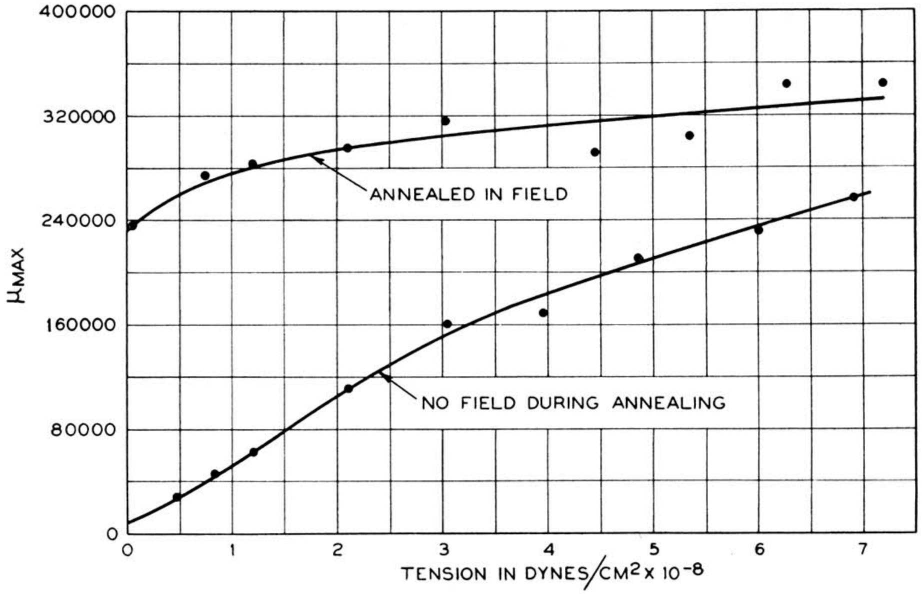

Fig. 5—The effect of tension upon the maximum permeabilities of samples of 65 permalloy annealed with and without an applied magnetic field.

Temperatures higher than those given in Fig. 6 have been used in some experiments. In this case solid rings of 65 permalloy 2.86 cm. O.D., 2.38 cm. I.D. and 0.63 cm. high were treated in hydrogen[11] at 1400° C. for 18 hours and later reheated to 650° C. and cooled in a magnetic field. Hysteresis loops and μ, H curves for one of these samples are given in Figs. 7 and 8. Curves (a) were taken after the high temperature treatment but before the treatment in a magnetic field. Curves (b) were taken after both treatments. The value of maximum permeability for this sample is the highest of which we have

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[11] By P. P. Cioffi, as described in *Phys. Rev.*, **39**, 363 (1932).

any record and the coercivity (0.012 œrsted) is the lowest. The area of the loop indicates an energy loss of 50 ergs/cm.^{3}/cycle which is also

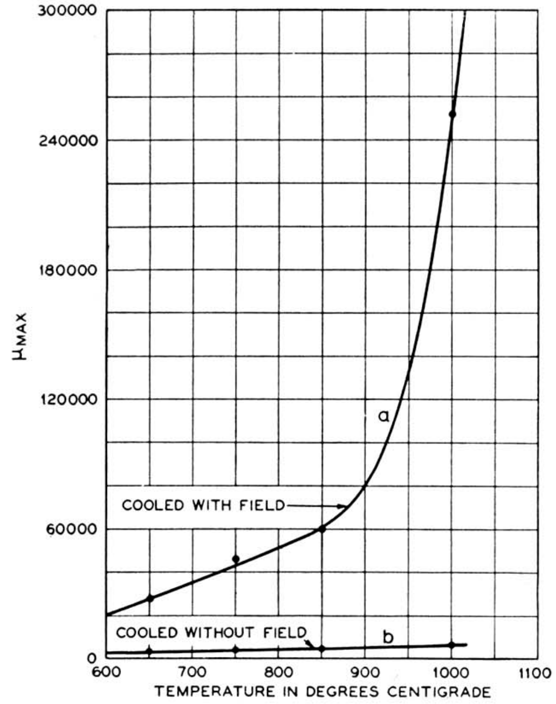

Fig. 6—The maximum permeabilities of samples of 65 permalloy, originally hard worked, after annealing at various temperatures for one hour and cooling with or without an applied field.

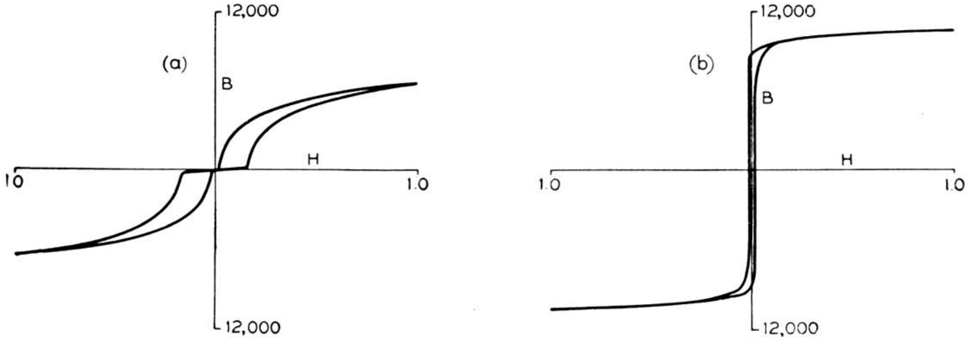

Fig. 7—Hysteresis loops of 65 permalloy: (a) After anneal at 1400° C. for 18 hr. in hydrogen; (b) After further heating to 650° C. and cooling in a magnetic field.

a record low value for hysteresis losses. The time required for the flux to change along each of the vertical sides of this loop was about three minutes.

