Magnetic Crystal Anisotropy and Magnetostriction of Iron-Nickel Alloys

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Single crystals of a number of iron-nickel alloys were prepared, and measurements made of the magnetic crystal anisotropy, and of the magnetostriction at saturation in different crystallographic directions, as dependent on the rate of cooling of the specimens after annealing. There is a large effect of the cooling rate on the anisotropy, for compositions near $FeNi_3$, where atomic ordering occurs. There is a definite but smaller effect of cooling rate on the magnetostriction. The composition for highest initial and maximum permeabilities is nearly that for which λ_{111}, the magnetostriction in the direction of easy magnetization, is equal to zero.

INTRODUCTION

In any theory of the magnetic properties of the iron-nickel alloys, one should take into account the magnetic crystal anisotropy and the magnetostriction at saturation in different crystallographic directions. Some data on these quantities have already been reported. Grabbe[1] showed that the anisotropy constants of quenched alloys are larger (more positive) than those of alloys slowly cooled, and Lichtenberger[2] measured the magnetostriction in the [111], [110], and [100] directions for alloys having one heat treatment.

In the present work the magnetic crystal anisotropy and the magnetostriction in various crystallographic directions are determined on the same specimens, which are subjected to different heat treatments. In confirmation of the work by Grabbe, K is found to depend markedly on heat treatment on account of the atomic ordering that occurs in these alloys, especially in those near in composition to $FeNi_3$. Some effect of heat treatment on saturation magnetostriction is also observed.

The results are discussed briefly in their relation to the "Permalloy problem" which is to explain the various peculiar magnetic properties of iron-nickel alloys having the face-centered cubic structure, especially the high permeabilities of commercial alloys containing about 78 percent nickel, and their variation with heat treatment. It is pointed out that when magnetization proceeds by domain boundary displacement, magnetization gives rise to a minimum of strain energy when the magnetostriction is zero in the *direction of easy magnetization*. In pure quenched alloys this occurs at 79 to 80 percent nickel. This is close to the optimum composition of the alloys of ordinary purity.

PREPARATION AND TREATMENT OF CRYSTALS

Specimens of alloys containing 35 to 90 percent nickel were prepared as described by Walker, Williams, and Bozorth,[3] by slow cooling of the melt in an atmosphere of pure dry hydrogen. The raw materials used were Armco iron and selected Mond nickel, in both of which the metallic impurity content is low; the nonmetallic impurities C, N, O, and S are known to be removed by dry hydrogen at the temperatures used.

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 as prepared for use. After careful grinding with a carborundum wheel, and deep etching, the orientation was checked using x-ray spectrometry and back-reflection Laue photographs, the latter for determining the position of the crystal axes in the plane of the disk. When a variation of the surface from the desired crystallographic plane was more than 0.5°, a further correction was made.

Specimens were cooled at two standard rates: (1) they were withdrawn from the furnace at about 700°C and placed quickly on a cold copper plate, or (2) they were cooled from above 600°C to below 300°C at 2°C/hr. In (1) the estimated cooling rate was 300 000°C/hr. The known order-disorder transformation of $FeNi_3$ is 506°C.[4] 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.

MAGNETIC CRYSTAL ANISOTROPY

The magnetic crystal anisotropy constant K of cubic crystals is defined by the expression for the energy density of crystal anisotropy,

$$E=K(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2),$$

the α's being the direction cosines of the magnetization with respect to the crystal axes. In our experiments K was determined by measuring the torque per unit volume L acting upon the crystal when it was placed in a uniform field sufficiently strong to saturate it. If θ is the angle, measured in the (100) plane between the direction of magnetization and the [001] axis,

$$L=-\partial E/\partial\theta=-\tfrac{1}{2}K\sin 4\theta,$$

and K is then twice the maximum amplitude of L. K and L are expressed in ergs/cm³.

