

Abstract

Measurements were made of the magnetostriction of single crystals of Fe-Si alloys varying in composition from 2.5 to 5 per cent silicon by weight. Calculations were made to show how the magnetostriction constants can be obtained accurately from the measurements.

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Abstract

Measurements were made of the magnetostriction of single crystals of Fe-Si alloys varying in composition from 2.5 to 8 per cent silicon by weight. Calculations were made to show how the magnetostriction constants can be obtained accurately from the measurements.

The "form effect" of the specimens was calculated and subtracted from the measured values.

For each of the alloys only two constants were found necessary to describe the strain. For annealed material the constant h_1 (in Becker's notation) was found to be zero at about 6 wt. per cent silicon and h_2 to be zero at about 5 wt. per cent. Measurements on quenched crystals show that at about 9 or 10 atomic per cent silicon a difference in the constant h_1 from that for the annealed crystals begins to appear. Presumably the order-disorder transformation begins at about this percentage.

In part II a survey is made of previous magnetostriction theories. The only one susceptible to calculation is the magnetic dipole theory. For silicon-iron this theory leads to values which are too small, but not always negligible.

In the absence of exact knowledge as to the principal cause of magnetostriction a semi-empirical theory is proposed.

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THE MAGNETOSTRICTION OF SINGLE CRYSTALS OF SILICON-IRON

Part I

Introduction

It has been known for some time that the magnetostriction of polycrystalline Si-Fe alloys is much different from that observed in pure iron and becomes zero for the composition by weight of about seven per cent silicon.¹ For compositions above seven per cent the magnetostriction is reported to have a different sign.

The purpose of the present investigation was to determine the fundamental magnetostriction constants of these alloys by measurements on single crystals. This information is not readily obtainable from polycrystalline measurements due mainly to lack of knowledge of orientation of the crystallites. Single crystals were prepared in the form of oblate spheroids of silicon composition varying from 2.5 to 8 per cent by weight.

Preparation of Spheroids and Method of Measurement

From single crystals² of various silicon compositions, discs were made which were in or near the (100) and the (110) planes and these either were machined or ground into oblate spheroids with a major axis of one inch and a minor of 0.1. Due to their brittleness, it was necessary to use the grinding procedure on the alloys of high silicon. Approximately one mil was etched off the surface of the specimens to prevent formation of new crystallites and they were annealed for five hours at 850°C, then furnace cooled. The results of a different heat treatment will be discussed later. Table I gives a description of the ellipsoids.

- - - - -

1. A. Schultze, Z. Phys., 50, 448-505 (1928)

2. Grown by Mr. R. K. McGeary at the Westinghouse Research Lab., East Pittsburgh, Pa.

Table I

<u>Wt. % Si</u>	<u>Plane</u>	<u>Deviation from Plane Indicated (Degrees)</u>
2.52	(100)	6
2.52	(110)	2
3.03	(100)	4-1/2
3.03	(110)	2
3.59	(100)	--
4.32	(100)	12
4.83	(110)	7
5.80	(100)	1
5.80	(110)	2
7.79	(110)	1

For a measurement the ellipsoid was mounted on a small turntable, as shown in Figure 1, by placing it between the two upright members shown and tightening a machine screw connecting them. Only enough pressure was applied as was necessary to hold the sample in place.¹ The turntable rotated on a brass plate which was calibrated in degrees and by this means the spheroid could be rotated to any angle with respect to the applied field. A brass cup was made to fit over the spheroid in order to reduce temperature fluctuations. The whole assembly, made entirely of brass, was placed between the poles of an electromagnet (Fig. 2).

A field of 3300 oersteds was used throughout the experiments.²

-
1. The initial strain produced by this was of the order of 10^{-6} .
 2. In appendix A it is shown that this is large enough to make the magnetization practically parallel with the applied field.

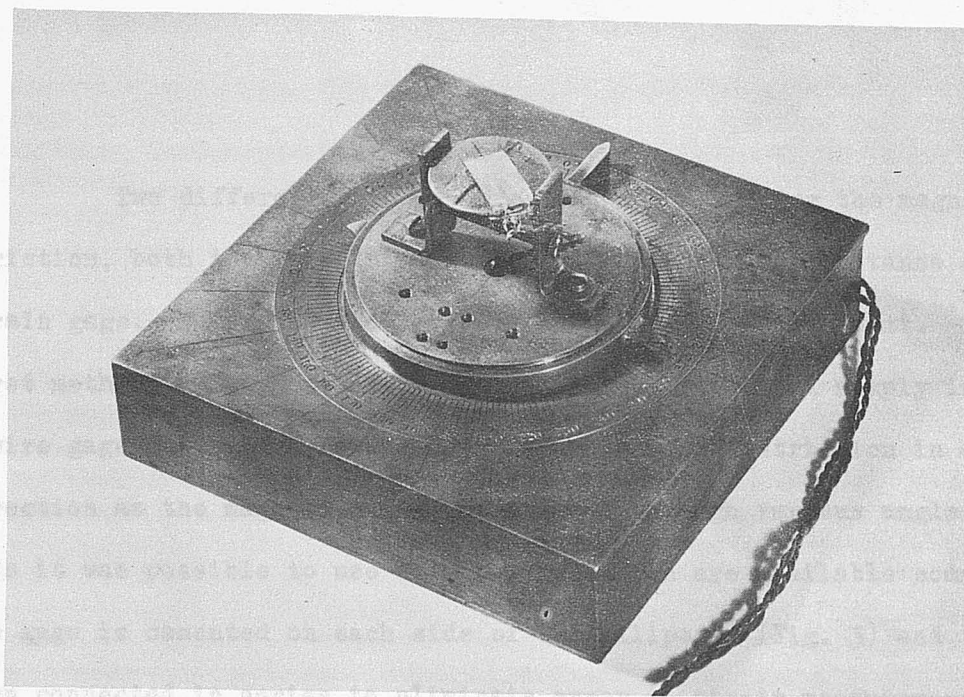


Figure 1

Spheroid Mounted on Turn Table

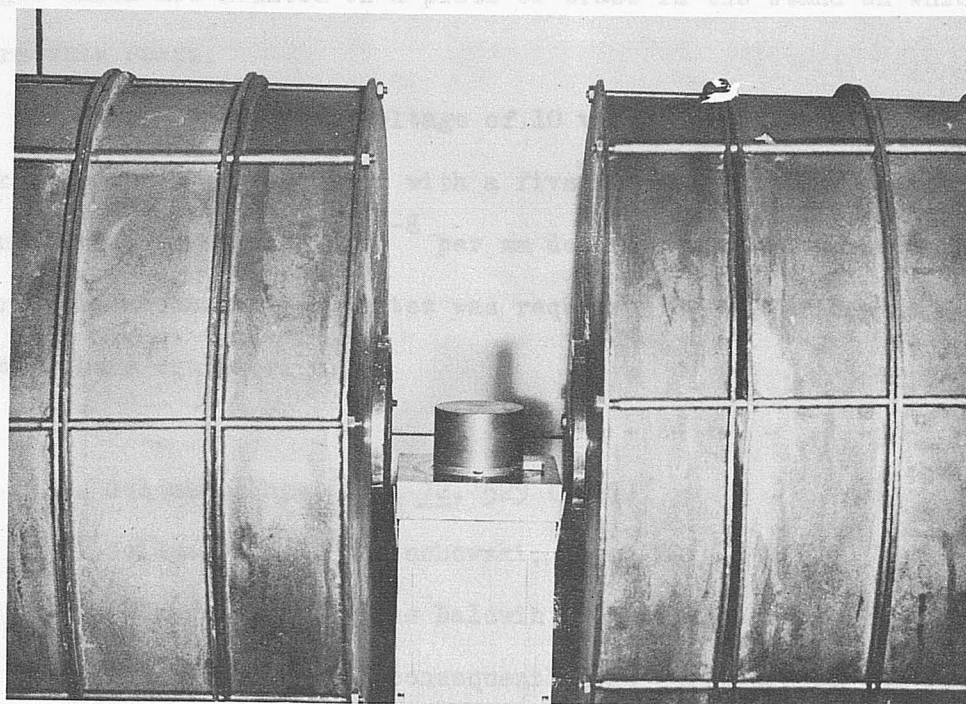


Figure 2

Assembly Between Poles of Electromagnet

Two different methods were used for measuring the magnetostriction, both involving a measure of the change in resistance of a wire strain gage. This offers a very simple method of measurement.^{1,2} The first method, which proved the most accurate, consisted simply in cementing a wire gage on the sample and measuring the magnetostriction in a given direction as the magnetization was rotated through various angles. For this it was possible to use strain gages which are available commercially.³ One gage is cemented on each side of the ellipsoid (Fig. 3) and the two then connected in series to eliminate error in strain measurement due to any bending of the sample. These two gages form the active arm of a Wheatstone bridge, the other three arms being composed of identical dummy gages which are mounted on a piece of brass in the stand on which the turntable rests.

With a battery voltage of 10 volts, and the output of the bridge connected to a galvanometer with a five meter light beam a strain sensitivity of about 5×10^{-8} per mm deflection was obtained. A warm up period of about thirty minutes was required for the bridge to attain temperature equilibrium.

- - - - -
1. J. E. Goldman, Phys. Rev. 72, 529 (1947)
 2. J. E. Goldman and R. Smoluchowski, Phys. Rev. 75, 140 (1949)
 3. A8, SR-4 gages made by the Baldwin Locomotive Works. These gages are only 1/8" in length and consequently the curvature of the spheroid is unimportant in the strain measurement.

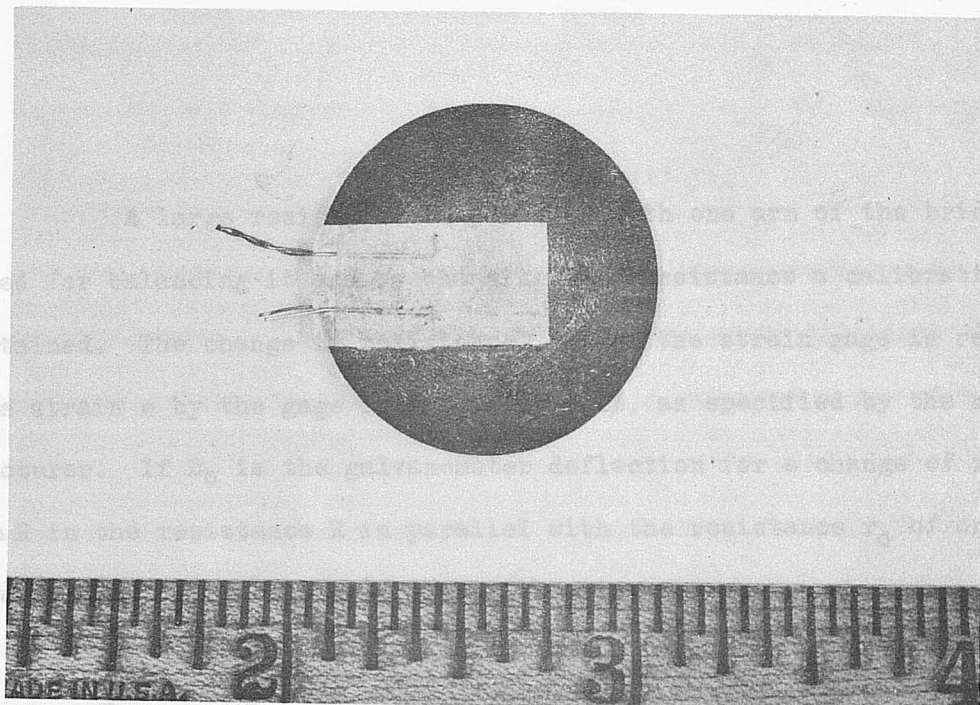


Figure 3

Strain Gauge Cemented on Spheroid

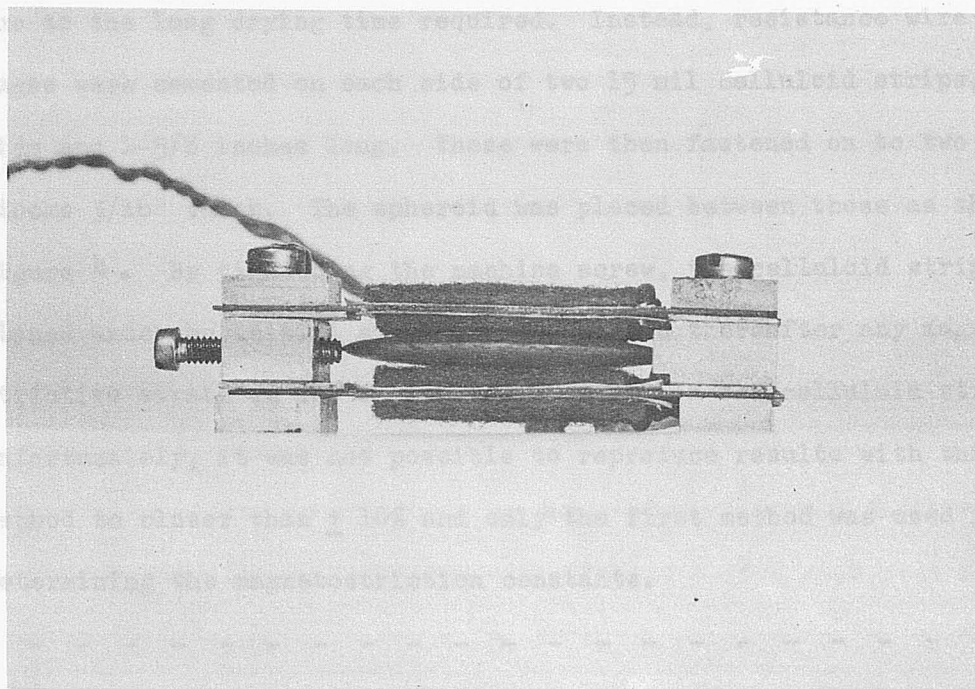


Figure 4

Spheroid Placed in Apparatus Described under "Method Two"

A large resistance in parallel with one arm of the bridge was used for balancing it and by changing this resistance a calibration was obtained. The change in resistance $\frac{\Delta R}{R}$ of the strain gage is related to the strain ϵ by the gage factor, $1.83 \pm 2\%$, as specified by the manufacturer. If D_0 is the galvanometer deflection for a change of resistance ΔR in the resistance R in parallel with the resistance r_0 of one arm of the bridge ($r_0 = 240$ ohms), the calibration is:

$$\frac{\epsilon}{D} = \frac{\Delta R r_0}{(G.F.) D_0 R (r_0 + R) \left(1 + \frac{\Delta R}{r_0 + R}\right)}$$

For the purpose of making strain measurements in many directions on the single crystal spheroids a method was devised making it unnecessary to cement gages directly on the spheroid, since this was time consuming due to the long drying time required. Instead, resistance wire strain gages were cemented on each side of two 15 mil celluloid strips, 1/2 inch wide and 1-5/8 inches long. These were then fastened on to two brass blocks 3/16" thick. The spheroid was placed between these as shown in Figure 4. By tightening the machine screw, the celluloid strips were placed under an initial strain of 10^{-4} , and thereafter any magnetostrictive strain in the sample was observed in the celluloid strips.

Unfortunately, it was not possible to reproduce results with this latter method to closer than $\pm 10\%$ and only the first method was used for determining the magnetostriction constants.

-
1. The strain in the crystal produced by this procedure was about one per cent of that in the celluloid strips.

The Magnetostriction Constants

A detailed discussion of magnetostriction processes is given by Becker and Döring.^{1,2} They consider magnetostrictive strains to be of three types: (1) a magnetostriction which is observed above technical saturation in strong fields and which for most materials is quite small, and will be considered negligible for the field used in these experiments. (2) A magnetostrictive "form effect" which depends upon the shape of the sample or more precisely upon the energy associated with the demagnetizing field. The ellipsoids used in these experiments have small demagnetizing factors and only a small correction is necessary to make the results correspond to those for zero demagnetizing factor, i.e. no form effect. (3) The third and most important is the strain produced in the crystal lattice by the spontaneous magnetization. This is the process to be studied in these experiments and the fundamental information desired is a knowledge of how the crystal lattice is distorted as the magnetization is rotated from an easy direction of magnetization to any other crystallographic direction.

In the past most of the measurements on single crystals have had limited value because the reference state from which the change in dimensions was measured was an ambiguous demagnetized state. Since holo-demagnetization can be achieved for any number of domain distributions it is obvious that the demagnetized state is not a good reference for

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1. R. Becker and W. Döring, *Ferromagnetismus*, (Julius Springer 1939) pp. 270-311.

2. Also see Francis Bitter, *Introduction to Ferromagnetism*, (McGraw-Hill 1937) Chap. VII.

measuring the magnetostrictive strains. Becker and Döring¹ attempt to overcome this uncertainty in demagnetized state by subtracting the magnetostriction measured in a given direction, with the crystal magnetized to saturation in the transverse direction, from the magnetostriction measured in that same direction with the magnetization parallel.

It seems, however, much better to avoid the demagnetized state entirely and use for the reference dimensions those of the crystal when saturated in an easy or $[100]$ direction. One then has essentially a single domain and the crystal lattice distortion which occurs as the magnetization is rotated out of the easy direction can be measured directly.

Due to symmetry of the crystal it is necessary to make only a small number of measurements to obtain the magnetostriction in any direction, having direction cosines $\beta_1, \beta_2, \beta_3$, for the magnetization in any direction $\alpha_1, \alpha_2, \alpha_3$. The strain in the $\beta_1, \beta_2, \beta_3$ direction in a saturated crystal in terms of the strains A_{ii} along the coordinate axes and the shearing strains A_{ik} about these axes is (using the convention of summing over subscripts that appear twice)

$$\epsilon = A_{ij} \beta_i \beta_j \quad (i \text{ and } j \text{ summed over } 1, 2, 3) \quad (1)$$

where the components of the symmetric strain tensor A are functions of the direction of magnetization, $\alpha_1, \alpha_2, \alpha_3$. If the unstrained cubic axes of the crystal are selected as the coordinate axes, and one of the A_{ii} and A_{ik} are known as a function of α_1, α_2 , and α_3 the other A 's can immediately be written from symmetry.