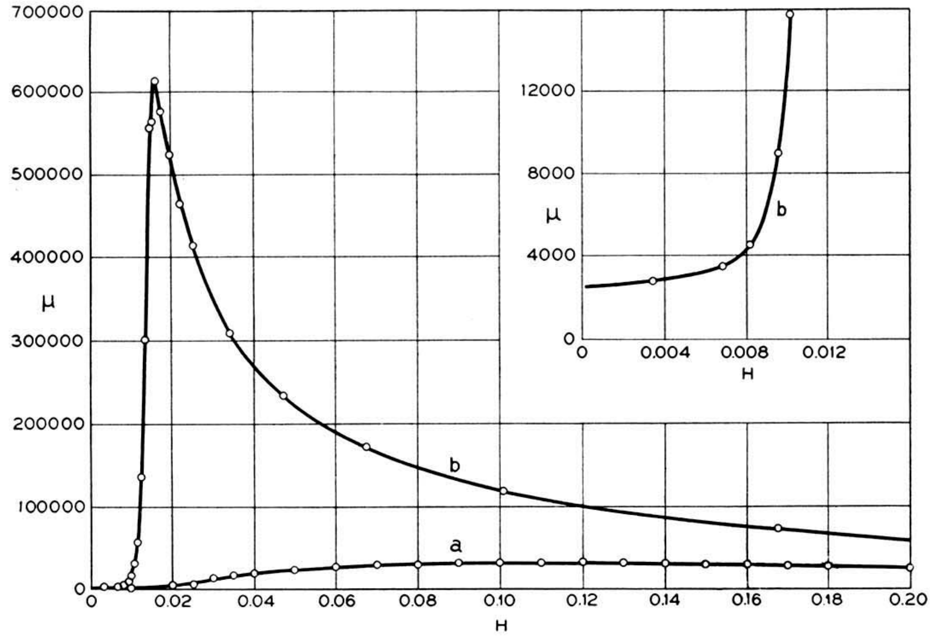

Fig. 8—Permeability curves for 65 permalloy, corresponding to the loops of Fig. 7.

ANISOTROPY

The magnetic properties of these materials are not the same in all directions. In the work described above, the magnetic field that was applied during measurement was parallel to the field applied during heat treatment. The effect of applying these two fields in mutually perpendicular directions was investigated for a seamless tube of perminvar (30 per cent iron, 25 per cent cobalt, 45 per cent nickel) 60 cm. long, 0.48 cm. outside diameter, and 0.025 cm. thick. This tube was annealed at 1000° C. in hydrogen, and again at 650° C. in a magnetic field applied parallel to the axis of the tube. Measurements were made with applied fields perpendicular to the axis by using a search coil wound through the ends of the tube and back along the out-

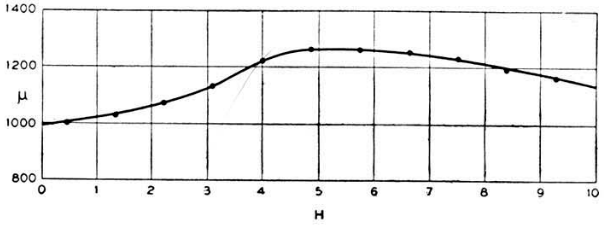

Fig. 9—Permeability curve for a tube of perminvar (30 per cent Fe, 25 per cent CO, 45 per cent Ni) after cooling in a magnetic field applied at right angles to the field used for measurement.

side and a magnetizing current passed through an insulated conductor along the axis of the tube. The values of permeability measured in this way are given in Fig. 9. The maximum permeability under these conditions is about 1250 and the hysteresis loop is nearly a straight line. If, however, the measuring field and the field applied during the anneal are parallel (circular) the maximum permeability on this sample is about 190,000 and the hysteresis loop is nearly a rectangle. The loops for these two situations are shown on the same scale in Fig. 10.

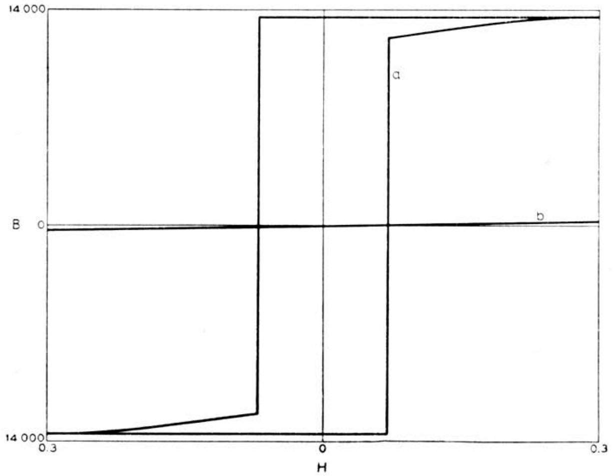

Fig. 10—Hysteresis loops of a tube of perminvar after cooling in a magnetic field: (a) Loop measured with field parallel to the field applied during heat treatment; (b) Loop measured with field perpendicular to the field applied during heat treatment.