Measurement of L was carried out with apparatus

[1] E. M. Grabbe, Phys. Rev. **57**, 728 (1940).
[2] F. Lichtenberger, Ann. Physik **15**, 45 (1932).
[3] Walker, Williams, and Bozorth, Rev. Sci. Instr. **20**, 947 (1949).
[4] O. Källbäch, Arkiv Mat. Astron. Fysik **34B** (17), 1 (1947).

designed by Williams,[5] and similar in principle to that used by Sixtus.[6] The torsion fiber was a phosphor-bronze wire 0.38 mm in diameter and was found to have a constant of 59 dyne-cm/degree of torque. Reproducible settings could be made to less than 0.1° in the torque angle, therefore in a crystal having a volume of 0.1 cm² the value of K could be determined with an accuracy of about 100 ergs/cm³, provided there were no other errors.

The magnetic field strength normally used was 9000 oersteds. This was more than sufficient to saturate the specimens of alloys measured, as shown by the constancy of the torque upon reduction of the field to one-half or less.

Often the final specimen was found to have a section that was not exactly circular; consequently, the maxima and minima of the torque curve did not have equal amplitudes. In such a case, only the 4θ component of the curve was used, as determined by a simple procedure: the observed torque curve was displaced horizontally by 90° and superposed on the original, and the average of the two curves taken. Such a procedure also allows for any small holes or imperfections that may or may not be visible on the surface. A specimen of one alloy (74 percent Ni) was prepared parallel to the (110) plane, the geometrical form of the disk being maintained with great care. The torque curve was found to depart only slightly from the theoretical form calculated with the second anisotropy constant K_2 equal to zero.[7] (The data gave $K_2/K<0.06$.)

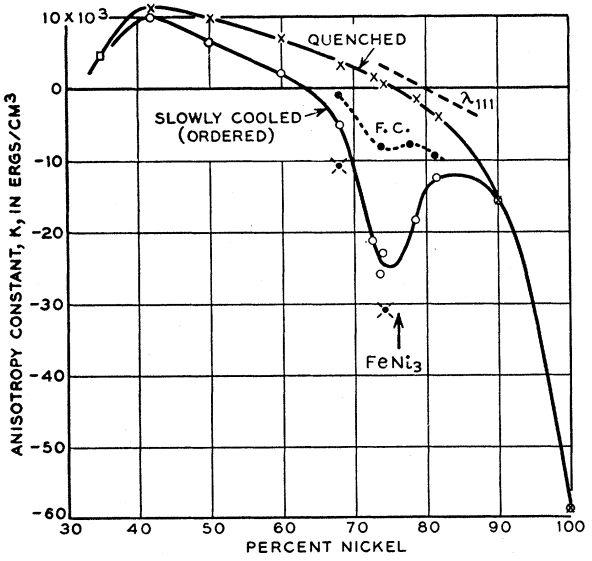

FIG. 1. Magnetic anisotropy constants of quenched and of slowly cooled alloys. Approximate rates of cooling, 10^5 and 2.5°C/hr, respectively, from 600 to 300°C. Broken line F.C. shows values for 55°C/hr. Line λ_{111} shows composition at which magnetostriction in [111] direction goes through zero. Single low points at 68 and 74 percent nickel are for cooling rate of about 1.5°C/hr.

[5] See R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., New York, 1951), p. 556.
[6] L. P. Tarasov, Phys. Rev. **56**, 1224 (1939).
[7] See reference 5, p. 563, for expression containing K_2.

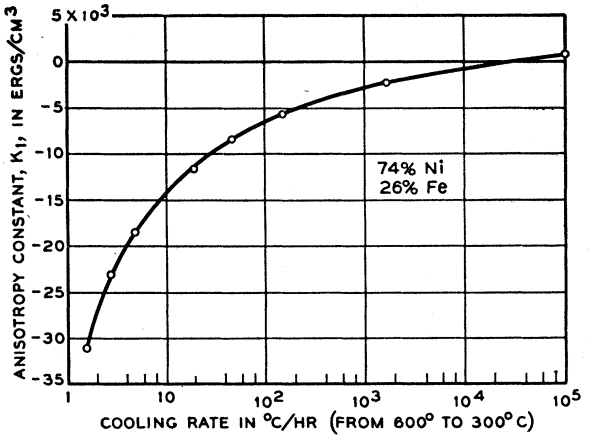

FIG. 2. Anisotropy constants of 74 percent nickel crystal as dependent on rate of cooling from 600 to 300°C. Highest cooling rate is approximate only (quenched on copper plate).