The strain components A_{ii} and A_{ik} can be expressed as a power series of terms in α_1, α_2 , and α_3 . However, because of cubic symmetry

1. Ferromagnetismus, (Julius Springer, 1939) p. 276.

many of the coefficients must be zero. From these considerations of symmetry Becker and Döring¹ give

$$A_{ii} = f_i(\alpha_i^2, S) \quad (1)$$

$$A_{ik} = \alpha_i \alpha_k f_2(\alpha_l^2, S) \quad (i \neq k \neq l)$$

where

$$S = \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_1^2 \alpha_3^2$$

Upon expanding these

$$A_{ii} = a_0 + a_1 \alpha_i^2 + a_2 S + \dots$$

$$A_{ik} = \alpha_i \alpha_k [b_0 + b_1 \alpha_l^2 + b_2 S + \dots]$$

(2)

These expressions can be expanded to as many terms as desired and by making measurements from which the a's and b's can be obtained, the magnetostriction is completely determined.

In making measurements it is desirable to use samples in the shape of thin oblate ellipsoids of revolution, for with these samples the magnetization is uniform, and within the plane of the major axes the demagnetizing factor is small, which likewise makes the magnetostrictive form effect small. If one chooses a crystallographic plane in which none of the α 's is identically zero, the a's and b's, with precise measurements, in principle, can be determined from this single plane. Consider the plane (110). If θ is the angle from the [001] direction,

$$\alpha_1 = -\frac{\sin \theta}{\sqrt{2}}, \alpha_2 = \frac{\sin \theta}{\sqrt{2}} \text{ and } \alpha_3 = \cos \theta.$$

In the [001] direction, ($B_1 = B_2 = 0, B_3 = 1$), the magnetostriction is

1. Loc. Cit. p 274.

$$e_{[001]} = A_{33} = a_0 + a_1 \cos^2 \theta + a_2 (\cos^2 \theta \sin^2 \theta + \frac{\sin^4 \theta}{4}) + \dots \quad (3)$$

The a_1, a_2, \dots obviously can be evaluated by rotating the magnetization into various angles θ .

If the magnetostriction in the direction $[\bar{1}11]$

$$(\beta_1 = -\frac{1}{\sqrt{3}}, \beta_2 = \beta_3 = \frac{1}{\sqrt{3}}) \quad \text{is measured}$$

$$\begin{aligned} e_{[\bar{1}11]} &= \frac{A_{11} + A_{22} + A_{33}}{3} + \frac{2}{3}(A_{23} - A_{12} - A_{13}) \\ &= a_0 + \frac{a_1}{3} + a_2 (\cos^2 \theta \sin^2 \theta + \frac{\sin^4 \theta}{4}) + \dots \\ &\quad + \frac{2}{3} b_0 \left(\frac{\sin^2 \theta}{2} + \sqrt{2} \sin \theta \cos \theta \right) + \frac{2}{3} b_1 \left(\frac{\sin^2 \theta \cos^2 \theta}{2} + \frac{\sin^3 \theta \cos \theta}{\sqrt{2}} \right) \\ &\quad + \frac{2}{3} b_2 \left(\frac{\sin^2 \theta}{2} + \sqrt{2} \sin \theta \cos \theta \right) \left(\cos^2 \theta \sin^2 \theta + \frac{\sin^4 \theta}{4} \right) + \dots \quad (4) \end{aligned}$$

Since the a 's are known the b 's can be determined in a like manner.

In none of the measurements in the (110) does evidence appear that constants beyond a_1 and b_0 are important for describing the magnetostriction. For example, equation (3) for strain in the $[001]$ is an even function in $\sin \theta$ and $\cos \theta$. It is symmetrical about 90 degrees. An examination of the curves in Appendix B for strain in the $[001]$ shows the deviation of measured curves from a squared sinusoid does not have this symmetry and thus cannot be due to additional terms in equation (3). It is shown in the next section that the discrepancies can be explained, or at

least the largest part of them, by the fact that the major planes of the ellipsoids are slightly out of the crystallographic plane indicated, and the direction of strain measurement may be slightly different from that indicated. Angles of the order of one or two degrees can account for the discrepancy.

For strain in the $[\bar{1}11]$ in (110) planes (see Appendix B again) the difference between the experimental curves and the term in b_0 , which is plotted as a dotted line, appears greater. It will be observed, however, that these curves approach zero and 180 degrees as the b_0 term. The other terms in (4) fall off much more rapidly near these points. Thus b_0 is the predominate term. An examination of the other terms in equation (4) will show that, at least by itself, none seems to account for the difference between the measured values and the b_0 term. The best fit of this difference seems to be a sine squared term, and again it is possible for this to arise as explained above.

A similar discussion applies for measurements made in the (100) plane. Figures 24 and 25 are two possible exceptions, however. Here, the accuracy of the measurement seems good enough to indicate a $\sin^2 2\theta$ term in addition to the $\sin 2\theta$ curve shown. An expansion of the power series for strain in the $[011]$ direction for magnetization in the (100) plane shows that a term of this type can arise for non-vanishing values of some of the higher terms in the a 's in equation (2) (a_2 for example). Nevertheless, even if this term is real (as it appears to be), and not due to some unknown error in the measurement, it still has little effect on the magnetostriction and will be neglected in the following discussion.¹ In the curves mentioned it introduces a maximum effect of about 5×10^{-7} .

1. It would be important, however, if the change in volume were being measured.

Evaluating the Constants

In the expressions for strain that have been used it is to be understood that equation gives the strain from a hypothetical unmagnetized state (such as would exist above the Curie temperature) to a state in which the crystal is magnetically saturated. Thus the constant a_0 is of no interest here since it really cannot be evaluated experimentally without taking the material through its Curie point. If instead of an unmagnetized state, the reference dimensions are those of a crystal saturated in the direction $\gamma_1, \gamma_2, \gamma_3$ then the expression for strain becomes

$$e' = A_{ij}(\alpha_1, \alpha_2, \alpha_3) B_i B_j - A_{ij}(\gamma_1, \gamma_2, \gamma_3) B_i B_j$$

Dropping the terms beyond a_1 and b_0 in each of the strain components and letting $a_1 = h_1$ and $b_0 = h_2$ to conform with a notation established by Becker, gives

$$e' = h_1(\alpha_i^2 B_i^2 - \gamma_i^2 B_i^2) + h_2(\alpha_i \alpha_j B_i B_j - \gamma_i \gamma_j B_i B_j) \quad i \neq j$$

Hereafter the prime will be dropped. If the expression is written so that i and j are summed over all the values 1, 2, 3 then

$$e = (h_1 - h_2)(B_i^2 \alpha_i^2 - B_i^2 \gamma_i^2) + h_2(\alpha_i \alpha_j B_i B_j - \gamma_i \gamma_j B_i B_j) \quad (5)$$

Consider now the magnetostriction in any arbitrary plane, Figure 5, where $I(\alpha_1, \alpha_2, \alpha_3)$ is the magnetization vector, $I_0(\gamma_1, \gamma_2, \gamma_3)$ the magnetization in the reference state and $e(\beta_1, \beta_2, \beta_3)$ is the strain being measured.

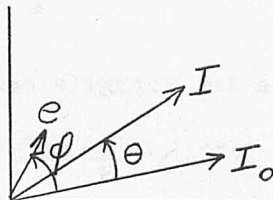


Fig. 5

Since $\cos(\varphi - \theta) = \alpha_i \beta_i$, $\cos^2(\varphi - \theta) = \alpha_i \beta_i \alpha_j \beta_j$

and $\cos^2 \varphi = \beta_i \gamma_i \beta_j \gamma_j$. Thus (5) becomes

$$e = (h_1 - h_2)(\beta_i^2 \alpha_i^2 - \beta_j^2 \gamma_j^2) + h_2 [\cos^2(\varphi - \theta) - \cos^2 \varphi] \quad (6)$$

Further, if $\delta_1, \delta_2, \delta_3$ are the direction cosines of a normal to the plane then:

$$\begin{aligned} \alpha_i \delta_i &= 0 \\ \alpha_i \gamma_i &= \cos \theta \\ \alpha_i \alpha_i &= 1 \end{aligned} \quad (7)$$

$$\begin{aligned} \beta_i \delta_i &= 0 \\ \beta_i \gamma_i &= \cos \varphi \\ \beta_i \beta_i &= 1 \end{aligned} \quad (8)$$

from which one can solve for the α 's and β 's and express e in terms of the angles θ and φ for a given plane, $\delta_1, \delta_2, \delta_3$ and given initial direction of magnetization, $\gamma_1, \gamma_2, \gamma_3$.

The expression for strain will now be evaluated for planes which are only slightly out of the (100) and (110), since these were used in the experiments, and for initial directions of magnetization slightly out of the [001]. Thus if χ, η, ξ are small angles

$$\begin{aligned} \gamma_1 &= \cos\left(\frac{\pi}{2} - \chi\right) \simeq \chi, \quad \gamma_2 = \cos\left(\frac{\pi}{2} - \eta\right) \simeq \eta \\ \text{and} \\ \gamma_3 &= \cos \xi \simeq 1 \end{aligned}$$

Now for a plane slightly out of the (100),

$$\delta_1 = \cos \psi \simeq 1, \quad \delta_2 = \cos\left(\frac{\pi}{2} - \lambda\right) \simeq \lambda \quad \text{and} \quad \delta_3 = \cos\left(\frac{\pi}{2} - \epsilon\right) \simeq \epsilon$$

where ψ, λ and ϵ are again small angles.

Using these in (7) and (8) one can readily establish that to a first approximation

$$\alpha_1 = -(\epsilon\alpha_3 + \lambda\alpha_2)$$

$$\alpha_2 = \sin\theta + \eta\cos\theta$$

$$\alpha_3 = \cos\theta - \eta\sin\theta$$

and the β 's are similar with φ replacing θ . Substituting these in (6) gives

$$e = -h_1 \sin^2\theta \cos 2\varphi + \frac{h_2}{2} \sin 2\theta \sin 2\varphi + (h_1 - h_2)\eta(2\sin^2\theta \sin 2\varphi - \sin 2\theta \cos 2\varphi) \quad (9)$$

Also, since there may be a small uncertainty in the direction in which the strain gages lie, one should replace φ with $\varphi + \Delta\varphi$ so (9) becomes

$$e = -h_1 \sin^2\theta \cos 2\varphi + \frac{h_2}{2} \sin 2\theta \sin 2\varphi + (h_1 - h_2)\eta(2\sin^2\theta \sin 2\varphi - \sin 2\theta \cos 2\varphi) + \Delta\varphi(2h_1 \sin^2\theta \sin 2\varphi + h_2 \sin 2\theta \cos 2\varphi) \quad (10)$$

Two cases of interest are $\varphi = 0$ (the strain is measured in the direction in which the crystal was initially saturated, i.e., nearly in the $[001]$ for which

$$e = -h_1 \sin^2\theta - (h_1 - h_2)\eta \sin 2\theta + h_2 \Delta\varphi \sin 2\theta \quad (11)$$

and $\varphi = 45$ degrees (the strain is measured close to the $[011]$) for which

$$e = \frac{h_2}{2} \sin 2\theta + 2(h_1 - h_2)\eta \sin^2\theta + 2h_1\Delta\varphi \sin^2\theta \quad (12)$$

From (11) it is observed that at $\theta = 90^\circ$ $e = -h_1$ and from (12) the difference in strain for the magnetization at 45° with that at 135° is h_2 . The constants measured in this way are obtained to about the same accuracy that the strain is measured (\pm about 3%) except in the special case where the one constant becomes very small compared with the other. In this case it is not correct to neglect higher powers of the small angles η , etc. For a plane slightly out of the (110) let

$$\delta_1 = \cos\left(\frac{\pi}{4} - \sigma\right) \approx \frac{1+\sigma}{\sqrt{2}}$$

$$\delta_2 = \cos\left(\frac{\pi}{4} - \tau\right) \approx \frac{1+\tau}{\sqrt{2}}$$

$$\delta_3 = \cos\left(\frac{\pi}{2} - \rho\right) \approx \rho$$

Proceeding in the previous way one finds in this plane

$$\begin{aligned} e = & -h_1 \sin^2\theta + \frac{(3h_1 + h_2)}{2} \sin^2\theta \sin^2\varphi + \frac{h_2}{2} \sin 2\varphi \sin 2\theta \\ & + \frac{(\eta - \chi)(h_1 - h_2)}{2\sqrt{2}} (3 \sin 2\theta \sin^2\varphi + 3 \sin 2\varphi \sin^2\theta - 2 \sin 2\theta) \\ & + \Delta\varphi \left[\frac{(3h_1 + h_2)}{2} \sin 2\varphi \sin^2\theta + h_2 \cos 2\varphi \sin 2\theta \right] \quad (13) \end{aligned}$$

For $\varphi = 0$ (strain near $[001]$)

$$e = -h_1 \sin^2\theta - \frac{(\eta - \chi)(h_1 - h_2)}{\sqrt{2}} \sin 2\theta + \Delta\varphi h_2 \sin 2\theta \quad (14)$$

and at $\theta = 90^\circ$ $e = -h_1$.

For $\varphi = 90^\circ$ (strain near $[\bar{1}10]$ direction)

$$e = \frac{(h_1 + h_2) \sin^2 \theta}{2} + \frac{(\eta - \chi)(h_1 - h_2) \sin 2\theta}{2\sqrt{2}} - \Delta\phi h_2 \sin 2\theta \quad (15)$$

and at $\theta = 90^\circ$ $h_2 = 2e - h_1$

This is not an accurate way to evaluate h_2 if $h_1 \sim 2e$. A better way is to measure the strain for $\phi = 54^\circ 44'$ (near $[\bar{1}11]$). Then

$$e = \frac{h_2 (\sin^2 \theta + \sqrt{2} \sin 2\theta)}{3} + \frac{(\eta - \chi)(h_1 - h_2) \sin^2 \theta + \Delta\phi [\sqrt{2}(3h_1 + h_2) \sin^2 \theta - h_2 \sin 2\theta]}{3} \quad (16)$$

At $\theta = 45^\circ$

$$e = \frac{h_2 (\frac{1}{2} + \sqrt{2})}{3} + \frac{(\eta - \chi)(h_1 - h_2)}{2} + \frac{\Delta\phi}{3} \left[\frac{(3h_1 + h_2)}{\sqrt{2}} - h_2 \right] \quad (17)$$

At $\theta = 135^\circ$

$$e = \frac{h_2 (\frac{1}{2} - \sqrt{2})}{3} + \frac{(\eta - \chi)(h_1 - h_2)}{2} + \frac{\Delta\phi}{3} \left[\frac{(3h_1 + h_2)}{\sqrt{2}} + h_2 \right] \quad (18)$$

and the difference between these two is $\frac{2}{3} h_2 (\sqrt{2} - \Delta\phi) \approx \frac{2\sqrt{2}}{3} h_2$

This gives an accurate measure of h_2 in this plane. (Except of course when $h_2 \rightarrow 0$)

The values obtained from the curves in the Appendix B are given in Table II.

Correction for Form Effect

It has been shown by several writers and verified by Becker¹ and Kornetski² that the shape of the sample measured is important in determining its magnetostriction. To compare the measurements made here

1. R. Becker, Z. Phys. 87, 547 (1934)

2. M. Kornetski, Z. Phys. 87, 560 (1934)

with a sample of the same material having no demagnetizing field, one must subtract the form effect. A method of calculating this is as follows: write expressions for the elastic energy and demagnetizing energy as a function of strain. Minimize the sum of these energies, with strain.

An expression for energy of the demagnetizing field of a spheroid as a function of strain already has been calculated by Powell, for the special case where the axes of the spheroid coincide with the cubic axes.¹ Taking his results and changing the notation somewhat, one finds that the energy to be minimized is (in so far as it depends upon strain and direction of magnetization)

$$E = \frac{C_{11}}{2} (A_{11}^2 + A_{22}^2 + A_{33}^2) + C_{12} (A_{11}A_{22} + A_{11}A_{33} + A_{22}A_{33}) + 2C_{44} (A_{12}^2 + A_{13}^2 + A_{23}^2) + \frac{\mu I^2}{2} (\alpha_2^2 A_{22} + \alpha_3^2 A_{33}) + \frac{\mu I^2}{6} (\alpha_2^2 A_{33} + \alpha_3^2 A_{22}) + \frac{2\mu I^2}{3} \alpha_2 \alpha_3 A_{23} \quad (19)$$

This expression is for spheroid cut and magnetized in the (100) plane. The C's are elastic constants, I the magnetization and for an oblate spheroid Powell gives

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

In terms of the principal axes, a, b and c, the eccentricity ϵ is given by

$$a = b = \frac{c}{\sqrt{1 - \epsilon^2}}$$

1. F. C. Powell, Proc. of the Cambridge Phil. Soc., 27, 561, (1931). We are not concerned with Powell's complete expression (6) since this is based on a special theory. We take, here, only that part which was

obtained from the expression $\frac{1}{2} \lambda_{ij} I_i I_j$ for the "demagnetizing energy"

μ should really be written $\mu = V \frac{\partial \lambda_{22}}{\partial A_{22}}$ (V, the volume).