SUMMARY

Heat treatment of the binary iron-nickel alloys in a magnetic field is effective in a broad region near 65 per cent nickel. Many of the iron-cobalt-nickel alloys are affected by this treatment. Specimens so treated are relatively insensitive to strains. Record values of maximum permeability, hysteresis loss and coercivity for a ferro-magnetic material were obtained by applying this method to 65 permalloy with the field applied for measurement parallel to the field applied during heat treatment. The magnetic properties so obtained are conspicuously anisotropic.

In general, we have found that if this method of annealing in a magnetic field is to be effective the material must have a Curie point somewhat above 500° C., and must not have a phase change between about 500° C. and room temperature. In any material, thorough preliminary annealing is essential for the best results.

We are inclined to the conclusion that the forces arising from magnetostriction produce plastic deformation if the Curie point is sufficiently high and that this results in magnetic orientation with high permeability in one direction and low permeability in directions at right angles. A complicated type of magnetostriction such as that existing in iron apparently reduces the efficacy of this type of treatment, since relatively small changes have been obtained in this material. The smallness of the changes produced in iron may also be affected by its small magnetostriction and by its greater resistance to plastic flow.

A more detailed discussion of these and other results is given in part II of this paper.

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HEAT TREATMENT OF MAGNETIC MATERIALS IN A MAGNETIC FIELD--II

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EXPERIMENTS ON THE MAGNETIZATION OF TWO ALLOYS AS AFFECTED BY HEAT TREATMENT IN A MAGNETIC FIELD AT VARIOUS TEMPERATURES

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Heat Treatment of Magnetic Materials in a Magnetic Field

II. Experiments with Two Alloys

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The magnetization of two alloys, as affected by heat treatment in a magnetic field at various temperatures, is examined in some detail in order to elucidate the nature of the accompanying changes which result in some cases in a 30-fold increase in maximum permeability. The *experiments* show that these alloys (one containing approximately 35 per cent iron and 65 per cent nickel, the other 20 per cent iron, 60 per cent cobalt and 20 per cent nickel) can be effectively heat treated in a magnetic field of 10 œrsteds if the temperature is above 400° C. and below the Curie point of the alloy. The time during which the magnetic properties change has been measured at different temperatures and is found to vary according to the equation $\tau = Ae^{W/kT}$. The experiments are *interpreted* in terms of the domain theory of ferromagnetism. The changes which occur are due to the relief of magnetostrictive stresses which arise when the material becomes ferromagnetic upon cooling through the Curie point or when an external magnetic field is applied, and the relief comes about by plastic flow or diffusion in the separate domains. The values of A (about 10^{-12} second) and W (2.1 electron volts) are the same as those determined by Bragg and Williams for the above equation which also gives the time necessary for the establishment of a superstructure in alloys. The relation between the two processes, establishment of superstructure and the relief of magnetostrictive strains, is pointed out.

INTRODUCTION

PREVIOUS experiments [1] have established the fact that the magnetic permeabilities of some of the permalloys are considerably increased by heat treatment in a magnetic field of the order of 10 œrsteds. The procedure is first to anneal the material in the usual way at about 1000° C. and cool slowly, and then to apply a magnetic field to the specimen and maintain it while the specimen is reheated to about 500° C. or higher and cooled again to room temperature.

The preceding paper describes an investigation of this phenomenon for a variety of iron-cobalt-nickel alloys, including especially the binary alloys of iron and nickel. The experiments described in the present paper have been done to examine in more detail the specific behavior of two interesting alloys, permalloy having 65 per cent nickel and 35 per cent iron (65 permalloy), and perminvar containing 20 per cent iron, 60 per cent cobalt and 20 per cent nickel. They have to do

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[1] See part I of this paper.

mainly with the changes in magnetic properties that occur in the temperature range about 500° C. The results show that these changes in magnetic properties are to be associated with the relief of magnetostrictive strains by local rearrangement of the material. This will be discussed in more detail below.

EXPERIMENTS WITH 65 PERMALLOY

An experiment showing particularly well the magnitude of the effect and the temperature range in which the application of the field is important, is illustrated in Fig. 1. Four specimens of 65 permalloy from the same casting were heated to 1000° C. and cooled to room temperature, and in each specimen the magnetic field (10 œrsteds) was applied during different temperature regions while cooling, as follows:

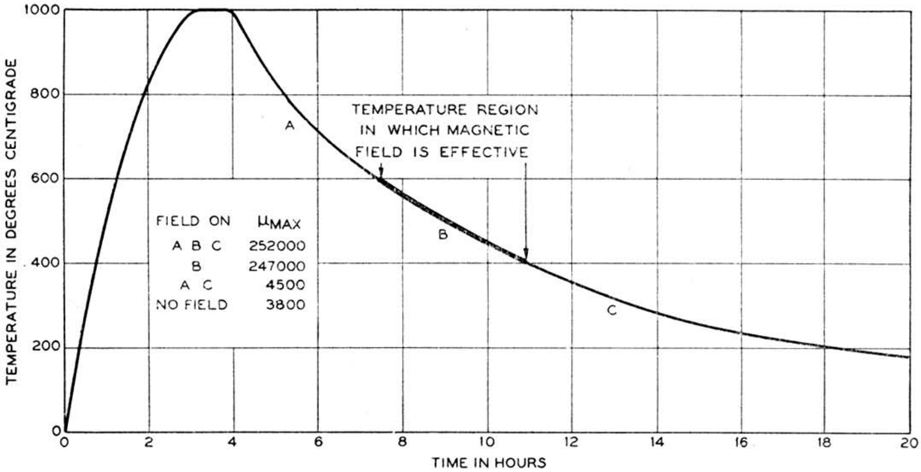

Fig. 1—Permalloy containing 65 per cent Ni, originally hard worked, is heated and cooled as shown. The maximum permeability is increased about 50-fold if a field of 10 œrsteds is applied in the temperature region indicated.