DISCUSSION OF ANISOTROPY

Data for alloys cooled at different rates are shown in Fig. 1. It is noted at once that the slowly cooled alloys have lower (more negative) anisotropy constants, especially in a range of composition near $FeNi_3$, at which a deep minimum occurs. Atomic ordering was detected by x-rays in the slowly cooled alloys containing 74 and 68 percent nickel, and it was not observed either in quenched alloys of the same compositions or in a slowly cooled specimen containing 90 percent nickel. It seems probable then that some ordering occurs in alloys containing as little as 40 percent nickel but not in those containing over 85 percent nickel.

Recently, Corliss, Hastings, and Weiss,[8] using neutron diffraction, have observed ordering in a single crystal of 68 percent nickel. After heat treatment in a magnetic field along a [110] direction, they observed ordering when the neutrons were reflected from the plane perpendicular to this [110] direction, and also when they were reflected from planes parallel to this direction. They thus find no indication of the "directional ordering" postulated by Chikazumi[9] as a possible explanation of the effect of magnetic anneal.

Alloys cooled at the intermediate rate of 50°C/hr, a rate often used in practice in "furnace cooling," have intermediate values of K, as shown by the broken line F.C. in Fig. 1. The alloy containing 74 percent nickel was cooled at various rates, and the resultant anisotropy constants are plotted in Fig. 2. This shows that the degree of ordering is still increasing as the cooling rate falls below 2°C/hr.

The composition at which the anisotropy is zero depends on cooling rate as follows:

Cooling rate (°C/hr)	2	55	10^5,
Composition for $K=0$ (percent Ni)	63	67	75.

[8] Corliss, Hastings, and Weiss, private communication.
[9] S. Chikazumi, Phys. Rev. **85**, 918 (1952).

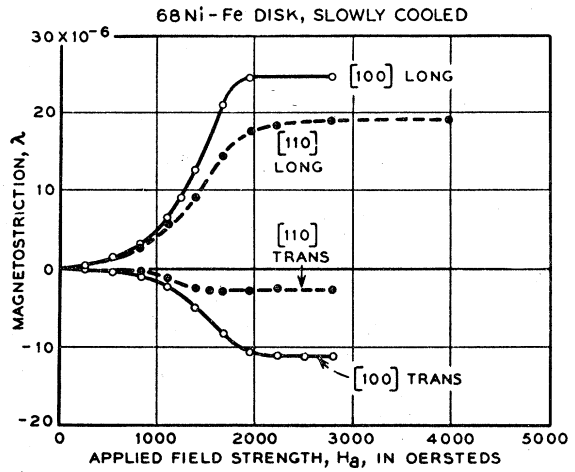

FIG. 3. Longitudinal and transverse magnetostriction of 68 percent nickel crystal, measured in [100] and [110] directions, as dependent on applied field strength. Ratio of diameter to thickness of specimen used is $m=4.4$.

It has previously been pointed out that a low value of the anisotropy constant is related to other magnetic phenomena, for example, low remanence,[10] high strain-sensitivity,[11] and susceptibility to magnetic anneal.[12] More quantitative tests of these relations can now be made.

MAGNETOSTRICTION

Magnetostriction was measured by strain gauges according to the method described by Goldman.[13] Usually $\frac{1}{8}$-in. gauges were employed, fastened to a disk surface with Duco cement and cured at 120 to 160°F as recommended by the maker.[14] The gauge was oriented so that it measured the change in length in a [001] or [011] direction in the (100) plane of the disk.