Table II

% Si	I	Plane	Measured		With form effect Subtracted	
			$h_1 \times 10^6$	$h_2 \times 10^6$	$h_1 \times 10^6$	$h_2 \times 10^6$
2.52	1640	(100)	+41.5	-10.4	39.7	-11.2
2.52		(110)	40.7	-10.7	38.9	-11.5
3.03	1620	(100)	39.9	- 8.0	38.1	- 8.7
3.03		(110)	40.2	- 7.28	38.4	- 8.0
3.59	1590	(100)	40.0		38.3	
4.32	1560	(100)	33.2	- 1.4	31.5	- 2.1
4.83	1535	(110)	27.7	- 0.43	26.1	- 1.1
5.80	1490	(100)	6.45	3.03	4.9	2.4
5.80		(110)	6.7	2.93	5.2	2.3
7.79	1370	(110)	-13.6	4.28	-14.9	3.7

The solutions of $\frac{\partial E}{\partial A_{ij}} = 0$ are

$$A_{11} = \text{const.}$$

$$A_{22} = \text{const.} - \frac{\mu I^2 \alpha_2^2}{3(C_{11} - C_{12})}$$

$$A_{33} = \text{const.} - \frac{\mu I^2 \alpha_3^2}{3(C_{11} - C_{12})}$$

$$A_{23} = -\frac{\mu I^2 \alpha_2 \alpha_3}{6C_{44}}$$

$$A_{12} = A_{13} = 0$$

In the initial state the spheroid is assumed to be magnetized in the $[001]$ direction ($\alpha_3 = 1, \alpha_2 = \alpha_1 = 0$); consequently with this as a reference the form effect strains become

$$A_{22} = -\frac{\mu I^2 \alpha_2^2}{3(C_{11} - C_{12})} = -\frac{\mu I^2 \sin^2 \theta}{3(C_{11} - C_{12})}$$

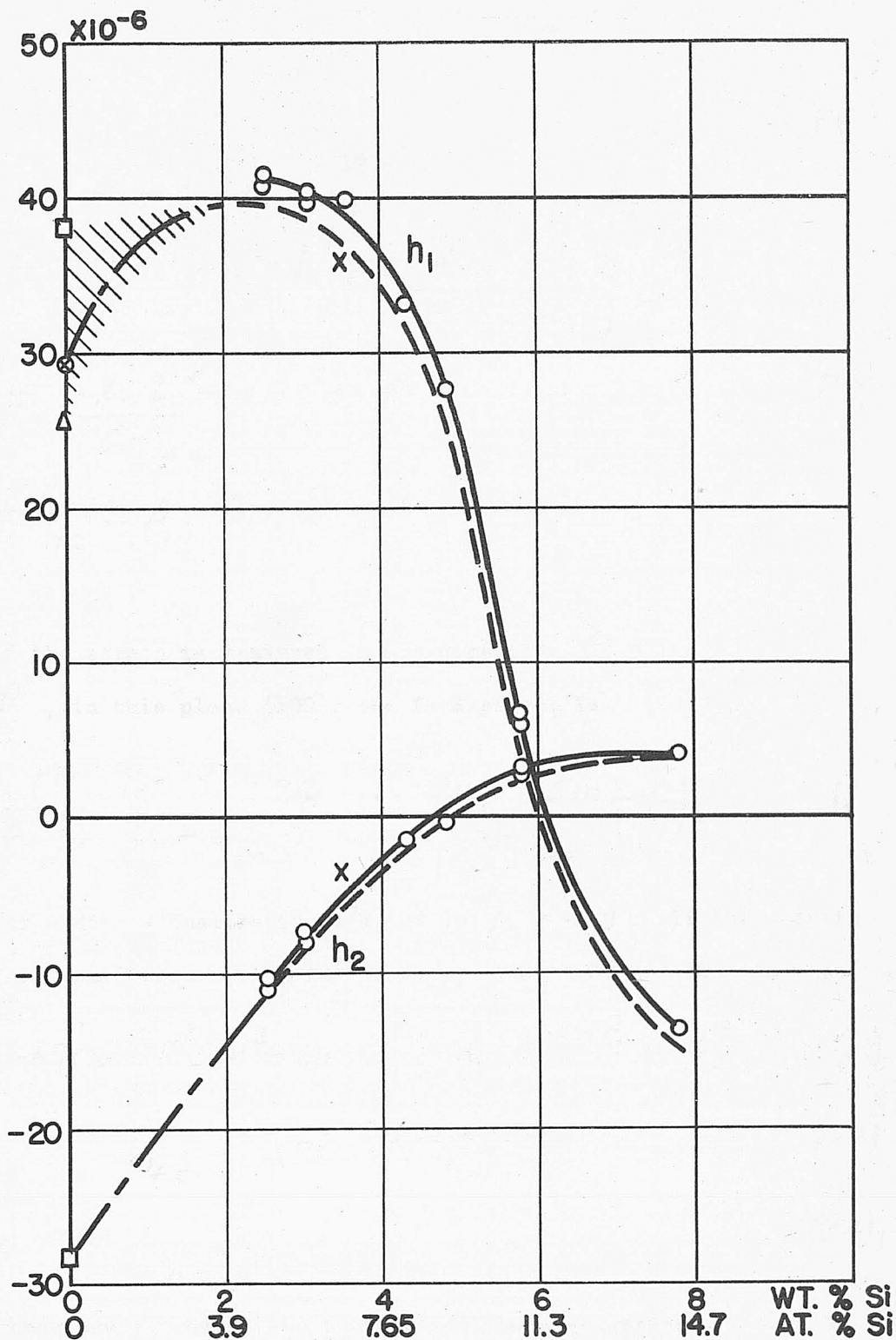


FIG.6

MAGNETOSTRICTION OF ANNEALED CRYSTALS

- \circ — MEASURED CURVE
- WITH FORM EFFECT SUBTRACTED
- \times — D.A. SHTURKIN, PHYSICS ABSTRACTS, 51, 1884 p 191 (1948)
- \square — S. KAYA AND H. TAKAKI, ANNIVERSARY VOL. DEDICATED TO PROF. HONDA (SENDAI 1936) 314
- \otimes — W. WEBSTER, PROC. ROYAL SOC. OF LONDON, 109, 570 (1925)
- Δ — K. HONDA AND Y. MASUYAMA, SCIENCE REPORTS TOHOKU IMP UNIV, 15 735 (1926)
- .- CURVE GOES TO VALUES BECKER CONSIDERS MOST ACCURATE.

$$A_{33} = -\frac{\mu I^2 (\alpha_3^2 - 1)}{3(C_{11} - C_{12})} = \frac{\mu I^2 \sin^2 \theta}{3(C_{11} - C_{12})}$$

$$A_{23} = \frac{-\mu I^2 \sin \theta \cos \theta}{6C_{44}} \quad (20)$$

$$A_{11} = A_{12} = A_{13} = 0$$

If the strain is measured in any direction, $B_2 = \sin \phi$,

$B_3 = \cos \phi$, in this plane (100), the form effect is

$$e_{f.e.} = \frac{\mu I^2}{3} \left(\frac{\sin^2 \theta \cos 2\phi}{C_{11} - C_{12}} - \frac{\sin 2\theta \sin 2\phi}{4C_{44}} \right) \quad (21)$$

For a dimensional ratio, a/c , of 10, $\mu = -1.9$. If the elastic constants for iron,¹ $C_{44} = 1.1 \times 10^{12}$ and $C_{11} - C_{12} = 0.95 \times 10^{12}$ are used

$$e_{f.e.} = -6.7 \times 10^{-13} I^2 \sin^2 \theta \cos 2\phi + 1.4 \times 10^{-13} I^2 \sin 2\theta \sin 2\phi \quad (22)$$

$$\text{Thus } h_{1f.e.} = 6.7 \times 10^{-13} I^2 \quad (23)$$

$$\text{and } h_{2f.e.} = 2.8 \times 10^{-13} I^2 \quad (24)$$

These are evaluated and subtracted from the measured results in Table II.

In evaluating the form effect for an oblate spheroid with major axes in an arbitrary plane, obviously the expression for energy of the

1. R. F. S. Hearmon, Rev. of Modern Phys., 18, 428 (1946). Since the elastic constants of Fe-Si apparently have not been measured, those for pure Fe are used as an approximation. The assumption that the elastic constants are independent of the direction of magnetization will be discussed in Part II.

demagnetizing field, which depends only upon the shape of the spheroid, will be of the same form as that in (19) if the direction cosines and strains involved in this part of the energy are referred to the principal axes of the spheroid rather than the cubic crystal axes. Thus in the general case

$$E = \frac{C_{11}}{2} (A_{11}^2 + A_{22}^2 + A_{33}^2) + C_{12} (A_{11}A_{22} + A_{11}A_{33} + A_{22}A_{33}) + 2C_{44} (A_{12}^2 + A_{13}^2 + A_{23}^2) + \frac{\mu I^2}{2} (\alpha_2'^2 A_{22}' + \alpha_3'^2 A_{33}') + \frac{\mu I^2}{6} (\alpha_2'^2 A_{33}' + \alpha_3'^2 A_{22}') + \frac{2\mu I^2}{3} \alpha_2' \alpha_3' A_{23}' \quad (25)$$

where the primed quantities refer to the spheroid axes and the unprimed to the cubic axes. It is necessary now to transform the unprimed quantities to the primed. Our interest here is only in a spheroid with principal axes in the (110) plane. This transformation can be obtained as follows. Choose the $[110]$, $[\bar{1}10]$ and $[001]$ directions as the X' , Y' , and Z' axes of the spheroid. Thus in relation to the cubic axes the primed axes are rotated 45° about Oz , and the relations

$$B_1' = \frac{B_1 + B_2}{\sqrt{2}}, \quad B_2' = \frac{B_2 - B_1}{\sqrt{2}} \quad \text{and} \quad B_3' = B_3$$

exist for the cosines of any direction. One can write equation (1) in the form

$$E = A_{ij} B_i B_j = A_{ij}' B_i' B_j' = \left(\frac{A_{11}' + A_{22}'}{2} - A_{12}' \right) B_1'^2 + \left(\frac{A_{11}' + A_{22}'}{2} + A_{12}' \right) B_2'^2 + A_{33}' B_3'^2 + \left(\frac{A_{11}' - A_{22}'}{2} \right) 2 B_1' B_2' + \left(\frac{A_{13}' - A_{23}'}{\sqrt{2}} \right) 2 B_1' B_3' + \left(\frac{A_{13}' + A_{23}'}{\sqrt{2}} \right) 2 B_2' B_3'$$

from which the transformation for the A's is immediately evident.

Substituting these in equation (25) gives

$$\begin{aligned}
 E = & \frac{C_{11}}{2} \left[\frac{(A'_{11} + A'_{22})^2}{2} + 2A'^2_{12} + A'^2_{33} \right] + C_{12} \left[\frac{(A'_{11} + A'_{22})^2}{4} - A'^2_{12} + (A'_{11} + A'_{22})A'_{33} \right] \\
 & 2C_{44} \left[\frac{(A'_{11} - A'_{22})^2}{4} + A'^2_{13} + A'^2_{23} \right] + \frac{\mu I^2}{2} (\alpha'^2_2 A'_{22} + \alpha'^2_3 A'_{33}) \\
 & + \frac{\mu I^2}{6} (\alpha'^2_2 A'_{33} + \alpha'^2_3 A'_{22}) + \frac{2\mu I^2}{3} \alpha'_2 \alpha'_3 A'_{23} \quad (26)
 \end{aligned}$$

Again, call θ the angle the magnetization makes with the $[001]$ so

$\alpha'_3 = \cos \theta$ and since the magnetization is confined in the (110) plane

$\alpha'_2 = \sin \theta$. As before, the solutions of the equations $\frac{\partial E}{\partial A'_{ij}} = 0$ are obtained.

$$A'_{11} = \text{const.} + \left[\frac{1}{C_{11} - C_{12}} - \frac{1}{2C_{44}} \right] \frac{\mu I^2 \cos^2 \theta}{6} \quad (28)$$

$$A'_{22} = \text{const.} + \left[\frac{1}{C_{11} - C_{12}} + \frac{1}{2C_{44}} \right] \frac{\mu I^2 \cos^2 \theta}{6} \quad (29)$$

$$A'_{33} = \text{const.} - \frac{\mu I^2 \cos^2 \theta}{3(C_{11} - C_{12})}$$

$$A'_{23} = - \frac{\mu I^2 \sin \theta \cos \theta}{6C_{44}} \quad (30)$$

$$A'_{13} = A'_{12} = 0$$

And since the reference state is still saturation in the $[001]$ the strain components referred to this state are

$$\begin{aligned} A'_{11} &= -\left[\frac{1}{c_{11}-c_{12}} - \frac{1}{2c_{44}}\right] \frac{\mu I^2}{6} \sin^2 \theta \\ A'_{22} &= -\left[\frac{1}{c_{11}-c_{12}} + \frac{1}{2c_{44}}\right] \frac{\mu I^2}{6} \sin^2 \theta \\ A'_{33} &= \frac{\mu I^2}{3(c_{11}-c_{12})} \sin^2 \theta \\ A'_{23} &= -\frac{\mu I^2 \sin \theta \cos \theta}{6c_{44}} \\ A'_{12} &= A'_{13} = 0 \end{aligned} \quad (27)$$

Whence, the strain in any direction ($\beta'_3 = \cos \varphi$, $\beta'_2 = \sin \varphi$, $\beta'_1 = 0$) in this plane is

$$\begin{aligned} e_{f.e.} &= A'_{ij} \beta'_i \beta'_j = \frac{\mu I^2}{3(c_{11}-c_{12})} \sin^2 \theta - \frac{\mu I^2}{6} \left[\frac{3}{c_{11}-c_{12}} + \frac{1}{2c_{44}} \right] \sin^2 \theta \sin^2 \varphi \\ &\quad - \frac{\mu I^2}{12c_{44}} \sin 2\theta \sin 2\varphi \end{aligned} \quad (28)$$

Finally, for $\varphi = 0$ (strain in $[001]$)

$$e_{f.e.} = \frac{\mu I^2 \sin^2 \theta}{3(c_{11}-c_{12})} \quad (29)$$

and for $\varphi = 54^\circ 44'$ (strain in $[\bar{1}11]$)

$$e_{f.e.} = -\frac{\mu I^2}{18c_{44}} (\sin^2 \theta + \sqrt{2} \sin 2\theta) \quad (30)$$

The appropriate corrections implied here are made in Table II. These corrections, in terms of h_1 and h_2 are the same as those for measurements in the (100) plane.

Longitudinal and Transverse Magnetostriction

In the past it has been the custom to make magnetostriction measurements by determining the so-called longitudinal and transverse magnetostriction, i.e. the strain in the direction of the magnetization and the strain in a direction transverse to the magnetization. Measurements of this type for a 3.03 and a 4.83 per cent alloy are shown in Appendix C and give further verification of the fact that two constants are adequate for the theoretical curve. The second, less accurate, method described under the section "Method of Measurement" was used to make the measurements since the direction of the strain measurement had to be changed for each point on the curve.

In one respect, the curves in Appendix C are still different from the usual magnetostriction curves for single crystals, in as much as the reference state is saturation in the $[001]$ rather than the demagnetized condition.

The equations for the longitudinal and transverse effect as used here are obtained as follows. From equation (6) the longitudinal magnetostriction is

$$\epsilon_l = (h_1 - h_2)(\alpha_1^2 \alpha_2^2 - \alpha_3^2) + h_2 \sin^2 \theta \quad (31)$$

since $\alpha_1 = \beta_1$ and $\gamma_3 = 1, \gamma_1 = \gamma_2 = 0$

In the (100) plane $\alpha_1 = 0, \alpha_2 = \sin \theta, \alpha_3 = \cos \theta$

curves. The calibration was obtained by making the peak values of the and measured curve agree with the calculated value.

$$e_\phi = 2(h_1 - h_2) \sin^4 \theta - (h_1 - 2h_2) \sin^2 \theta \quad (32)$$

In the (110) plane $\alpha_2 = \frac{\sin \theta}{\sqrt{2}} = -\alpha_1$, $\alpha_3 = \cos \theta$

and

$$e_\phi = \frac{3}{2}(h_1 - h_2) \sin^4 \theta - (h_1 - 2h_2) \sin^2 \theta \quad (33)$$

With the magnetization transverse to the direction in which the strain is measured ($\theta = \phi + \frac{\pi}{2}$)

$$e_\phi = (h_1 - h_2)(\beta_1^2 \alpha_1^2 - \beta_3^2) - h_2 \cos^2 \phi \quad (34)$$

In the (100) plane $\alpha_1 = \beta_1 = 0$, $\beta_2 = -\alpha_3 = \sin \phi$ and $\beta_3 = \alpha_2 = \cos \phi$

$$e_\phi = (h_1 - 2h_2) \cos^2 \phi - 2(h_1 - h_2) \cos^4 \phi \quad (35)$$

In the (110) plane $-\beta_1 = \beta_2 = \frac{\sin \phi}{\sqrt{2}}$, $\beta_3 = \cos \phi$
 $-\alpha_1 = \alpha_2 = \frac{\cos \phi}{\sqrt{2}}$, $\alpha_3 = -\sin \phi$ and

$$e_\phi = \frac{(h_1 - 3h_2)}{2} \cos^2 \phi - \frac{3}{2}(h_1 - h_2) \cos^4 \phi \quad (36)$$

Qualitatively there is good agreement between the magnetostriction calculated from these equations and the measured points. No independent calibration was made for the measurements shown in these

curves. The calibration was obtained by making the peak value of the measured curve agree with the calculated value.

Effect of Order and Disorder

It has been shown by Goldman and Smoluchowski¹ that ordering in an alloy can appreciably change the magnetostriction. Since, over a certain range of composition, the Fe-Si system is known to order one would expect to measure different constants for annealed and quenched crystals. The values given in Table II and plotted in Fig. 6 are for annealed material, furnace cooled, and presumably ordered for compositions for which ordering can take place.

Above 12.5 atomic per cent silicon ordering definitely has been observed by a number of investigators and the superstructure is of the Fe_3Si type^{2,3,4}. Below 12.5 per cent no superlattice lines have been detected in the X-ray photographs, but there are reasons to believe that ordering, at least of short range character, commences at about 9 or 10 atomic per cent since there is a discontinuity in the curve of lattice constant vs. per cent silicon near this composition^{5,6}. Also a discontinuity appears at 9.86 atomic per cent silicon in a plot of the magnetic anisotropy constant against per cent silicon.⁷

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1. J. E. Goldman and R. Smoluchowski, Phys. Rev. 75, 140 (1949)
 2. G. Phragmén, Journal of the Iron and Steel Inst. 114, 397 (1926)
 3. Eric R. Jette and Earl S. Greiner, Trans. A.I.M.M.E. 105, 259 (1933)
 4. Margaret C. M. Farquhar, H. Lipson and Adrienne R. Weill, Journal of the Iron and Steel Inst. 152, 457 (1945)
 5. Eric R. Jette and Earl S. Greiner, Loc. cit.
 6. Margaret C. M. Farquhar, etc., Loc. cit.
 7. L. P. Tarasov, Phys. Rev. 56, 1231 (1939)

Measurements of the magnetostriction for specimens quenched in oil, shown in Table III and plotted in Figure 7, add credence to the belief that ordering begins to take place in the region of 9 to 10 atomic per cent.