(1) Field applied during entire time of cooling.
(2) No field applied during entire cycle.
(3) Field applied only when cooling from 600° to 400° C.
(4) Field applied during cooling except from 600° to 400° C.

The effect upon the maximum permeability, as measured at room temperature after each cooling, is tabulated on Fig. 1. Application of the field as the specimen cools from 600° C. (just above the Curie point) to 400° C., increases the maximum permeability from about 5000 to 250,000, or by a factor of 50.

To determine more definitely the temperature region in which the field is effective, the following series of runs was made on another single specimen [2] from the same lot of material. The annealed specimen was heated to 600° C. and maintained there for an hour, a field of 10 œrsteds was then applied and the specimen cooled to a temperature t and maintained there for one hour. The field was then removed and at the same time the furnace began to cool to room temperature at the rate of about 150° C./hr. The maximum permeability μ_m was then determined and plotted against the temperature t as shown in Fig. 2,

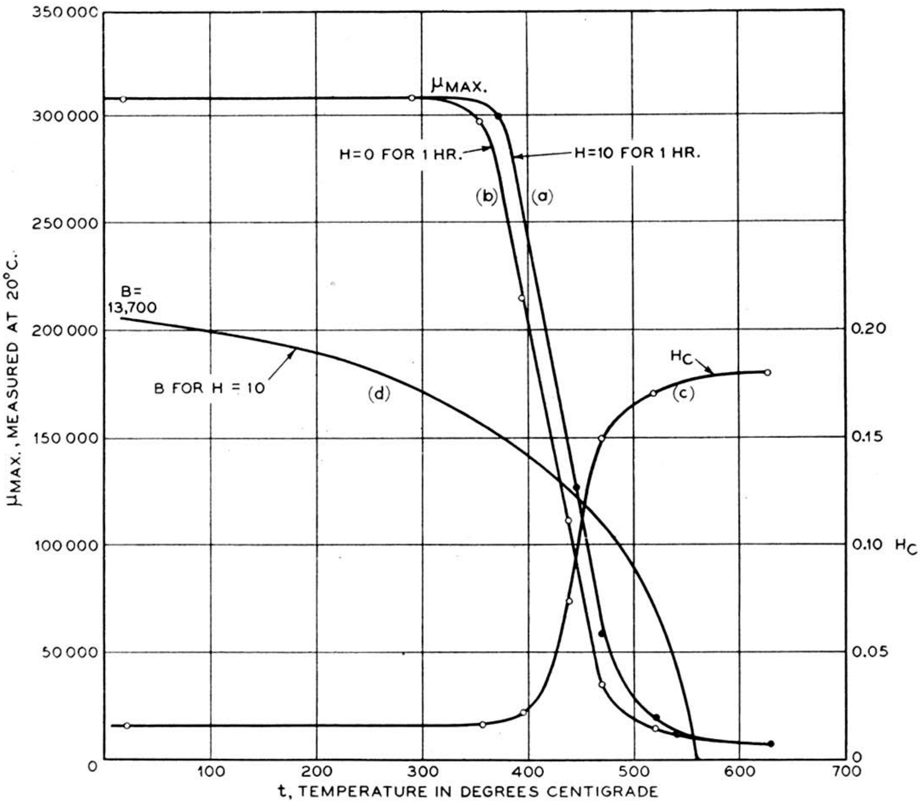

Fig. 2—Values of maximum permeability (curves *a* and *b*) and coercive force (curve *c*) of 65 permalloy measured at room temperature after heat treatment in a field. The field of 10 œrsteds was removed at the temperature, t, on cooling. Curve *d* shows for comparison the value of B for $H = 10$, measured at the temperature t.

curve (a). A similar curve (b) was obtained by applying the same field at 600° C., but removing it as soon as the specimen arrived at the temperature t, no field being present either during the hour at temperature t or during the cooling to room temperature. The corresponding coercivity H_c is shown in curve (c). In the same figure the curve (d) shows the flux density B corresponding to a field strength of 10 œrsteds,

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[2] The dimensions of this ring specimen were: 2.86 cm. O.D., 2.38 cm. I.D. and 0.63 cm. axial height.

measured at the temperature t in a separate series of experiments. In order to obtain these data a second winding of deoxidized copper spaced from the first (H-producing) winding by two lavite rings was connected to the ballistic galvanometer (fluxmeter).

These experiments show that for heat treating this material in a field the effective range is from the Curie point down to a temperature of about 400° C. They do not show whether the effect is associated with the Curie point, lying, for example, in that range of temperature in which the magnetization changes most rapidly with temperature, or whether it occurs at the lowest temperature at which rearrangement (recrystallization or diffusion or plastic flow) takes place rapidly enough to be noticeable during the course of the experiments.

To settle this doubt we selected a second alloy, a perminvar, which was known to be affected by heat treatment in a field and which has a Curie point considerably higher than that of 65 permalloy.