The magnetization was accomplished in an electromagnet having a 5-in. square section that was tapered to a $3\frac{1}{4}$-in. round section at the pole tips. A valuable increase in the uniformity of the field at the specimen was obtained by turning the pole tips in a lathe so that they presented to the specimen concave surfaces having a radius of curvature of 6 in. With a gap of 2 in., a specimen could be placed on a brass surface at the center, and the non-uniformity of the field would not move it against the friction resulting from its weight. Actually, the specimen was cemented at its edge to a small piece of brass which was held in place by a weight of about 100 g. The cement was allowed to harden with the field on so that the specimen was held parallel to the field; this was done to eliminate bending during the change in field during measurement. The maximum field readily attainable, with 20 amperes through the magnet coils, was about 5000 oersteds. Normally, 3000 oersteds was sufficient for obtaining the desired data.

Leads from the strain gauge led to the three other resistances of the bridge, and one of these, and also the strain gauge, were paralleled with precision resistance boxes that could be varied in steps of 1 ohm to 100 000 ohms. The detector was a Rubicon Photoelectric Galvanometer, with contained dc amplifier, the output of which was connected to a dc voltmeter used with 0–50 or 0–10 volt scales. After balancing the bridge with a given magnetic field applied, we observed the unbalance caused by reducing the field to zero.

Calibration was made by observing the unbalance caused by a known change in the resistance in parallel with the gauge. The change ΔR_g in the gauge resistance R_g that produces the same unbalance as a known change ΔR in the parallel resistance R is determined by

$$\Delta R_g/R_g^2=\Delta R/R^2,$$

provided $\Delta R \ll R$ and $\Delta R_g \ll R_g$. Since $\Delta R_g/R_g$ is strictly proportional to the fractional change in length of the specimen, $\Delta l/l$, we have

$$\frac{\Delta l}{l}=\frac{\Delta R_g}{R_g F}=\frac{R_g \Delta R}{R^2 F},$$

F being the "gauge factor," $(\Delta R_g/R_g)/(\Delta l/l)$, which is supplied by the manufacturer. The calibration curve, linear for small outputs, relates the output to ΔR. In our experiments $R_g \approx 120$, $R \approx 10\,000$ ohms and $F \approx 1.9$. Since the output voltage unbalance $V \approx 1.2\Delta R$, 0.1 volt corresponds to about 0.07×10^{-6}. A fraction of this change in length could be measured when desired.

The effect of the form of the specimen on its magnetostriction was determined as follows. Following Carr

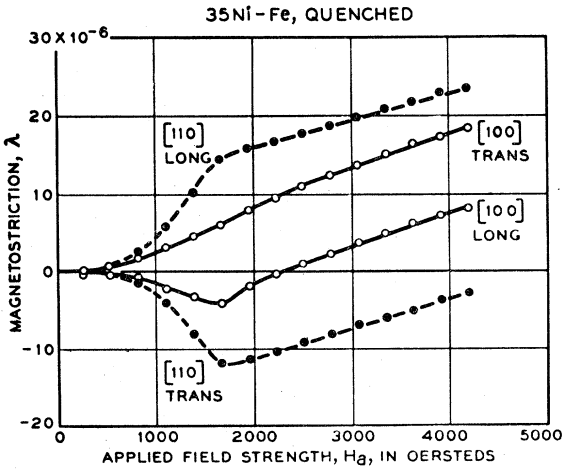

FIG. 4. Magnetostriction of 35 percent nickel crystal, showing large volume effect in high fields. Ratio $m=3.9$.