Due to the brittleness of the specimens it was not possible to subject these crystals to a severe quench. For the heat treatment indicated cracks appeared in the specimens which

TABLE III

<u>Wt. % Si</u>	<u>Plane</u>	<u>Quenching Temperature</u>	<u>$h_1 \times 10^6$</u>	<u>$h_2 \times 10^6$</u>
2.52	(110)	900°C	39.4	-10.7
4.32	(100)	900°C	34.0	- 1.0
5.80	(110)	900°C	12.0	3.3
5.80	(100)	700°C	13.0	4.9
7.79	(110)	900°C	No significant change from annealed material.	

MAGNETOSTRICTION CONSTANTS OF CRYSTALS QUENCHED IN OIL

made magnetostriction measurements difficult. For this reason the above values are not as accurate as measurements on the annealed specimens. Comparing the values with those of Table II, one sees that the only significant change that has occurred is the value of h_1 for the 5.8 wt. per cent alloy. The constant h_1 for the quenched alloy is nearly double that for the annealed; h_2 has changed but little and within the accuracy of the measurements no change has occurred in the constants for specimens with smaller amounts of silicon. Also no change appears in the values for the 7.79 wt. per cent alloy. The latter in the annealed condition should possess some order. Since no difference large enough to measure was detected in its magnetostriction after an oil quench from 900°C one may suppose either the magnetostriction of this alloy is insensitive to order or that

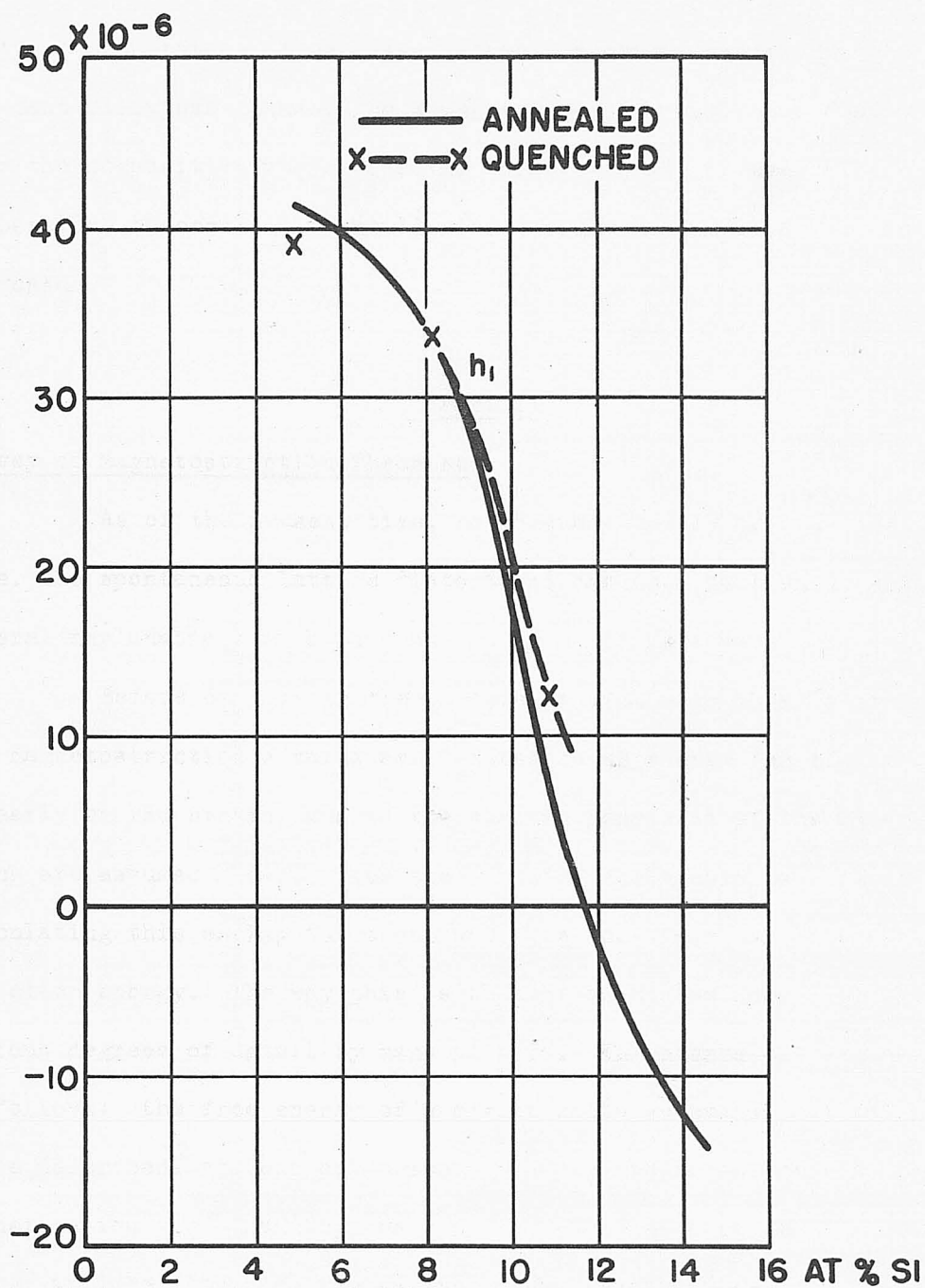


FIG. 7

900° is still below the critical temperature where disorder sets in. The critical temperature, from magnetostriction measurements, for the 5.8 wt. per cent alloy was found to be somewhere in the neighborhood of 450°C.

Near the composition where ordering first appears, however, the critical temperature theoretically should rise rapidly as the alloying element is increased.¹

Part II

Survey of Magnetostriction Theories

As of the present time, no adequate theory of magnetostriction (i.e. the spontaneous lattice distortion) has been published, although several explanations with some amount of merit have been proposed.

Before discussing these it is desirable to show in general how the magnetostriction strains are related to an energy function which depends linearly on the strain, and to the elastic constants of the material, which are assumed known. Thus the scope of the theory is reduced to calculating this energy function, which is sometimes called the magnetostriction energy. The way this is brought about has been explained in various degrees of detail by many authors. In essence the argument goes as follows: the free energy of a magnetically saturated crystal is assumed to be described, at constant temperature, by the direction cosines of the magnetization vector and by the components of the strain tensor. This energy will be thought of as an energy per unit volume of the unstrained crystal, or in other words, as referred to a given mass. For definiteness, the unstrained state will be taken as one for which the crystal lattice

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1. C. E. Easthope, Proc. Cambridge Phil. Soc. 33, 502 (1937)

still possesses its cubic shape. Only cubic crystals will be discussed here.

If one does work on the crystal by applying stresses σ_{ij} which distort it, the change in free energy, for the case in which the magnetization vector is kept fixed, is

$$dE = \frac{\partial E}{\partial A_{ij}} dA_{ij} = \sigma_{ij} dA_{ij}$$

For small stresses, a ferromagnetic material that is saturated obeys a Hooke's law, in that for a fixed direction of magnetization the change in strain is proportional to the change in stress. In other words,

$$\frac{\partial \sigma_{ij}}{\partial A_{lm}} = \frac{\partial^2 E}{\partial A_{lm} \partial A_{ij}} = C_{lmij} \quad \text{where the } C\text{'s are constants,}$$

the elastic constants, and in as far as this is true no powers of strain greater than two are involved in the expression for free energy.

One can write

$$E = E_0 + E_1 + E_2 \quad (1)$$

where E_0 is independent of strain and includes the ordinary crystalline anisotropy energy; E_1 depends linearly on the strain and may be referred to as the magnetostriction energy and E_2 is the elastic energy quadratic in the strain components. It is customary to assume the elastic constants to be independent of the direction of magnetization.¹

Under equilibrium conditions, subject to the constraints, the energy is a minimum. If the only constraint on the system is that the direction of magnetization is fixed, then

$$\frac{\partial E}{\partial A_{ij}} = 0 \quad (2)$$

For a cubic crystal the elastic energy is

1. There seems to be a paucity of experimental data on this. It appears reasonable to assume, however, that the orientation of the electron spins will have no great effect on the electrostatic forces which determine the elastic constants.

$$E_2 = \frac{C_{11}}{2}(A_{11}^2 + A_{22}^2 + A_{33}^2) + C_{12}(A_{11}A_{22} + A_{11}A_{33} + A_{22}A_{33}) + 2C_{44}(A_{12}^2 + A_{13}^2 + A_{23}^2) \quad (3)$$

Only six components appear because $A_{ij} = A_{ji}$.

The solutions of (2) are

$$A_{11} = \frac{C_{12}}{(C_{11} + 2C_{12})(C_{11} - C_{12})} \left(\frac{\partial E_1}{\partial A_{22}} + \frac{\partial E_1}{\partial A_{33}} \right) - \frac{(C_{11} + C_{12})}{(C_{11} + 2C_{12})(C_{11} - C_{12})} \frac{\partial E_1}{\partial A_{11}} \quad (4)$$

$$A_{12} = -\frac{1}{4C_{44}} \frac{\partial E_1}{\partial A_{12}}$$

etc.

It is worth mentioning here that since E_1 is a linear function of strain the strain components due to any particular part of E_1 can be calculated separately, as was done for the form effect in Part I.

One can now write E_1 as a series involving only the terms in the direction cosines which crystal symmetry will allow. For a cubic crystal, in the manner of Becker and Doring,¹

$$E_1 = A_{11}(b_0 + b_1\alpha_1^2 + b_2S + b_3\alpha_1^4 + \dots) + A_{12}\alpha_1\alpha_2(m_1 + m_2\alpha_3^2 + m_3S + m_4\alpha_3^4 + \dots) + \text{etc. for } A_{22}, A_{33}, A_{13} \text{ and } A_{23} \text{ using the same constants} \quad (5)$$

where $S = \alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2$ constants.

and the k's and m's are

Equation (5) was obtained, however, from considerations of the lattice symmetry only. But for a finite crystal, the shape of the boundary should be considered too. It, of course, should not be cubic since an ellipsoidal boundary is required for uniform magnetization. Of these, only

1. Becker and Doring, Ferromagnetismus, (Julius Springer, Berlin, 1939) p 136

the sphere will have all the symmetry of a cube, so strictly speaking (5) is for a crystal with a cubic lattice and a spherical boundary. For other boundaries one will have to understand (5) as the magnetostriction energy after the part depending on shape (i.e. the dimensions of the ellipsoid) has been subtracted. The shape part arises from the term in the energy contributed by the demagnetizing field and is therefore quadratic in the components of magnetization.¹

Our interest here is in the silicon-iron alloys, for which as the preceding experiments have shown, terms beyond k_1 and m_1 in (5) are unimportant, so

$$E_1 = A_{11}(b_0 + b_1 \alpha_1^2) + A_{22}(b_0 + b_1 \alpha_2^2) + A_{33}(b_0 + b_1 \alpha_3^2) + m_1(A_{12}\alpha_1\alpha_2 + A_{13}\alpha_1\alpha_3 + A_{23}\alpha_2\alpha_3) \quad (6)$$

A substitution in (4) gives

$$A_{ii} = \text{const.} - \frac{k_1 \alpha_i^2}{C_{11} - C_{12}} \quad (7)$$

$$A_{ik} = - \frac{m_1 \alpha_i \alpha_k}{4C_{44}}$$

Thus from the expressions for strain given in Part I, one obtains the rather familiar result

$$h_1 = \frac{-k_1}{C_{11} - C_{12}} \quad (8)$$

$$h_2 = \frac{-m_1}{4C_{44}}$$

The theoretical problem for iron (and Si-Fe alloys) is thus simplified to the following: one must find an energy which is quadratic in the direction cosines of magnetization, as in (6), and whose magnitude is

(1) For an oblate ellipsoid magnetized only in the equatorial plane, the part depending on shape that must be added to (5) is given in the second line of equation (25), Part I (in so far as it depends on direction of magnetization).

such as to satisfy (8) for the experimental values of h_1 and h_2 . We can now proceed to discuss some attempts to do this.

It is natural that the first attempts to develop a magnetostriction theory were based upon classical magnetic dipole interaction between atoms.^{1,2} The dipoles were assumed situated at the lattice points of the crystal, the energy between a parallel pair was written as a linear function of strain and the strain dependent part was summed over all pairs in the lattice, leading, correctly for the case of iron, to a quadratic energy expression in the direction cosines.

The lattice sum for a spherical crystal converges rapidly enough so that the sum can be performed by taking a moderate number of terms. McKeehan has made an accurate calculation of this.³ Using his result (0.36 instead of the value 0.4 which Becker uses for S)⁴ one observes from equation (4) of Becker's paper that according to this theory $k_1 = -6SI^2$ and $m_1 = 8SI^2$ or ⁵

$$h_1 = 6.65 \times 10^{-6}$$

$$h_2 = -1.9 \times 10^{-6}$$

(9)

(I is the intensity of magnetization.) F. C. Powell⁶ pointed out, however, that Becker's results were correct only for a spherical crystal.

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(1) N. Akulov, Z. Physik, 52, 389 (1928)

(2) R. Becker, Z. Physik, 62, 253 (1930)

(3) L. W. McKeehan, Phys. Rev., 43, 1022 (1933)

(4) R. Becker, Z. Physik, 62, 253, (1930)

(5) It should be noted that Becker's actual calculation of the strain is not quite correct since he uses an isotropic expression for the elastic energy instead of the one appropriate for a cubic crystal.

(6) F. C. Powell, Proc. of the Cambridge Phil. Soc., 27, 561 (1931)

The lattice sum for the energy between pairs of dipoles is only conditionally convergent. For the general case one should actually perform the sum over all pairs and the result will depend upon the boundary of the crystal. Powell did this by using the Lorentz method which replaces the summation for distant pairs by the interaction of a given atom with the field of the distant atoms, obtained by integration over the pole density: i.e. for a particular dipole, one first sums its interaction with all dipoles in a small sphere surrounding it, obtaining approximately Becker's result. Then to account for its interaction with dipoles outside the sphere, assume a pole density, $-I_n$, on the surface of the sphere and a pole density, I_n , on the surface of the crystal, where I_n is the normal component of the magnetization. The density on the crystal boundary is precisely what leads to the "form effect".

For a spherical boundary the effects of the two pole densities cancel; measurements, however, are always made on crystals of small demagnetizing factor rather than the spherical shape. Powell showed that Akulov's calculations are the appropriate ones for the case of no demagnetizing factor.¹

In Part I the magnetostriction due to the "poles" on the boundary of the crystal, the form effect, was calculated. The form effect (in the sense of Becker) gives the difference between the magnetostriction of an arbitrary spheroid and that for the limiting case of a spheroid with no demagnetizing factor in the direction of magnetization. The correction which Powell makes is from a sphere to an arbitrary spheroid. But

1. Akulov's numerical values are incorrect due to a mistake which Powell points out.

obviously, $C_{\text{spheroid}} = C_{\text{sphere}} - C_{\text{f.e. sphere}} + C_{\text{f.e. spheroid}}$

so Powell's correction also can be explained in terms of form effect which can be calculated from rather fundamental principles without any consideration of the atomic nature of the interaction.¹

The measurements in Part I are given for zero form effect. To determine the form effect of a sphere, equation (19) in Part I can be referred to. This gives the elastic energy and the anisotropic part of the demagnetizing energy for the special case of an oblate ellipsoid magnetized in the (100) plane (i.e. $\alpha_1 = 0$). By comparing this with (6) and using $\mu = \frac{-12\pi}{5}$ appropriate for a sphere one obtains $k_1 \text{ f.e. sphere} = \frac{-4\pi I^2}{5}$ and

$m_1 \text{ f.e. sphere} = \frac{-8\pi I^2}{5}$, so from (8) for an iron crystal

$$h_1 \text{ form effect, sphere} = 7.75 \times 10^{-6} \quad (10)$$

$$h_2 \text{ form effect, sphere} = 3.34 \times 10^{-6}$$

These can be subtracted from (9) to compare the theory with

measurements. The result is

$$h_1 = -1.1 \times 10^{-6} \quad (11)$$

$$h_2 = -5.2 \times 10^{-6}$$

Comparing these with the values for iron on Fig. 6, Part I, one observes that the h_1 is of the wrong sign and almost negligible. The h_2 calculated is too small by a factor of 5, although the corresponding calculation for iron-silicon in the region where h_2 is small would give an

1. R. Becker, Z. Physik, 87, 547 (1934)

appreciable value.¹

Actually these figures may be somewhat misleading. A better test of Becker's calculations is to add (10) to the measured constants, thereby obtaining what one would expect to measure on a spherical crystal, and to compare this with the calculations of (9). Thus h_1 and h_2 of (9) both have the correct sign for iron but are too small by factors of 6 and 13 respectively.

Goldman and Smoluchowski² have made calculations which, for an iron-cobalt alloy, show the effect of order on the magnetic dipole interactions. They modify the theory so as to associate with each atom a dipole moment that depends not alone upon the type of atom but also upon its atomic environment. Only the effects of nearest and second nearest neighbors are considered. By this means, they calculate, for the $[100]$ direction, the difference between the magnetostriction of an ordered and a disordered alloy. Percentagewise, it agrees within a factor of two with measurements they made on polycrystalline material. In actual magnitude, however, the theory, as for iron, would apparently lead to values much too small.

It is sometimes suggested that calculations based on a distribution of dipole moment over the atom as given by the actual wave functions of the electrons responsible for magnetism (in place of considering it concentrated at the lattice point which is allowable only for non-overlapping, spherically symmetrical wave functions) might remove the quantitative defect of the magnetic dipole theory. This does not seem likely, however, at least

1. Powell reports the calculations for iron as being too small by a factor of 13. This is because he evaluates $h_1 - h_2$ to compare with experiment.
2. J. E. Goldman and R. Smoluchowski, Phys. Rev. 75, 140 (1949)

for iron and iron-silicon alloys, since the magnetostriction of these is quadratic in the direction cosines of magnetization. Neglecting any overlapping, the effect of distributing the dipole moment is to introduce quadrupole and higher order interactions.¹ These if large enough to affect the magnetostriction would probably lead to appreciable terms of higher order in the direction cosines and ruin the symmetry properties of the theory.