EXPERIMENTS WITH PERMINVAR

The perminvar, selected for its high Curie point of 850° C., has the approximate composition 20 per cent iron, 60 per cent cobalt and 20 per cent nickel. The first experiments, performed with 65 permalloy, were somewhat modified in the perminvar series. The specimen was heated to 950° C., 100° above the Curie point, and maintained there for one hour. The heating current in the furnace was then cut off and the material allowed to cool with the furnace, the cooling rate being approximately 200°/hr. at 900° C. and 150°/hr. at 500° C. The magnetic field was applied at the temperature t, on cooling. The maximum permeabilities measured at room temperature are plotted as curve (a) of Fig. 3. Hysteresis loops were also traversed between ± 10 œrsteds, and the discontinuous change in B, forming the vertical part of the side of such a loop, is plotted as curve (b) in the same figure. Curve (c) shows the value of B for $H = 10$, measured at the temperature t.

In another series of experiments the specimen was heated for one hour as before at 950° C. Then the field of 10 œrsteds was applied and cooling began. At the temperature, t, on cooling, the field was removed and remained zero till room temperature was reached. The maximum permeabilities, plotted against the temperature t, are shown in Fig. 4.[3]

In further experiments, measurements of relaxation time were made between 400° and 600°, the range is which magnetic changes occur as

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[3] The highest values of $\mu_{\max}$ in Figs. 3 and 4 are not the same; that for the latter is higher because it was measured after the specimen had been subjected to many more heating and cooling cycles and was therefore much more thoroughly annealed.

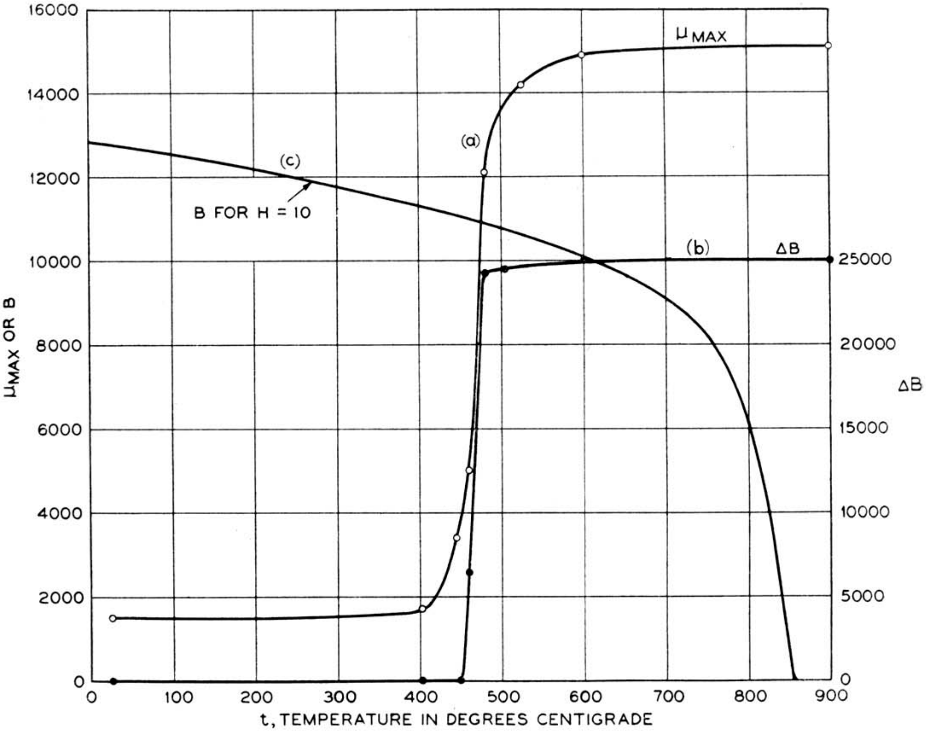

Fig. 3—Curve *a* shows the maximum permeabilities of a specimen containing 20 per cent Fe, 60 per cent Co and 20 per cent Ni, measured at room temperature after cooling from 950° C., a magnetic field having been applied at the temperature, *t*, on cooling. Curve *b* indicates the change in *B* which occurs for constant *H*, the height of the vertical part of the hysteresis loop.

shown in Figs. 1 to 4. The specimen was heated to a temperature *t* and this temperature maintained constant within two or three degrees. At a recorded time a field of 10 œrsteds was applied. At the end of one minute the field was reduced to zero and the galvanometer (fluxmeter) deflection read, after which the field was again applied and the opposite

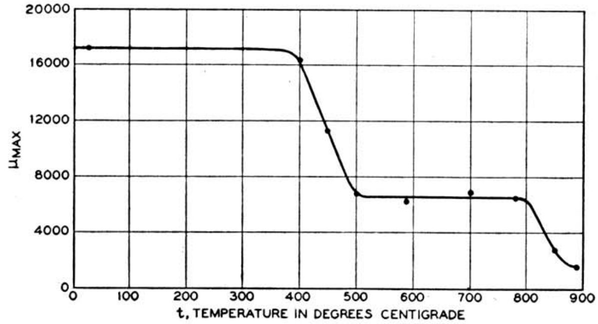

Fig. 4—Data similar to those of Fig. 3, except that here the magnetic field was applied during cooling from 950° C. to temperature *t* and was then removed.

deflection read. The average of the two deflections was recorded, and at the end of each minute thereafter similar observations were made. The total time during which the field was off was about five seconds per minute. The data for 480° C. are shown in Fig. 5, curve "O." The last deflections here shown are due almost entirely to the air-flux, and were found to be nearly equal to the deflection for the same change of H when the specimen was above the Curie point.