[10] R. M. Bozorth, Z. Physik **124**, 519 (1948).
[11] R. M. Bozorth and H. J. Williams, Revs. Modern Phys. **17**, 72 (1945).
[12] 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. 214–19.
[13] J. E. Goldman and R. Smoluchowski, Phys. Rev. **75**, 140 (1949).
[14] The Baldwin-Lima-Hamilton Corporation, Philadelphia, Pennsylvania.

and Smoluchowski,[15] we calculate from the axis of an oblate ellipsoid its eccentricity ϵ

$$a=b=c/(1-\epsilon^2)^{\frac{1}{3}},$$

and the quantity

$$\mu=(3\pi/2\epsilon^4)[(1-\epsilon^2)(3+2\epsilon^2)-(3/\epsilon)(1-\epsilon^2)^{\frac{1}{2}}\sin^{-1}\epsilon].$$

The magnetostriction caused by the form is then

$$\lambda_f=\mu I^2/3(c_{11}-c_{12})$$

for the [100] direction, I being the intensity of magnetization and the c's the elastic constants. In the [111] direction $2c_{44}$ replaces $c_{11}-c_{12}$. The form effect λ_f is an expansion and must be subtracted from the observed change in length to give the required magnetostriction of an infinite sheet. In the crystals here measured $a=b\approx 5c$, $\mu\approx 0.8$, $I\approx 1000$, $c_{11}-c_{12}\approx 10^{12}$, and $\lambda_f\approx 0.2\times 10^{-6}$. In most cases the precision of the measurements does not warrant subtraction of this amount.

Measurements of magnetostriction were usually made in the [100] and [110] directions on the same specimens as those used to measure the magnetic anisotropy. After measuring $\Delta l/l$ parallel to the field, the crystal and attached gauge were turned 90° in their plane and the "transverse" magnetostriction recorded. We denote the longitudinal magnetostriction by λ_l, the transverse by λ_t. For the [100] measurements we then have the desired constant

$$\lambda_{100}=2(\lambda_l-\lambda_t)/3,$$

a result that is independent of the domain distribution before magnetization. Similarly, but not quite so obviously, when λ_l and λ_t are the measured changes in length in the [110] direction when the field is applied, respectively, parallel and perpendicular to [110], we have

$$\lambda_{111}=2(\lambda_l-\lambda_t)/3.$$

These expressions follow from the two-constant formula[5] for magnetostriction:

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

λ_{100} and λ_{111} being the saturation magnetostriction in the [100] and [111] directions, for material with domains oriented initially at random, and the α's and β's the direction cosines of the magnetization and the measured change in length, respectively, with respect to the crystal axes. Also, $\lambda_{110}=(3\lambda_{111}+\lambda_{100})/4$.

A typical curve showing $\Delta l/l$ as dependent on the magnetizing field H_a, generated by the electromagnet is shown in Fig. 3 for the specimen containing 68 percent nickel. The demagnetizing field of the specimen must be taken into account if one desires to know H inside of the specimen. However, in order to determine the desired constants λ_{100} and λ_{111}, it is only necessary to know the limit which $\Delta l/l$ approaches in high fields. For the case illustrated, $10^6\lambda_{100}=\frac{2}{3}(24.5+11.3)=23.9$, and $10^6\lambda_{111}=\frac{2}{3}(18.7+2.8)=14.3$.

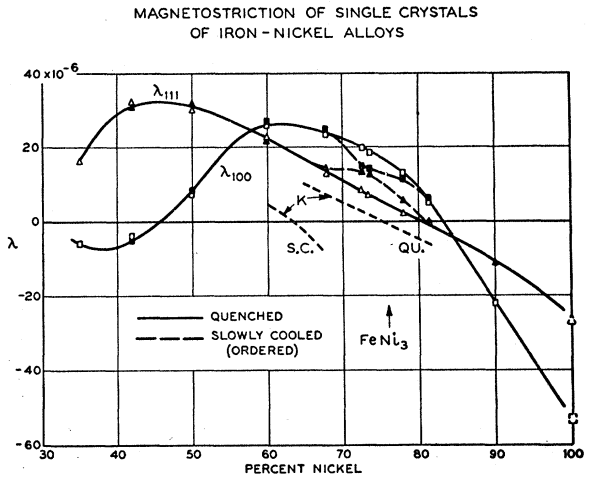

FIG. 5. Magnetostriction of quenched and of slowly cooled alloys. Broken lines show compositions at which anisotropy of quenched (QU.) and slowly cooled alloys (S.C.) goes through zero.