Thus one is led to look for some other interaction as the pre-dominate cause of magnetostriction. In doing this it is necessary to pass over exchange interaction, which causes ferromagnetism, since this, in the usual approximation, has no anisotropy and as such could produce only a volume change when the magnitude of the spontaneous magnetization changes.

A suggestion of Van Vleck² that magnetostriction arises from spin-orbit coupling in the atom in the manner that magnetic anisotropy is thought to arise seems quite plausible. The idea here is that the anisotropic electrostatic coupling of orbital momenta between atoms in the crystal is the important factor. This is communicated to the electron spins by the spin-orbit coupling and thus the energy becomes a function of the direction of magnetization. Apparently, no calculations on this have been attempted. The magnetic anisotropy calculations have been tried by Van Vleck² and Brooks³ and their approximations leave the result uncertain within a factor of 10. The magnetostriction calculation would require a derivative of this energy.

(1) For example see L. W. McKeehan, Phys. Rev., 52, 18 (1937)

(2) J. H. Van Vleck, Phys. Rev. 52, 1178 (1937)

(3) Harvey Brooks, Phys. Rev. 58, 909 (1940)

A recent observation by Snoek¹ concerning magnetostriction is that several alloys made up of ferromagnetic components and possessing a simple close packed lattice structure have zero magnetostriction at the composition giving one mean Bohr magneton. He postulates that this is due to the integral number of magnetons, but as yet, no theory concerning this has been published.

Magnetic Dipole Interaction for Fe-Si

In general, if the free energy of the crystal, or at least the part depending upon the magnetization vector, can be obtained by a summation over all pairs of atoms of an expression \mathcal{E}_{MN} for the interaction of the M^{th} and N^{th} atoms in their respective lattice positions, then the magnetostriction energy is

$$E_i = \frac{1}{2V_0} \sum_{M,N} \frac{\partial \mathcal{E}_{MN}}{\partial X_k} \Delta X_k = \frac{1}{2V_0} \sum_{M,N} \frac{\partial \mathcal{E}_{MN}}{\partial X_k} A_{kl} X_l \quad (12)$$

The X_k are the coordinates, taken along the unstrained cubic axes, of the N^{th} lattice point with the M^{th} taken as the origin; V_0 is the volume of the unstrained crystal. The repeated small subscripts are to be summed over 1, 2, 3, and the summation sign indicates a sum over all pairs in the crystal.

For the present problem of magnetic dipole interaction

$$\mathcal{E}_{MN} = \frac{-3\sigma^2 \cos^2 \varphi}{r_{MN}^3} = \frac{-3\sigma^2}{r_{MN}^5} (\alpha_1 X_1 + \alpha_2 X_2 + \alpha_3 X_3)^2 \quad (13)$$

giving the energy between two parallel dipoles of moment σ and direction cosines α_1, α_2 , and α_3 .

1. J. L. Snoek, Nature 163, 837 (1949)
2. The first term in the energy $\frac{\sigma^2}{r^3}(1-3\cos^2\varphi)$ is of no interest here since it is isotropic.
3. For this problem the dipoles are all assumed parallel, which of course, is strictly true only at absolute zero of temperature.

It will now be assumed that the dipoles are associated only with the iron atoms so (13) applies when M and N are both iron and is zero otherwise.

Thus for the Fe-Si alloy

$$E_i = \frac{1}{2V_0} \sum_{M,N} P_M P_N \frac{\partial \epsilon_{MN}}{\partial X_R} A_{kl} X_l \quad (14)$$

where P_M and P_N are the respective probabilities of finding iron atoms at the M^{th} and N^{th} lattice positions.

For a completely random alloy having atomic fraction of silicon f , $P_M = P_N = (1-f)$ and for the approximation being made, $(1-f) = \frac{I}{I_0}$, the ratio of intensity of magnetization to that for iron, a relation which is nearly true for small amounts of silicon. So the constants in both (9) and (11) become multiplied by $\left(\frac{I}{I_0}\right)^2$.

In Fig. 8, the contribution this makes to the constant h_2 is subtracted from the measured values, leaving the resultant for the random alloys which is yet to be explained. The contribution to h_1 is considered negligible.

To carry out the calculation for the ordered alloys it would be necessary to know the order parameters.

A Semi-empirical Theory for Fe-Si

In the absence of exact knowledge as to the predominate cause of magnetostriction one can only attempt to explain the resultant curves of Fig. 8 in a semi-empirical manner.

It is supposed that short range forces are involved, which simplify the lattice sum in as much as only immediate neighbors are important. If so desired one may think of these forces arising in the manner

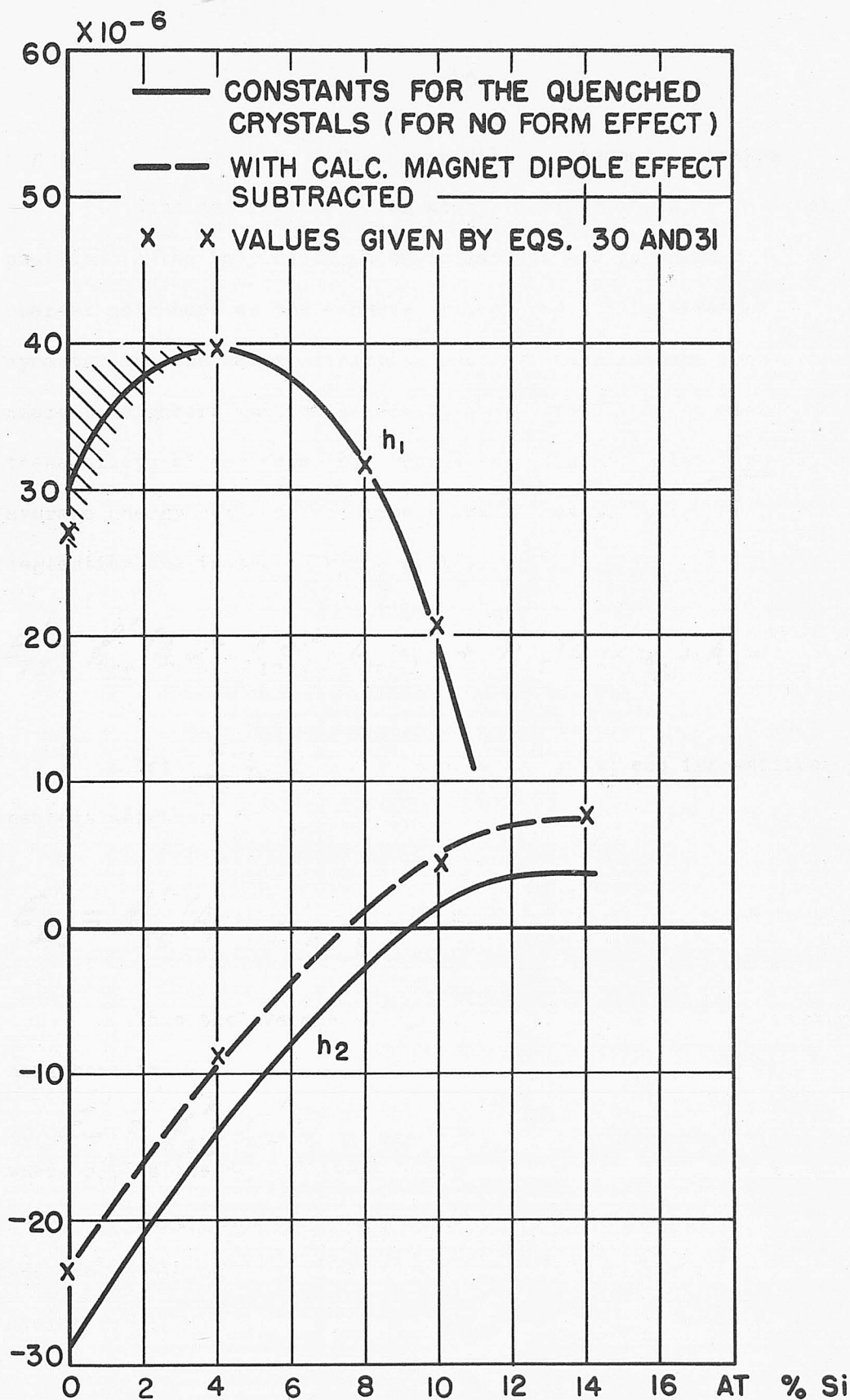


FIG. 8

suggested by Van Vleck.¹ The atoms will be assumed to interact in pairs.

Consider first an iron atom, imagined as being in a body centered position in the body centered cubic lattice and surrounded by its eight nearest neighbors on the corners of the cube. This arrangement has cubic symmetry and the magnetostriction energy of the average iron atom with its nearest neighbors must be generally of the form (5), or specifically for these alloys of the form (6). For a specific alloy let $\frac{E'_{II}}{8}$ represent the average energy between iron-iron pairs of nearest neighbors, and, neglecting the isotropic k_0 term, let

$$E'_{II} = k'_{II} (A_{11}\alpha_1^2 + A_{22}\alpha_2^2 + A_{33}\alpha_3^2) + m'_{II} (A_{12}\alpha_1\alpha_2 + A_{13}\alpha_1\alpha_3 + A_{23}\alpha_2\alpha_3) \quad (15)$$

Let $\frac{E'_{IS}}{8}$ be the average energy between iron-silicon pairs of nearest neighbors and

$$E'_{IS} = k'_{IS} (A_{11}\alpha_1^2 + A_{22}\alpha_2^2 + A_{33}\alpha_3^2) + m'_{IS} (A_{12}\alpha_1\alpha_2 + A_{13}\alpha_1\alpha_3 + A_{23}\alpha_2\alpha_3) \quad (16)$$

Thus the average energy between an iron atom and its eight nearest neighbors is

$$E'_I = p'_{II} E'_{II} + (1 - p'_{II}) E'_{IS} \quad (17)$$

where p'_{II} is the probability that a nearest neighbor of an iron atom is iron.

Next, consider the six second nearest neighbors of an iron atom and use double primes to describe the corresponding quantities here. The average energies between pairs are $\frac{E''_{II}}{6}$ and $\frac{E''_{IS}}{6}$. Also

$$E''_I = p''_{II} E''_{II} + (1 - p''_{II}) E''_{IS} \quad (18)$$

1. Loc. cit.

Only the nearest and next nearest neighbors will be considered although one could readily include more. This makes the magnetostriction energy of an iron atom with its neighbors given by

$$E_I = E'_I + E''_I = p'_{II} E'_{II} + p''_{II} E''_{II} + (1-p'_{II}) E'_{IS} + (1-p''_{II}) E''_{IS} \quad (19)$$

One must now express the same thing for a silicon atom. In doing this, silicon-silicon interactions will be ignored since it is assumed that these do not depend upon the magnetization. Thus

$$E_S = E'_S + E''_S = p'_{SI} E'_{IS} + p''_{SI} E''_{IS} \quad (20)$$

where p_{SI}^I is the probability a nearest neighbor of a silicon atom is iron, p_{SI}^{II} the probability any one of the second nearest neighbors is iron.

For N_I iron atoms per unit volume and N_S silicon atoms, the anisotropic magnetostriction energy per unit volume is

$$E_1 = \frac{N_I}{2} E_I + \frac{N_S}{2} E_S \quad (21)$$

The one-half is the usual factor which prevents each pair from being counted twice. This expression can be simplified by observing that $8N_S p_{SI}^I$ gives the number of pairs of nearest neighbors having one iron and one silicon atom; $8N_I (1-p_{II}^I)$ gives the same thing, so these are equal. Also $N_S p_{SI}^{II} = N_I (1-p_{II}^{II})$.

Therefore the magnetostriction energy becomes

$$E_1 = \frac{N_I}{2} \left[p'_{II} (E'_{II} - 2E'_{IS}) + p''_{II} (E''_{II} - 2E''_{IS}) + 2E'_{IS} + 2E''_{IS} \right] \quad (22)$$

For an annealed crystal the probabilities p_{II}^I and p_{II}^{II} are determined both by the composition and the state of order of the material, for which additional assumptions would have to be made. For the present,

only the quenched, presumably random, alloys will be considered. For these

$p_{II}^I = p_{II}^{II} = 1-f$ where f is the atomic fraction of silicon. Also replace

N_I by $N(1-f)$ where N is the total number of atoms per unit volume.

Thus for a random alloy

$$E_1 = \frac{N(1-f)}{2} [E'_{II} + E''_{II} - f(E'_{II} + E''_{II} - 2E'_{IS} - 2E''_{IS})] \quad (23)$$

and from (8), (15) and (16)

$$h_1 = -\frac{N(1-f)}{2(c_{11}-c_{12})} [k'_{II} + k''_{II} - f(k'_{II} + k''_{II} - 2k'_{IS} - 2k''_{IS})] \quad (24)$$

$$h_2 = -\frac{N(1-f)}{8C_{44}} [m'_{II} + m''_{II} - f(m'_{II} + m''_{II} - 2m'_{IS} - 2m''_{IS})] \quad (25)$$

It is understood that the parameters k and m , and also the elastic constants refer to values for a particular alloy, and may be different for different alloys. The k 's and m 's will most certainly depend upon the distance between the interacting atoms, which varies with composition. This change in lattice constant with composition is large, compared with that produced by the magnetostrictive strain, and may be large enough to appreciably change the interaction energy between pairs. Further, one might imagine the interaction of a pair to be dependent upon the surroundings of the pair and consequently upon composition. Also the elastic constants may be expected to vary.

In view of this, one can imagine the dependence on f of the parameters in (24) and (25) as being expressed by a power series, and it is hoped that over the small range of silicon considered here only a constant and a linear term in f are needed. As far as effects due to

changes of the lattice constant are concerned this is reasonable because, at least up to 9 or 10 atomic per cent silicon, the lattice constant varies linearly with the percentage of silicon.^{1,2} Beyond 10 per cent the lattice constant apparently has not been measured for the disordered alloys.

To simplify the notation let $k_{II} = k_{II}^I + k_{II}^{II}$, etc. Then according to the above assumption

$$\begin{aligned}\frac{b_{II}}{c_{II}-c_{I2}} &= \frac{b_{II0}}{(c_{II}-c_{I2})_0} (1 + K_{II}f) \\ \frac{b_{IS}}{c_{II}-c_{I2}} &= \frac{b_{IS0}}{(c_{II}-c_{I2})_0} (1 + K_{IS}f) \\ \frac{m_{II}}{c_{44}} &= \frac{m_{II0}}{c_{440}} (1 + M_{II}f) \\ \frac{m_{IS}}{c_{44}} &= \frac{m_{IS0}}{c_{440}} (1 + M_{IS}f)\end{aligned}\quad (26)$$

where the K's and M's are four new constants and the subscript zero refers to values for nearly pure iron.

The equations for h become

$$h_1 = -\frac{N(1-f)}{2(c_{II}-c_{I2})_0} \left[b_{II0} + (b_{II0}K_{II} + 2b_{IS0} - b_{II0})f + (2b_{IS0}K_{IS} - b_{II0}K_{II})f^2 \right] \quad (27)$$

$$h_2 = -\frac{N(1-f)}{8c_{440}} \left[m_{II0} + (m_{II0}M_{II} + 2m_{IS0} - m_{II0})f + (2m_{IS0}M_{IS} - m_{II0}M_{II})f^2 \right] \quad (28)$$

As shown in Fig. 8 these fit the measured curves (with the calculated magnetic dipole effect subtracted) very well if

1. Eric R. Jette and Earl S. Greiner, Trans. A.I.M.M.E. 105, 259 (1933).
2. Margaret C. M. Farquhar, H. Lipson and Adrienne R. Weill, Journal of the Iron and Steel Inst. 152, 457 (1945)

$$h_{II_0} = -6 \times 10^{-16}$$

$$h_{II_0} K_{II} + 2h_{IS_0} - h_{II_0} = -138 \times 10^{-16}$$

$$2h_{IS_0} K_{IS} - h_{II_0} K_{II} = 1450 \times 10^{-16}$$

(29)

$$m_{II_0} = 23.8 \times 10^{-16}$$

$$m_{II_0} M_{II} + 2m_{IS_0} - m_{II_0} = -423 \times 10^{-16}$$

$$2m_{IS_0} M_{IS} - m_{II_0} M_{II} = 1340 \times 10^{-16}$$

giving the equations

$$h_1 = (1-f)(27 + 620f - 6550f^2) \times 10^{-6} \quad (30)$$

$$h_2 = (1-f)(-23 + 410f - 1300f^2) \times 10^{-6} \quad (31)$$

Thus, at least, one can say that (27) and (28) have a form capable of explaining the measured results, even though this in itself is not very convincing. If the expressions are correct, the values (29) tend to indicate that the iron-silicon interactions are large compared with the iron-iron.

It would be of interest to measure the magnetostriction of some of these alloys under large initial strains, near the elastic limit, for in this way one could determine whether the change in lattice constant alone could produce expressions such as (26) with the required magnitude. It would be necessary, also, in future work to know if the elastic constants for these alloys vary significantly with composition.

ACKNOWLEDGEMENT

I wish first to express appreciation to Professor R. Smoluchowski for his help and encouragement, while acting as thesis advisor for this work; secondly, to acknowledge the help of Dr. J. K. Stanley, of the Westinghouse Research Laboratories, in preparing the ellipsoidal specimens.

In addition I wish to express appreciation to the Westinghouse Laboratories for a fellowship making this work possible, and to them, and in particular J. E. Goldman, for the use of his electromagnet for some of these measurements.

The contribution of Mr. R. K. McGeary in supplying the single crystal material was mentioned previously.

APPENDIX A

The angle θ , the abscissa of the magnetostriction curves is, strictly, the direction of the applied field. The angle ψ between the external field H and the saturation magnetization I for the case of magnetization in the plane of the major axes of an oblate spheroid is¹

$$\sin \psi = -\frac{1}{IH} \frac{\partial F}{\partial \psi}$$

Where

$$F = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2$$

and the K 's are the anisotropy constants; the α 's the direction cosines of I .

In the (100) plane

$$F = K_0 + K_1 \sin^2 \psi \cos^2 \psi$$

where ψ is the angle between the $[001]$ direction and the magnetization.