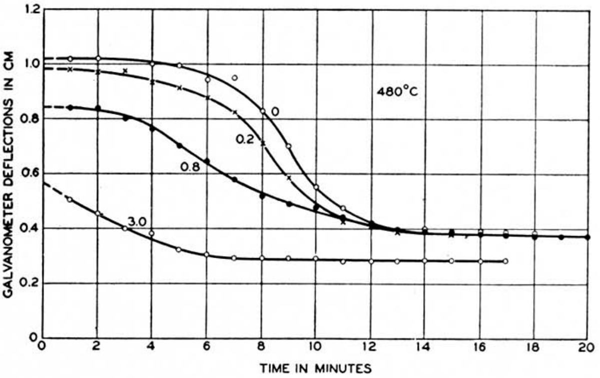

Fig. 5—Measurements made while the specimen (20 per cent Fe, 60 per cent Co, 20 per cent Ni) was maintained at 480° C. A field $H = 10$ was applied at zero time and held constant except for momentary changes, once each minute, to 0, 0.2, 0.8 or 3.0 œrsted. The galvanometer deflections caused by these changes in H indicate changes in B and show how the magnetic properties change with time.

We designate by τ the relaxation time, the time necessary for that part of the deflection which is to disappear, to change to $1/e$ of its original value. This relaxation time was determined for a series of temperatures, in this case always by changing H from $+ 10$ to $+ 0.2$, instead of from 10 to 0. Then log τ was plotted against the temperature t. The results are approximated by the straight line shown in Fig. 6(a). A plot of log τ against the reciprocal of the absolute tem-

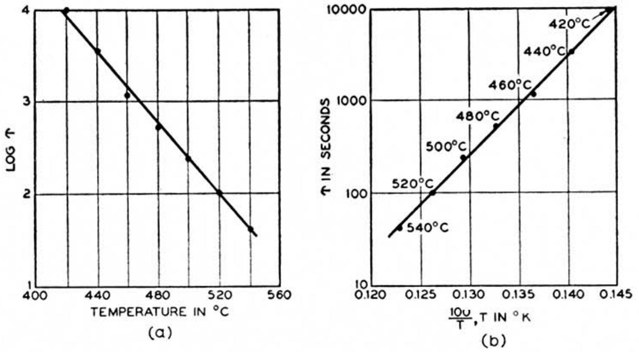

Fig. 6—The relaxation time, τ as dependent upon temperature.

perature T is also substantially a straight line as shown in Fig. 6(b).[4] This ambiguity results from the relatively small range in T.

Instead of changing H from 10 to 0 when taking ballistic deflections, in some experiments at 480° H was changed from 10 to either 3, 0.8 or 0.2 œrsted. The corresponding deflections, shown by the curves of Fig. 5, were converted into flux density, B, and the latter plotted as a function of the time. Values of flux density and time from these curves were used in turn to plot parts of the hysteresis loops as they were at the times 0, 5, 9 and 15 minutes after the first application of the field of 10 œrsteds. Fig. 7 shows these loops and the way in which they change gradually from the usual rounded form to the square form characteristic of material heat treated in a field.

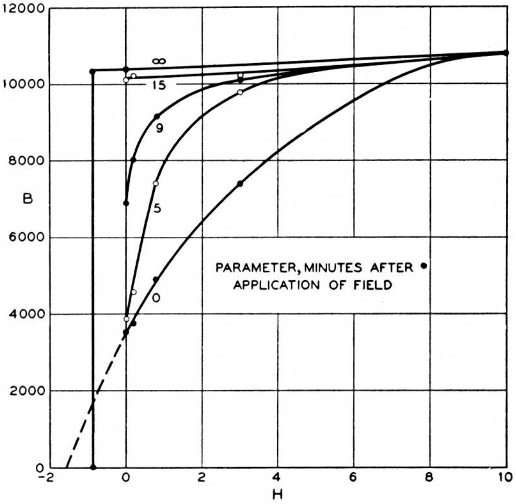

Fig. 7—Portions of the hysteresis loop measured at 480° C., at various times after the application of a field of 10 œrsted.

Discussion of Results

The results on 65 permalloy (Figs. 1 and 2) showed the heat treatment in a field to be effective when the field is applied in that range of temperature which is below the Curie point and above, say, 400° C. The results of Fig. 3 show that in spite of the higher Curie point this statement applies also to perminvar. For both materials the largest effects are produced when the field is present between 400° and 500°

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[4] In a paper on time lag in magnetization at high temperatures, *Physik. Zeits.*, **32**, 860 (1931), H. Kühlewein has described slow changes in magnetization at 400° C. and above, which he attributes to the slow relief of magnetostrictive stresses by plastic flow.

C., temperatures removed from the Curie point of the perminvar by 300° or 400°. It was thus established that the lower limit of 400° is independent of the Curie point, the only requirement being that the latter must be substantially higher than 400° C.