Figure 4 shows the magnetostriction *vs* field strength curves for the single crystal containing 35 percent nickel. All of the curves continue to rise in high fields, a trend that is obviously the result of volume magnetostriction, an expansion that is known to be equal in all directions independent of the direction of the field but approximately proportional to the increase in field-strength. Since this is also known to be most marked near the Curie point, it is not surprising that the material with the lowest Curie point should show the greatest effect.

The volume expansion calculated from the average slopes of the curves in high fields is 12×10^{-9}/oersted, a value that compares reasonably well with the value 15×10^{-9}/oersted reported by Masiyama.[16]

The presence of a large volume magnetostriction does not interfere with the determination of the constants λ_{100} and λ_{111}, for the difference between the longitudinal and transverse magnetostrictions is the desired quantity, and this approaches a definite limit in the high fields used.

DISCUSSION OF MAGNETOSTRICTION

Values of λ_{100} and λ_{111} as dependent on composition are shown in Fig. 5. Results for the quenched alloys are similar to those of Lichtenberger,[2] but λ_{111} is substantially higher in the 40 to 50 percent nickel range.

The effect of ordering is definite but not large, as shown by the broken lines in Fig. 5. The greatest observed change caused by ordering is about 5×10^{-6} in λ_{100} and λ_{111}, but the ratio of the λ_{111}'s can be more

[15] W. J. Carr and R. Smoluchowski, Phys. Rev. **83**, 1236 (1951).

[16] Y. Masiyama, Sci. Repts. Tôhoku Imp. Univ. **20**, 574 (1937).

than 2/1. No definitely established change, attributable to ordering, was observed outside of the range 68 to 80 percent nickel.

Data for alloys in the interesting region near 79 percent nickel are shown on an enlarged scale in Fig. 6. For quenched alloys, λ_{111} goes through zero at about 80 percent nickel; however, the reliability of the two-constant formula for magnetostriction is not great enough to establish the composition for zero λ_{111} with any great precision, and separate measurements have been made[17] to establish this composition. These show that the composition for zero λ_{111} is slightly less than 80 percent nickel.

The change in magnetization by the moving-boundary process takes place most easily when the magnetostriction in the direction of easy magnetization approaches zero.[18] Since [111] is the direction of easy magnetization in quenched iron-nickel alloys containing more than 75 percent nickel, it is to be expected that the composition for highest permeability will coincide with that for $\lambda_{111}=0$. Experiment now shows that these compositions do not differ by more than 1 or 2 percent.

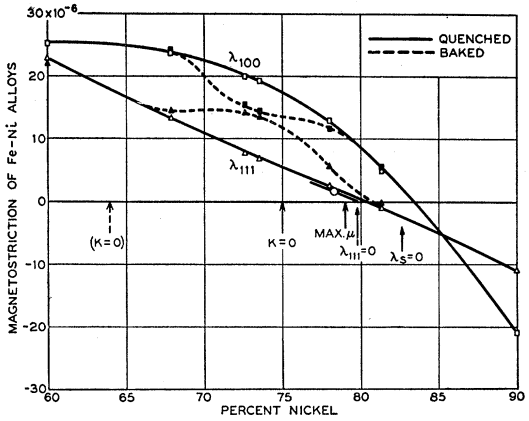

FIG. 6. Magnetostriction of alloys in range 60 to 90 percent nickel, showing unique points in series. Single point near λ_{111} curves is for specimen containing 0.5 percent manganese.

This small shift is probably effected by the nearby point for zero anisotropy at 75 percent nickel.

The relation of the magnetic anisotropy and the magnetostriction to the "Permalloy problem" has been discussed at length in another place.[18] Many of the changes in magnetic properties with composition and heat treatment of alloys of ordinary purity can now be explained in terms of the data here reported.

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

[18] R. M. Bozorth, Revs. Modern Phys. (to be published).