Since θ , the angle the applied field makes with $[001]$ direction, equals $\psi - \psi$

$$F = K_0 + K_1 \sin^2(\theta + \psi) \cos^2(\theta + \psi)$$

and
$$\sin \psi = -\frac{K_1}{2IH} \sin 4(\theta + \psi)$$

Taking $K_1 = 4 \times 10^5$ ⁽²⁾, $I = 1700$ and $H = 3300$ gives

$$\sin \psi = -0.036 \sin 4(\theta + \psi)$$

Thus ψ is always less than about two degrees and is zero at values of $\theta = 45$ and 90 degrees where the constants are measured.

1. Becker and Döring, Ferromagnetismus, (Julius Springer, 1939) p.122

(2.) This is approximately the value for a 2% Si alloy. It becomes smaller for higher Si content. L.P. Tarasov, Phy. Rev. 56 (1939) p.1234

A similar calculation for the (110) plane shows

$$\sin \psi = - \frac{\left[K_1 + \frac{K_2}{2} \sin^2(\theta + \psi) \right] \left[2 - 3 \sin^2(\theta + \psi) \right] \sin 2(\theta + \psi)}{2 I H}$$

The constant K_2 is small compared with K_1 and can be neglected.

Since ψ is small

$$\psi \approx - \frac{K_1}{2 I H} (2 - 3 \sin^2 \theta) \sin 2 \theta$$

which is the same order of magnitude as the previously calculated angle.