The lowest temperature at which the heat treatment is effective can be identified with that at which plastic flow begins to occur as the result of the forces brought into play by magnetostriction. These forces are of the order of magnitude of 0.1 to 1 kg./mm.^{2} as calculated from the magnetostrictive strain at saturation ($\approx 10^{-5}$) and Young's modulus ($\approx 2 \times 10^4$ kg./mm^2.). There are as yet no data on the plastic flow of the alloys we have studied, but the data for nickel indicate that forces of this magnitude begin to produce flow at 400 to 500° C.[5] This point of view is borne out by Fig. 6, which shows that the relaxation time is an exponential function of the temperature, a relation known to hold for plastic flow.[6]

Another point is to be noticed in the curves of Fig. 7. A comparison of the curve for the initial time and that for five minutes shows that at the latter time the flux density for $H = 0$ has changed very little (here $\tau = 9.5$ minutes) whereas the flux-density for $H = 3$ has changed 80 per cent of the total amount it has to change ($\tau = 3.0$ minutes). It is to be supposed that different parts of the specimen will be under strains of different magnitudes, and that during the annealing the largest strains will disappear more rapidly than the intermediate and smallest strains do. Since only the large strains affect the magnetic properties in high fields (*e.g.*, $H = 3$) the magnetization in high fields will change upon annealing before those in low fields ($H = 0$).

The changes in magnetic properties of iron-nickel alloys due to cold working, and their recovery by annealing at various temperatures, have been studied by Tammann and Rocha.[7] They find that complete recovery takes place only after annealing at 500° to 800° C., depending on the composition, but that the most rapid changes occur at 300° to 600° C. Analogous changes also occur for the electrical and mechanical properties. This range of temperature is roughly the same as that in which we observe the greatest magnetic effects due to annealing in a field, and the phenomena are undoubtedly very similar in nature.

Our data discussed above all support the plastic flow hypothesis provided we can show how plastic flow in the presence of a field in-

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[5] Recent data of P. Chevenard, *Rev. de Mét.*, **31**, 517 (1934), show that a stress of 1 kg./mm.^{2} will produce an extension of 10^{-5}/hr. at about 425° C. His data for iron indicate the corresponding temperature to be about 200° C. higher than for nickel.

[6] Plastic flow due to magnetostrictive stresses has already been mentioned by Kühlewein (reference 3) and by H. Kersten, *Zeits. f. Physik*, **71**, 560 (1931).

[7] G. Tammann and H. J. Rocha, *Ann. d. Physik*, **16**, 861 (1933). In a series of papers, Tammann and his students have reported also the recovery of various electrical and mechanical properties by annealing.

creases the permeability in the direction of that field. In order to understand this, a more detailed consideration must be given to the changes which occur in the local strains, and their relation to the changes in magnetization. These considerations are given in the next section.

An additional remark should be made concerning Fig. 4. When the field is reduced to zero on cooling at any point between 700° and 500° C., the permeability at room temperature has a nearly constant value intermediate between the two extremes. We know of no good reason why this "shelf" should exist. According to the simple theory given below we should expect the permeability to be higher in this range.

DOMAIN THEORY OF HEAT TREATMENT

We start with the idea, as first proposed by Weiss many years ago and recently discussed anew by Webster, Heisenberg, Becker and others, that a magnetic material is composed of small domains, in each of which the material is magnetized to saturation in some direction. In iron and 65 permalloy this direction is one of the cubic axes of the crystals. In an unmagnetized polycrystalline specimen, in which the crystals are oriented at random, some domains are magnetized in every possible direction. As the field is applied in a certain direction, the magnetization in most of the various domains changes by 90° or 180° to become more closely aligned with the field. The sudden changes from one of the six directions of easy magnetization to another is the explanation of the Barkhausen effect.

Now consider what will happen to these regions as the material is heated above the magnetic transformation point. More specifically let us take, as shown in Fig. 8 (a) a region assumed spherical, for convenience, which is above the magnetic transformation point, but which will form a magnetized domain when cooled below this temperature. As shown in (b), it will become magnetized along one of the cubic axes; but as it does so it will tend to change its shape. If time is allowed, it may actually assume by plastic flow of its neighbors the ellipsoidal shape appropriate to magnetostriction, as shown in an exaggerated form in (c); or, it may do the equivalent thing and flow plastically within its own boundaries relieving the magnetostrictive stresses in this way. Then when the material is cooled to room temperature and is magnetized in the direction indicated in (d), the magnetization in this domain will reverse by 180° to one of its other stable positions more closely aligned with the field. No change in shape will occur during this change in magnetization. If, however, still at room temperature, we apply the field in the direction indicated in (e), the domain will *tend* to change its shape as shown by the dashes. But this change will be opposed by the mechanical constraints due to the surrounding material. If in another experiment the material is cooled rapidly enough through the magnetic transformation point down to a low temperature ($< 400°$ C.), there will be insufficient time for any change in shape to occur by plastic deformation and the domain will maintain more or less its original spherical form. In such a condition (f) the magnetization of the domains may change by 90° or 180° with equal facility, whereas in slowly cooled material the domains can change by 180° C. easily but by 90° only with great difficulty. This case can occur, for example, in 78 permalloy the permeability of which is higher after rapid cooling than after slow cooling: the additional probability of 90° rotations increases the number of domains which can easily be aligned with the applied field.

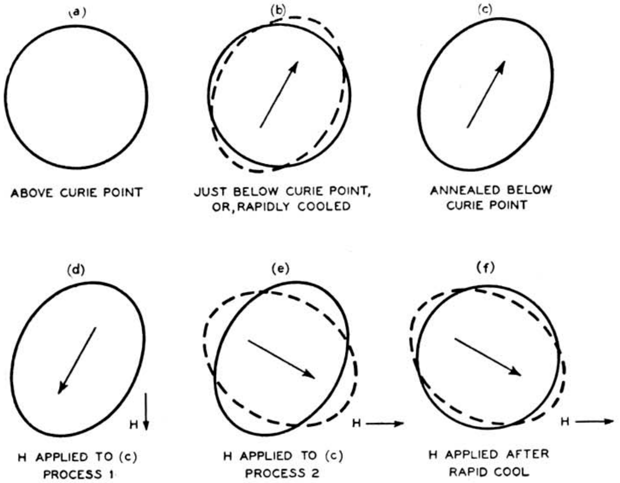

Fig. 8—Idealized domains (solid lines) under different conditions of heat treatment and magnetization. The dashed lined indicate in an exaggerated way the changes in shape which magnetostrictive forces tend to produce, and which do occur after plastic flow (or diffusion) of the material has taken place.

The effects of slow cooling both with and without a field, are illustrated in Fig. 9. Here in the first rectangle the domains are represented by circles and the directions of magnetization of the different domains are at random, as in an ordinary polycrystalline material. Actually in the figure all the directions of magnetization in a plane are shown in steps of 30°. When this material is magnetized as indicated the magnetization vectors will change by 0°, 90° or 180° to align themselves as nearly as possible with the field, consistent with the restriction that they must be magnetized in one of the easy directions of magnetization in the crystal, that is, along a cubic axis. Now when the material is annealed in a magnetic field, the domains will be oriented in the same ways but the strains produced by magnetostriction when it is cooled through the magnetic transformation point, will be relieved if the temperature and time allow (rectangle four). In steps of 30° in the two-dimensional model they will be oriented in just three ways. When the material is demagnetized at room temperature, these three positions will increase to six as shown in the third rectangle. When now it is magnetized again during measurement, each domain will change by 0° or 180°. Comparison of the second and fourth rectangles shows that when no field is present during heat treatment, half of the domains change by 90°, which they must do against the strong mechanical forces which oppose any change in shape, forces which are absent when the material is heat treated in a magnetic field.

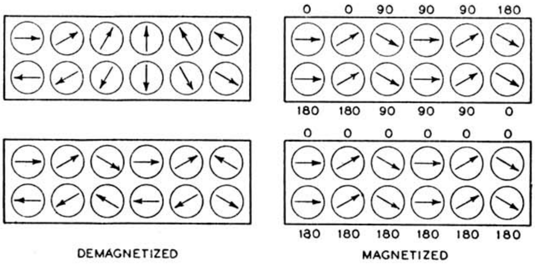

Fig. 9—The changes in the direction of magnetization in domains, by 90° or 180° as indicated, upon application of a field.

These diagrams illustrate an additional point: If we try to magnetize this material at right angles to the direction in which the field was applied during heat treatment, each domain must change by 90° and very large mechanical forces will be operative resisting this change. This simple picture, then, indicates why the permeability should be higher after heat treatment in a field when the field is applied in one direction (parallel), and lower when it is applied in the other (perpendicular).

The Relaxation Time

Within experimental error, the logarithm of the relaxation time is proportional to the reciprocal of the absolute temperature T, as shown in Fig. 6 (b); that is, $\tau = Ae^{W/kT}$, where $A = 2.8 \times 10^{-12}$ second, $W = 3.4 \times 10^{-12}$ erg (2.1 electron volts), and k is Boltzmann's constant. Or we may designate by T_1 the absolute temperature at which $\tau = 1$ second, and write

$$\tau = e^{-\ln A (T_1/T - 1)},$$

where $T_1 = 931°$ K. (658° C.).

Equations of these forms have already been used by Bragg and Williams[8] to designate the relaxation time for certain properties of alloys which form superstructures. W is interpreted by them as the "activation energy" required for the interchange of atomic position which takes place during the formation or disruption of the superstructure, and for $AuCu_3$ has the value 2.0 electron volts. For the same alloy A has a value of about 10^{-12}.

Since the equation connecting τ with T is the same, both as to form and to the value of the constants, for the magnetic experiments described here and for the alloys with superstructure, it is important to consider what the two physical processes have in common. As pointed out by Bragg and Williams, the rate of interdiffusion of two metals is intimately connected with the relaxation times which they determine. But this rate is also intimately connected with plastic flow, especially one due to magnetostriction where there is deformation without change in volume. According to the simple theory described in the preceding section, during heat treatment either the boundaries of a domain are changed by plastic flow or the material inside adjusts itself to the unchanged boundaries. In a domain in which magnetostrictive stresses have just arisen, because of constrained boundaries, similar forces act upon each element of volume of the domain, and it is only necessary for readjustment of atom positions to occur within regions containing a few atoms for the whole domain to become stress-free. This readjustment is accomplished by diffusion, and it is to be expected that these relaxation times will be of the same order of magnitude as those for β-brass and for the gold-copper and iron-aluminum alloys discussed by Bragg and Williams.

We conclude, then, that both in alloys with superstructure and in ferromagnetic materials, we find the same mechanisms by which the changes take place and the same potential barriers to be overcome. The difference between the two phenomena is that in the one case the interatomic forces tend to put different kinds of atoms near together and in the other case the magnetic (magnetostrictive) forces act to change the distances between the atoms.

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[8] W. L. Bragg and E. J. Williams, *Proc. Roy. Soc.*, A**145**, 699 (1934).

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