APPENDIX B

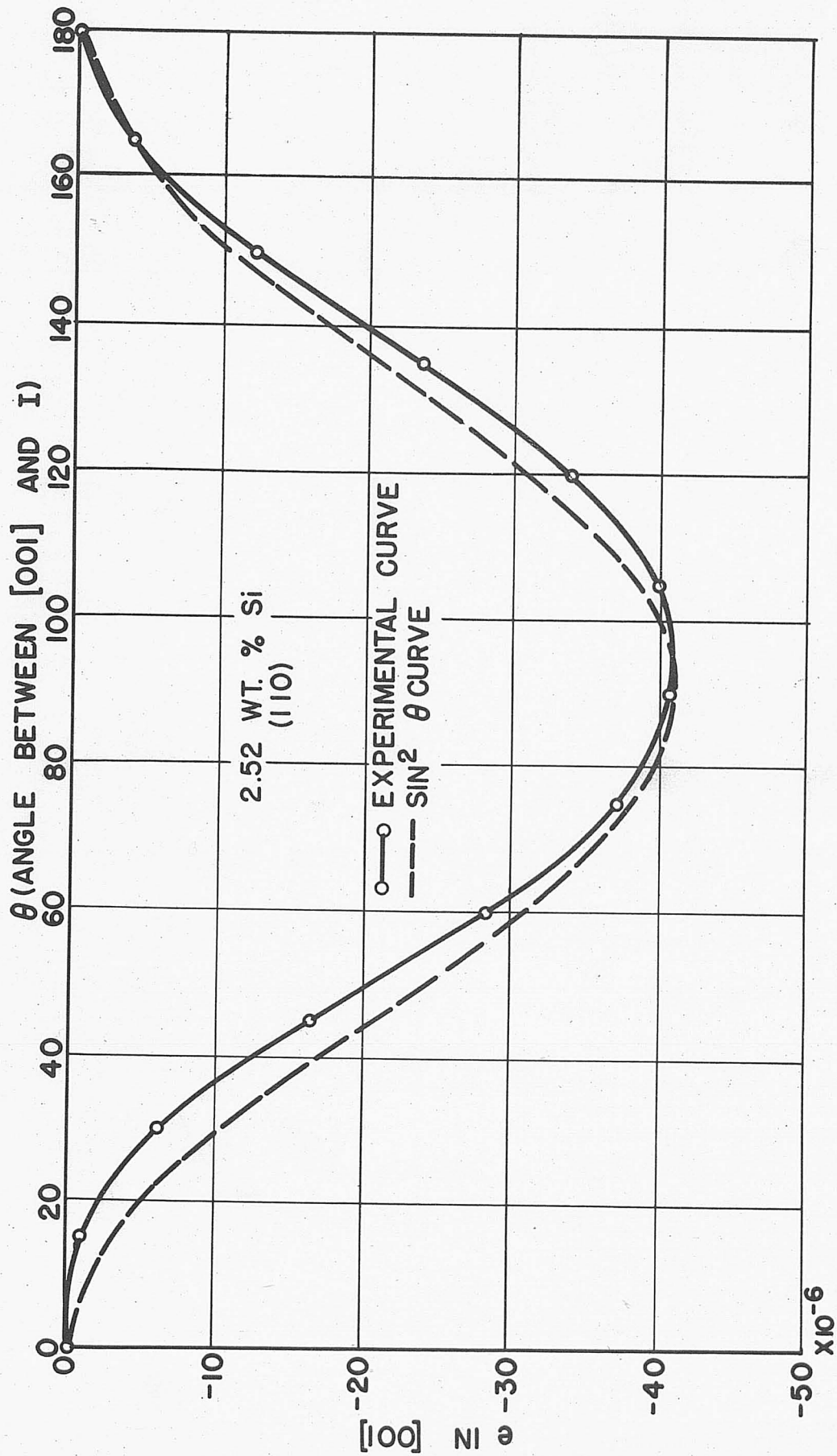


FIG. 8

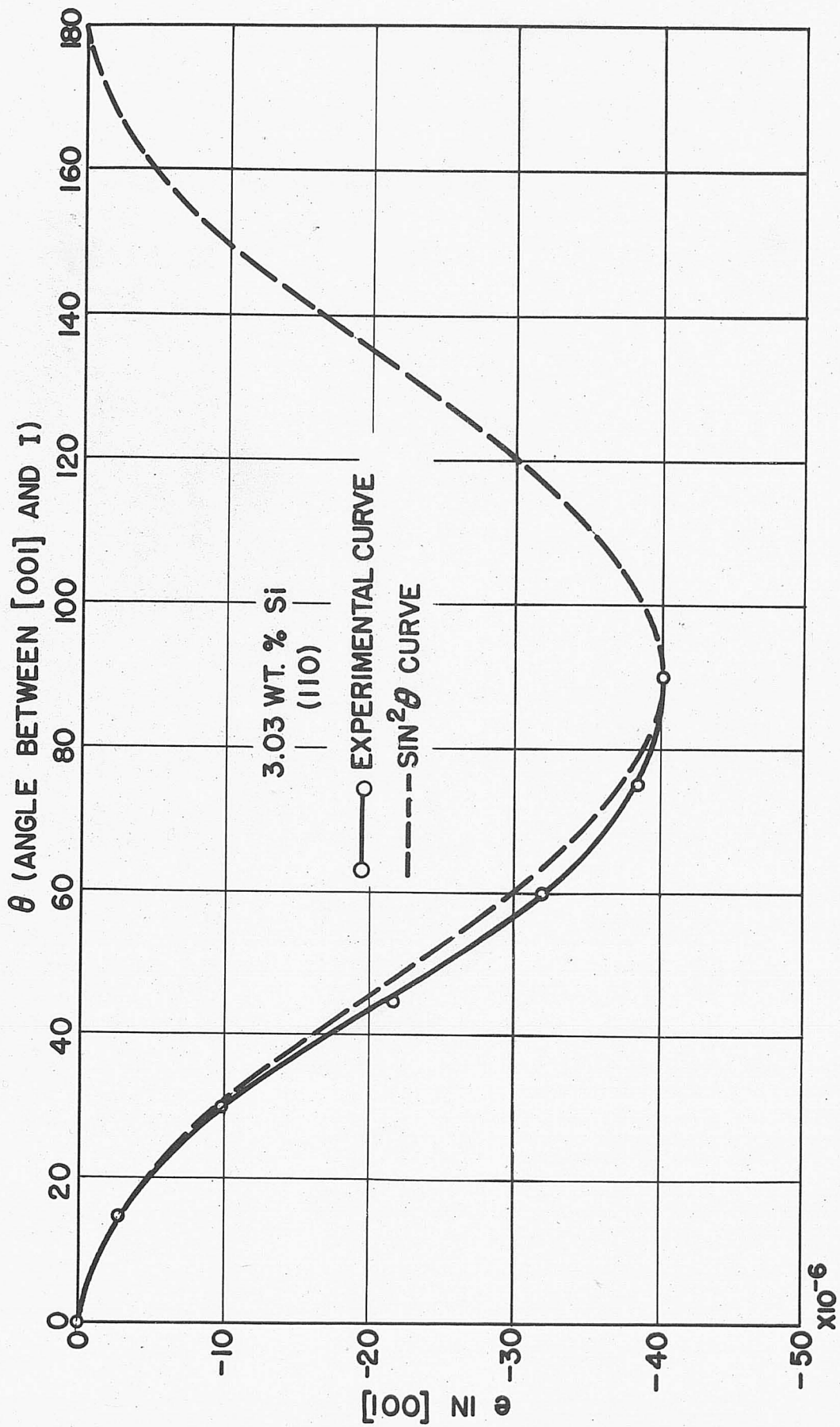


FIG. 9

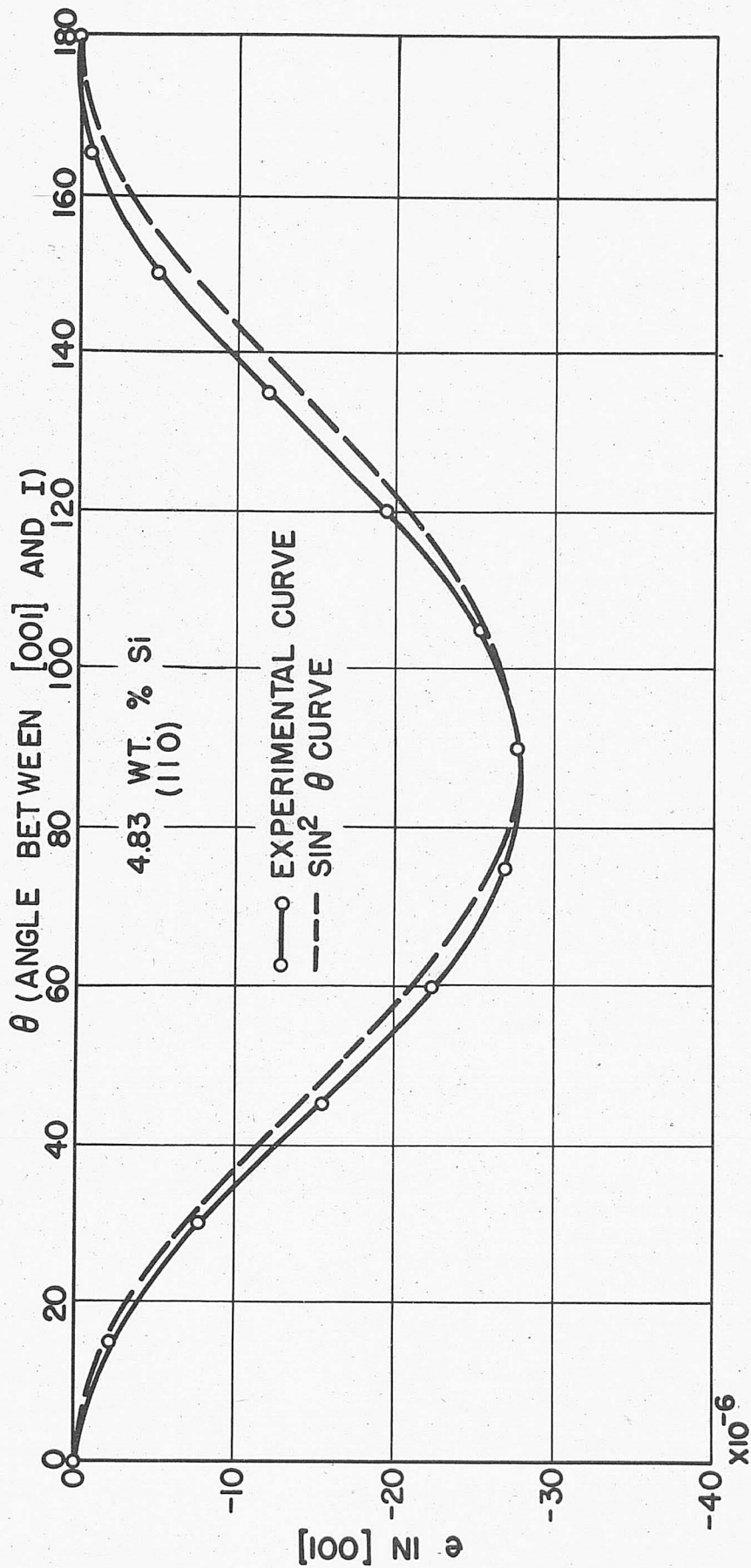


FIG. 10

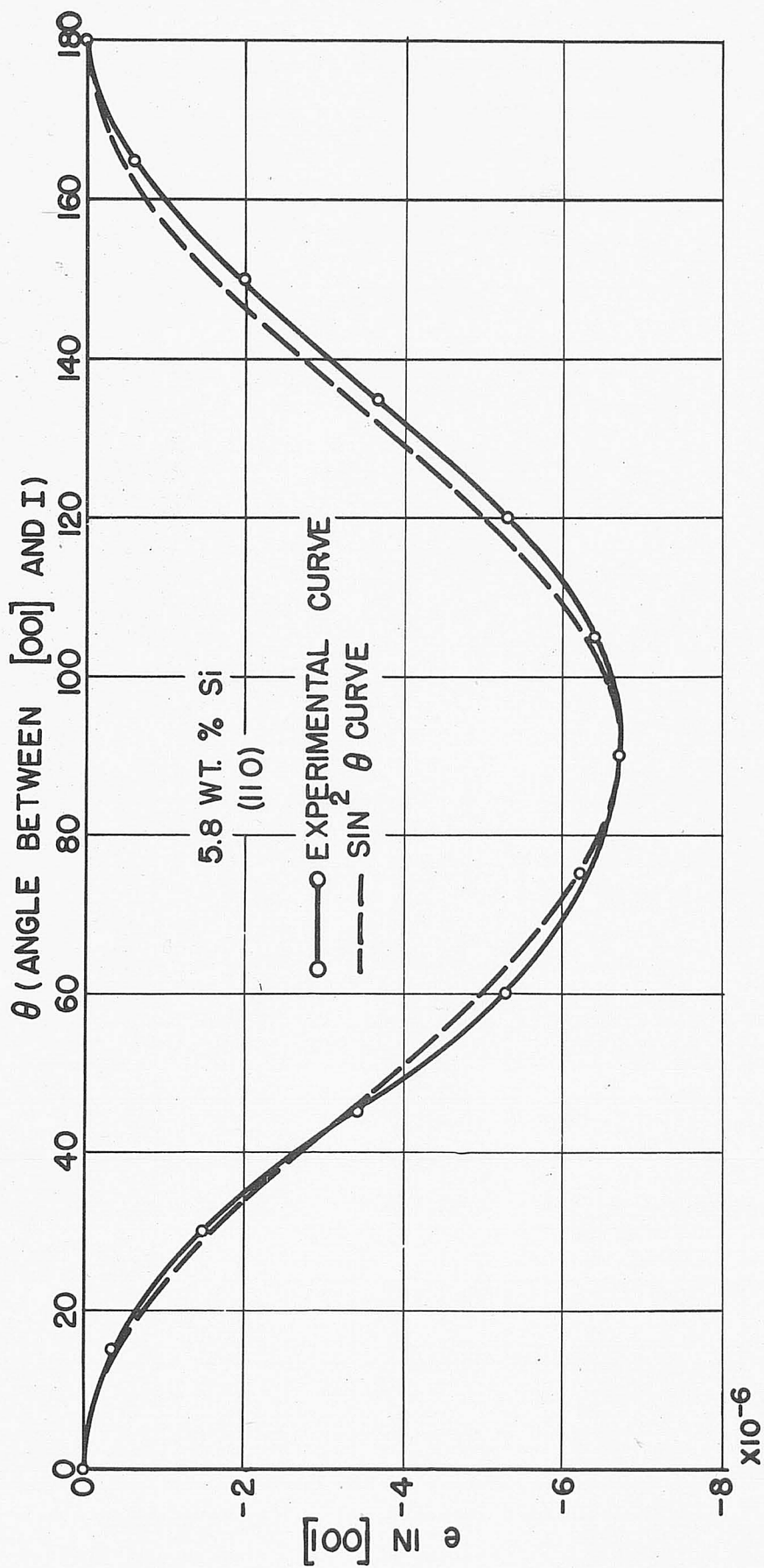


FIG. 11

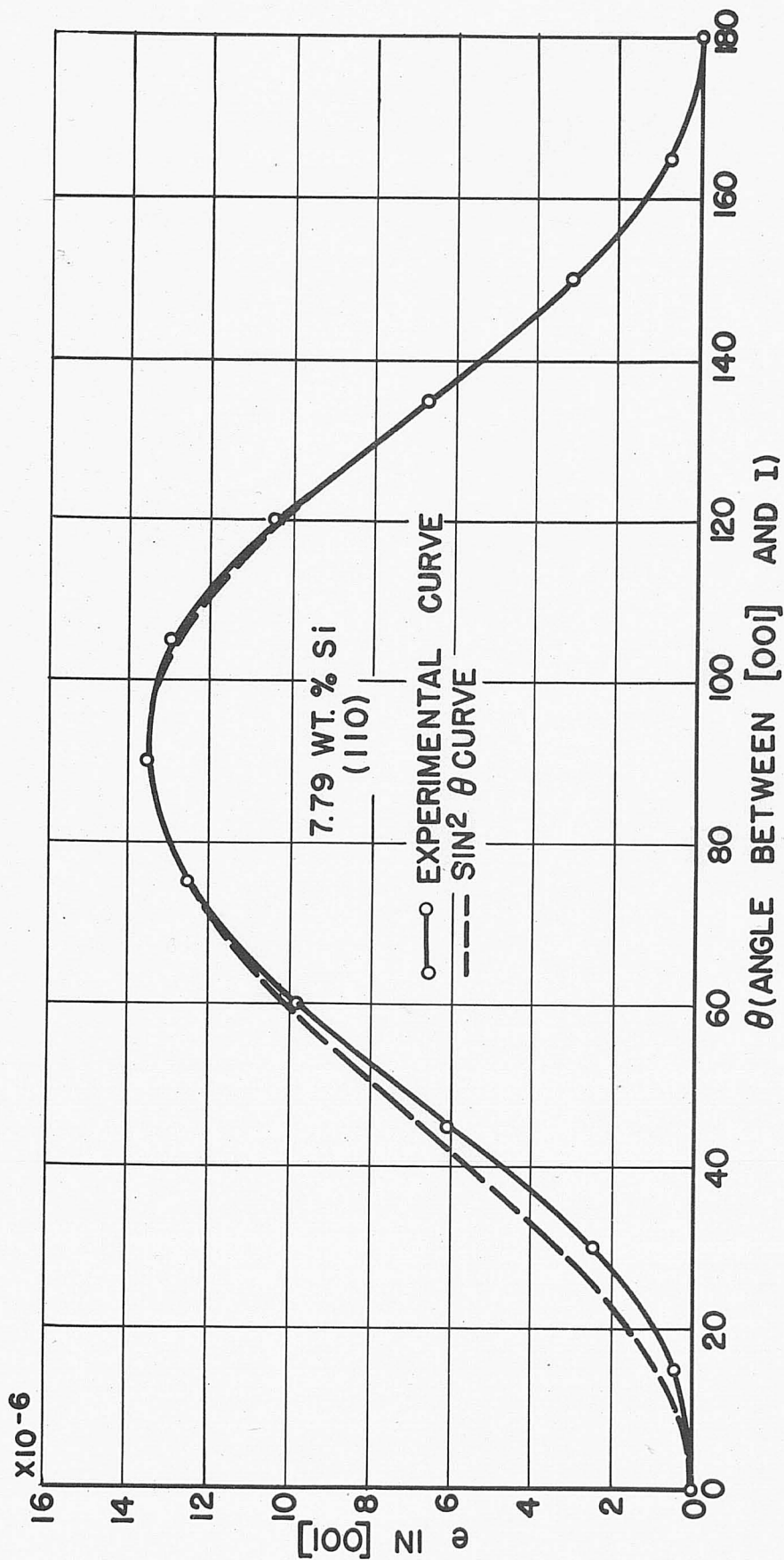


FIG. 12

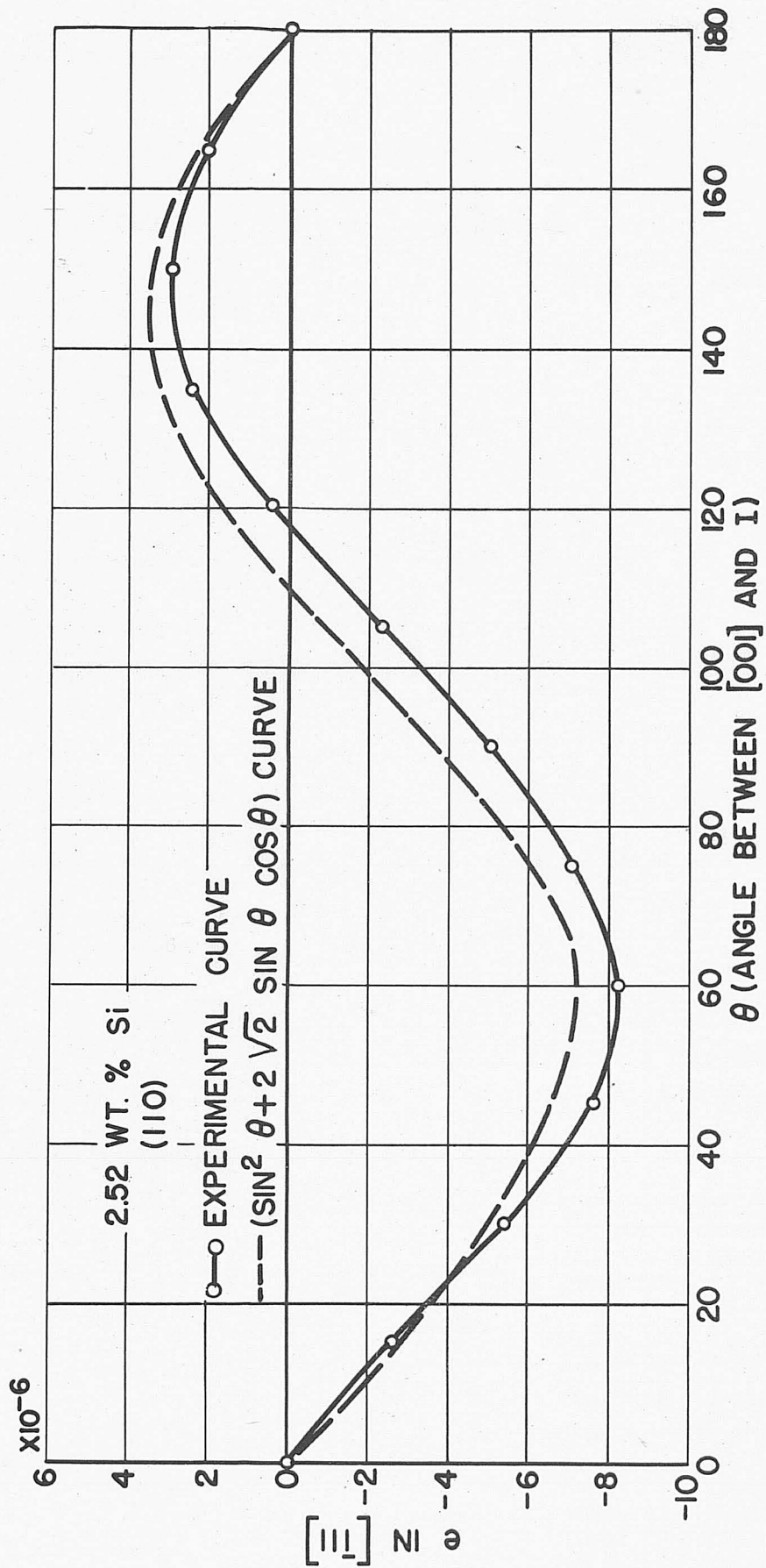


FIG. 13

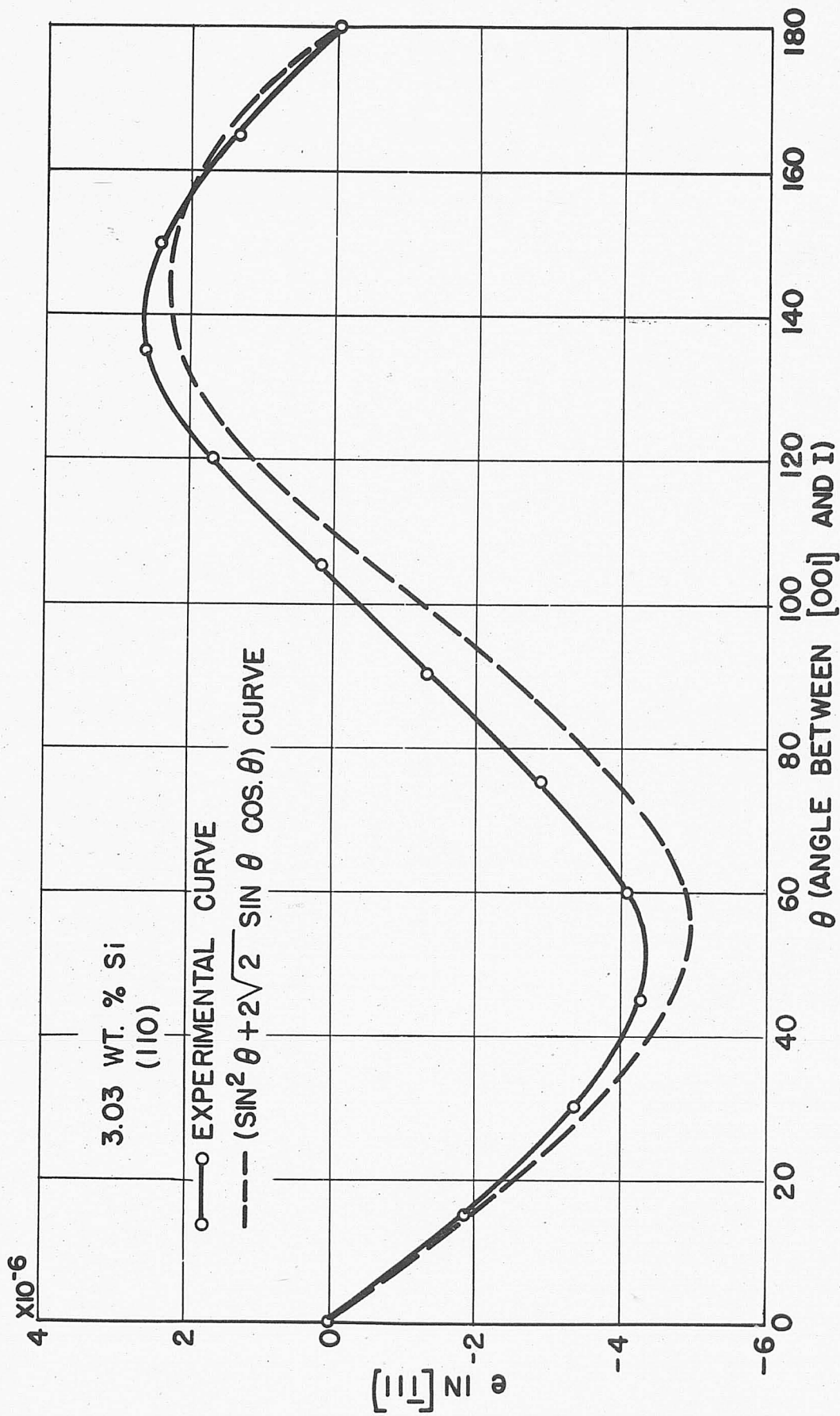


FIG. 14

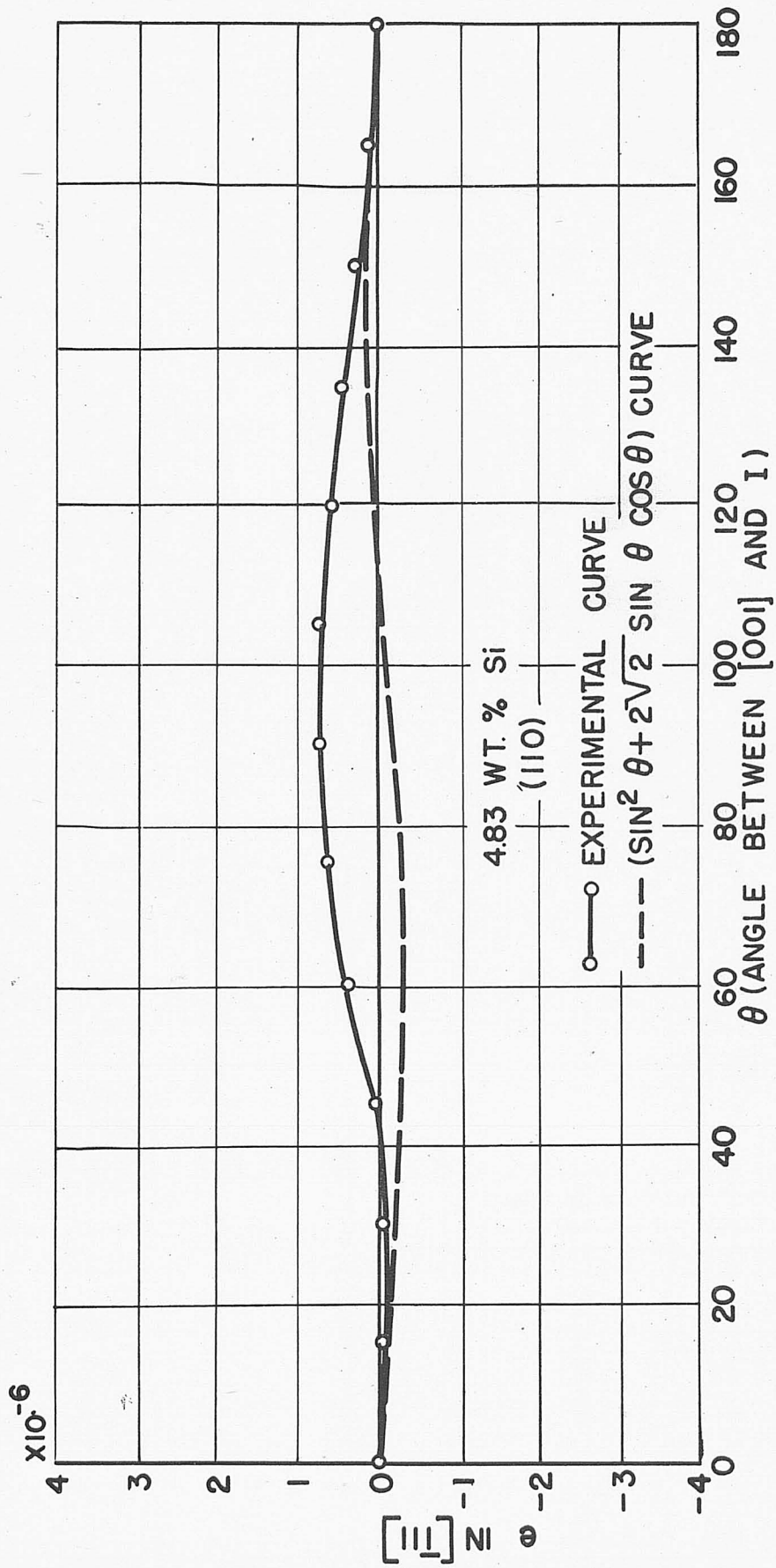


FIG. 15

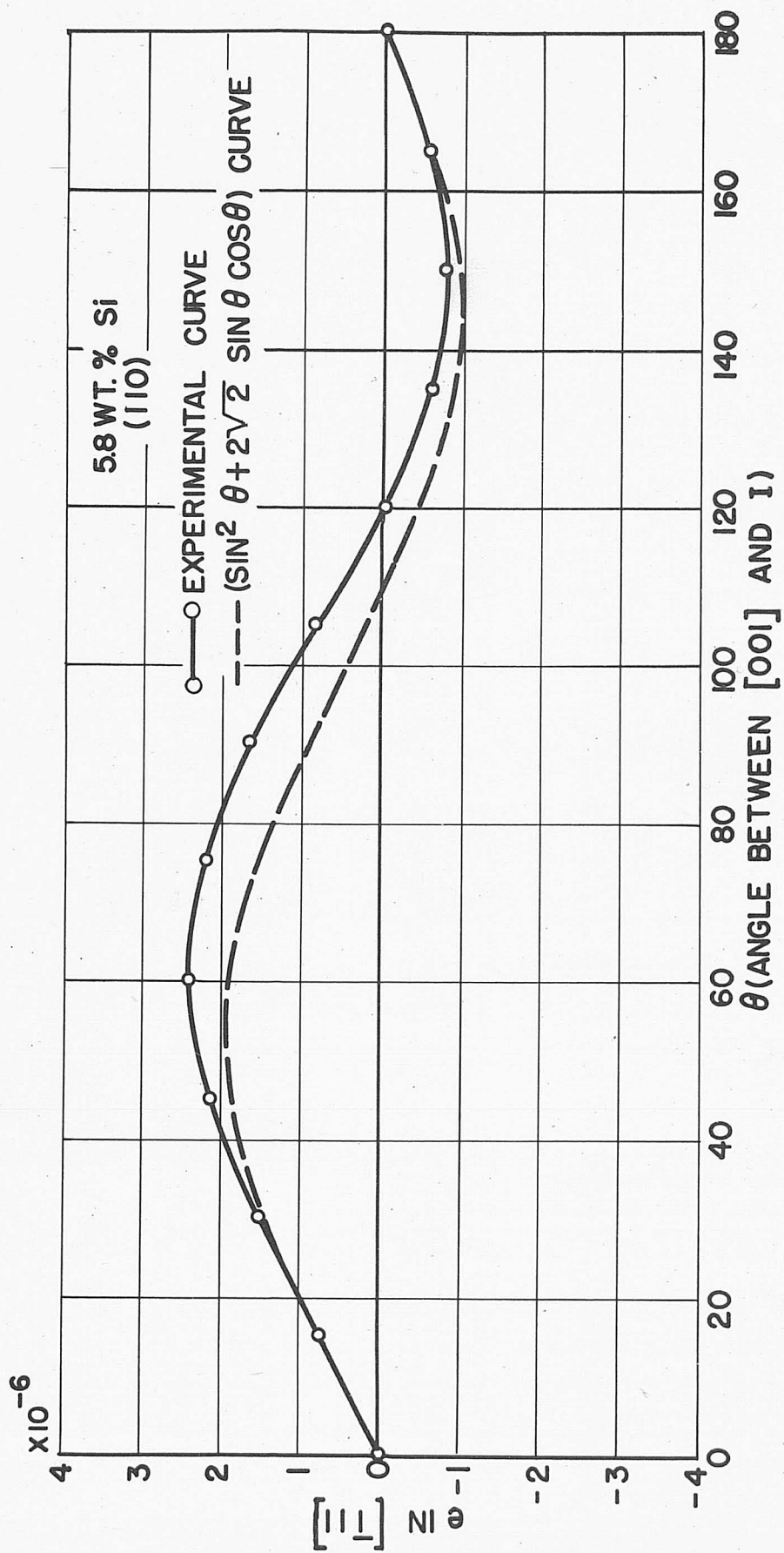


FIG. 16

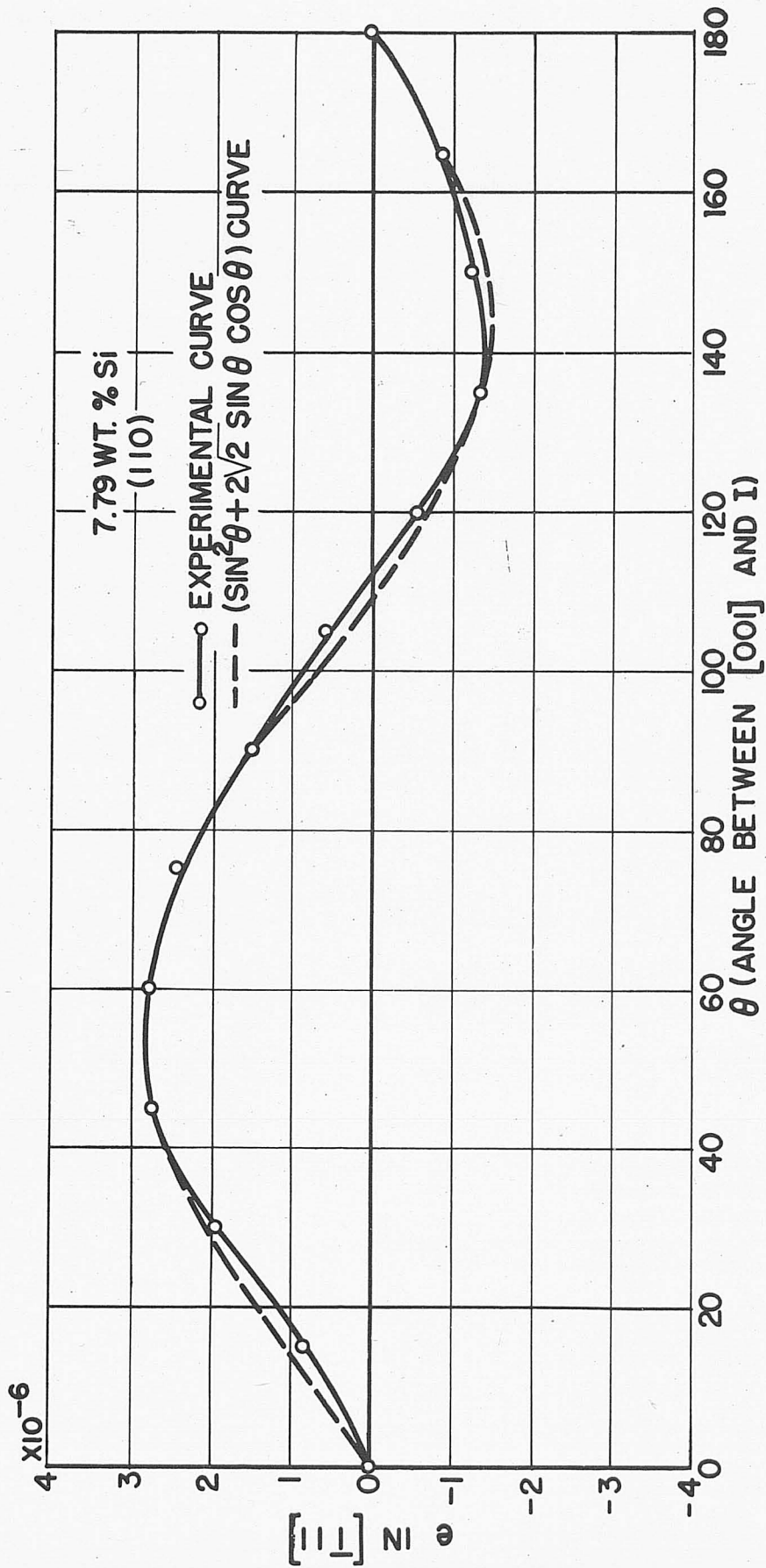


FIG. 17

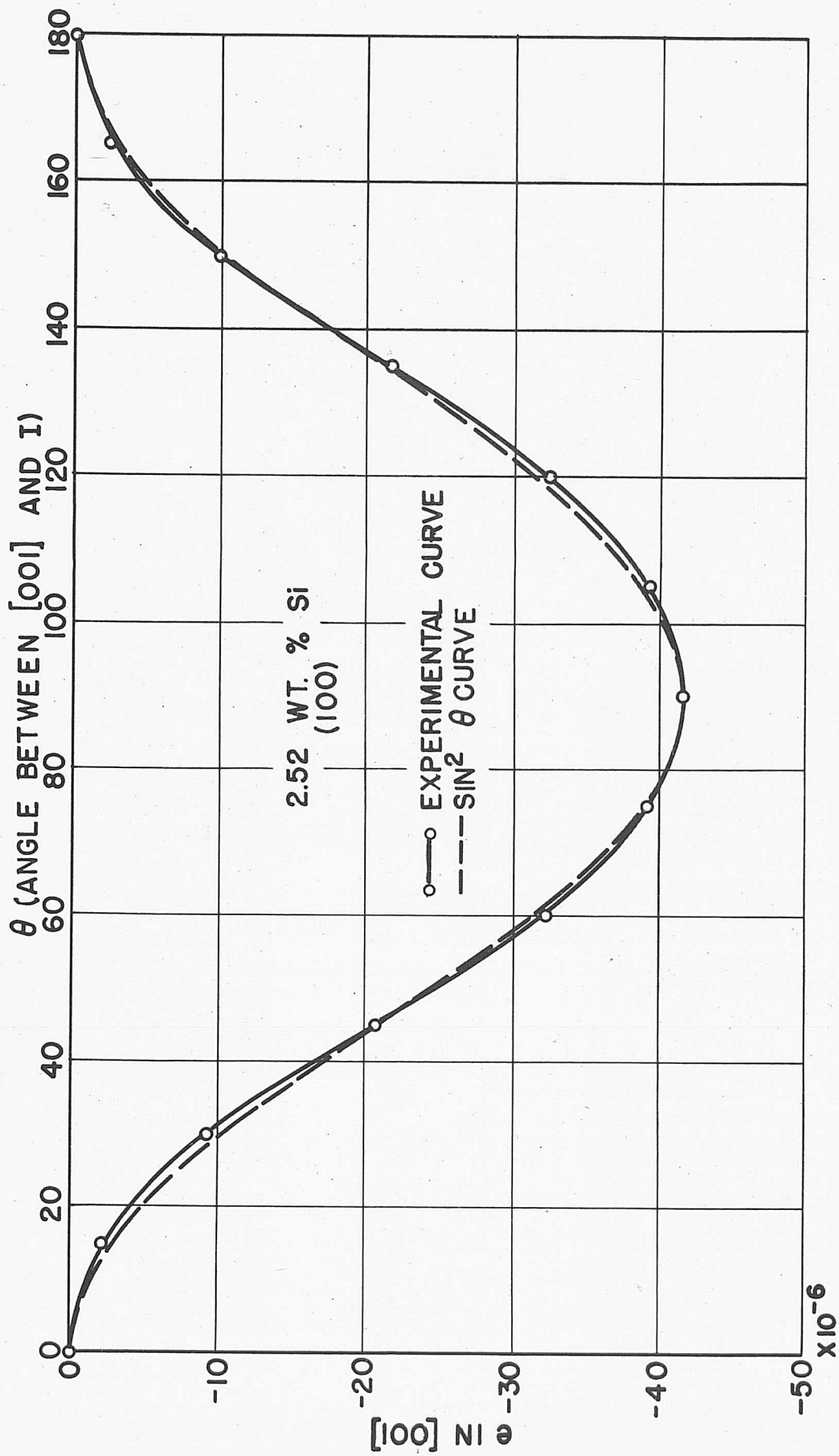


FIG. 18

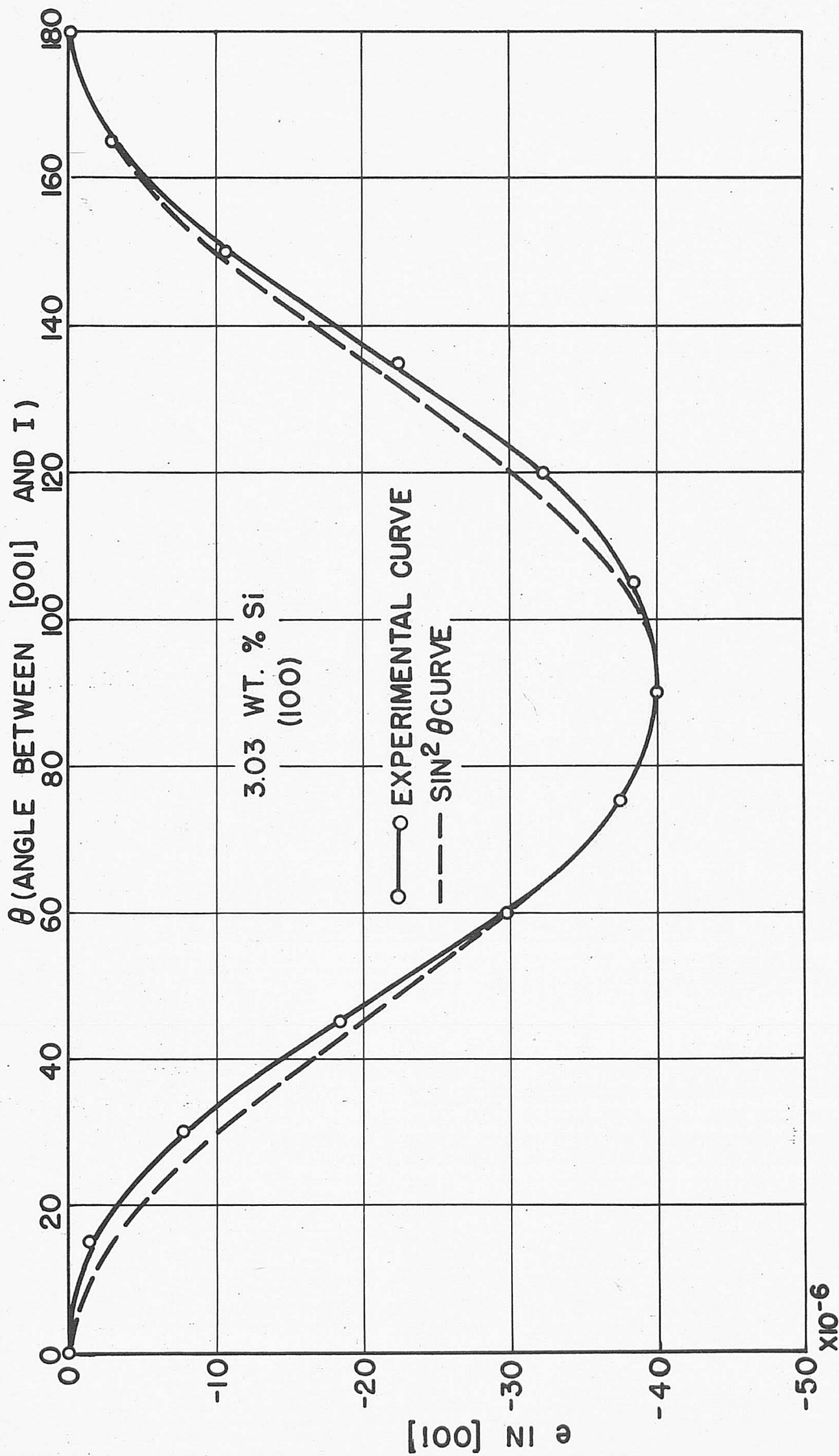


FIG. 19

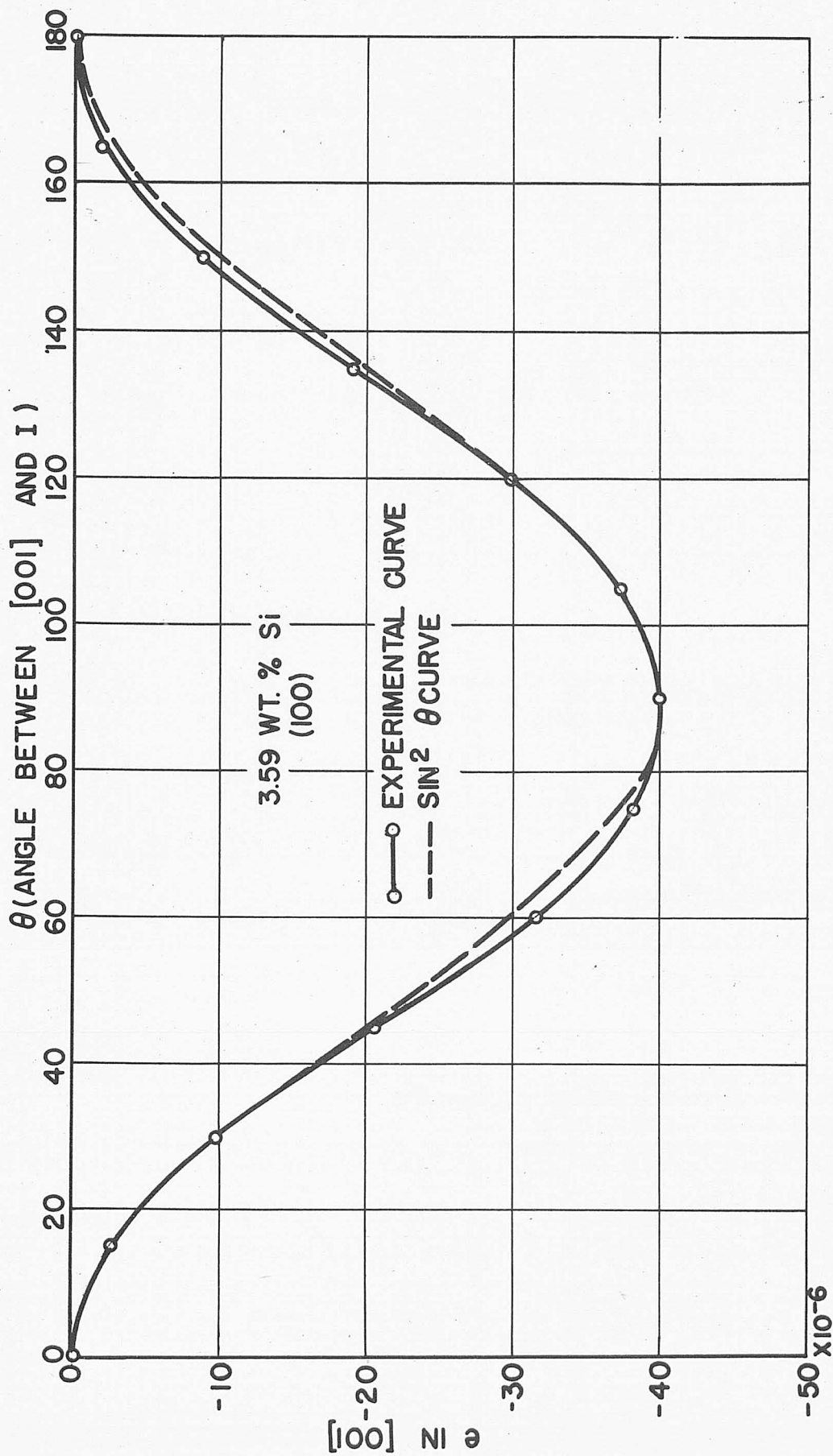


FIG. 20

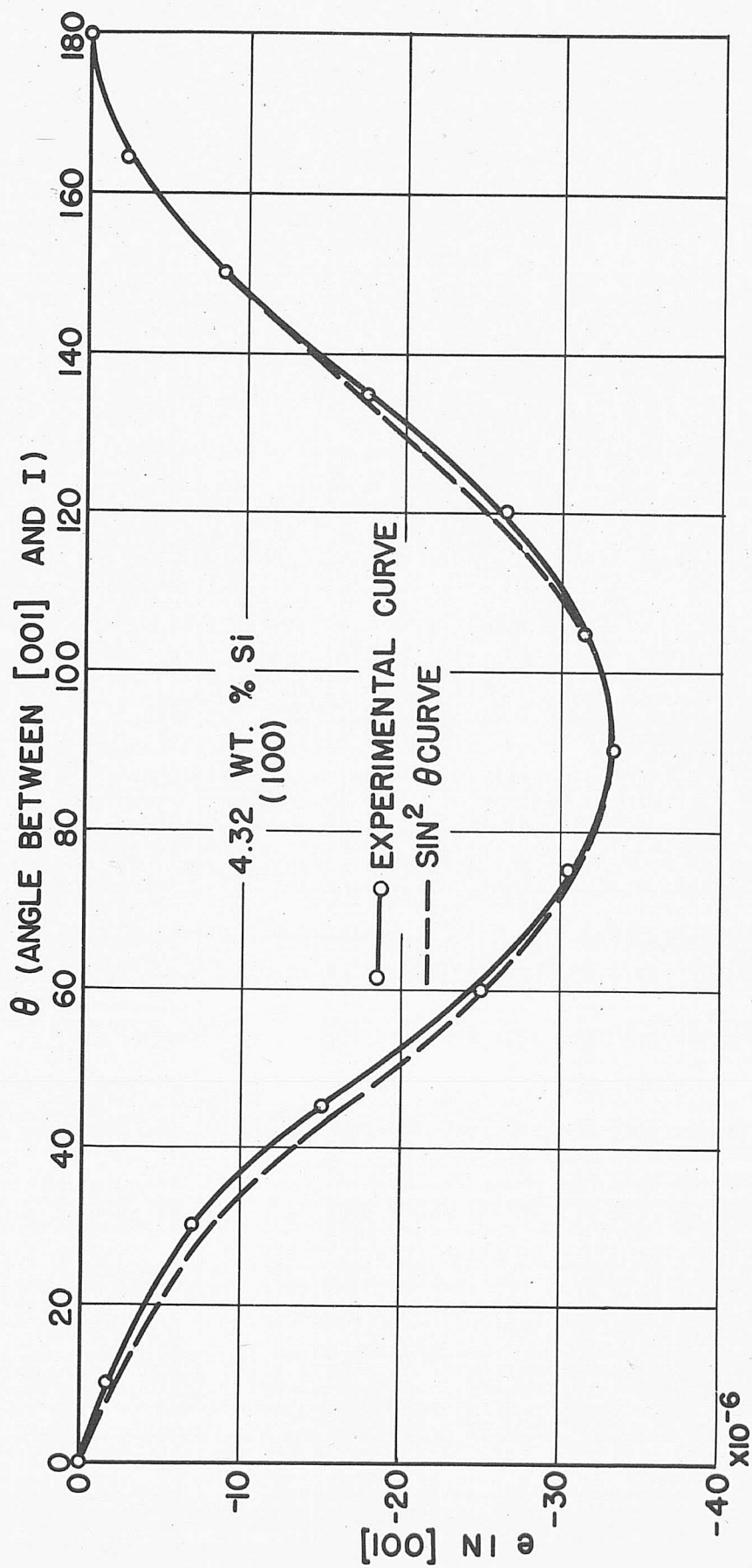


FIG. 21

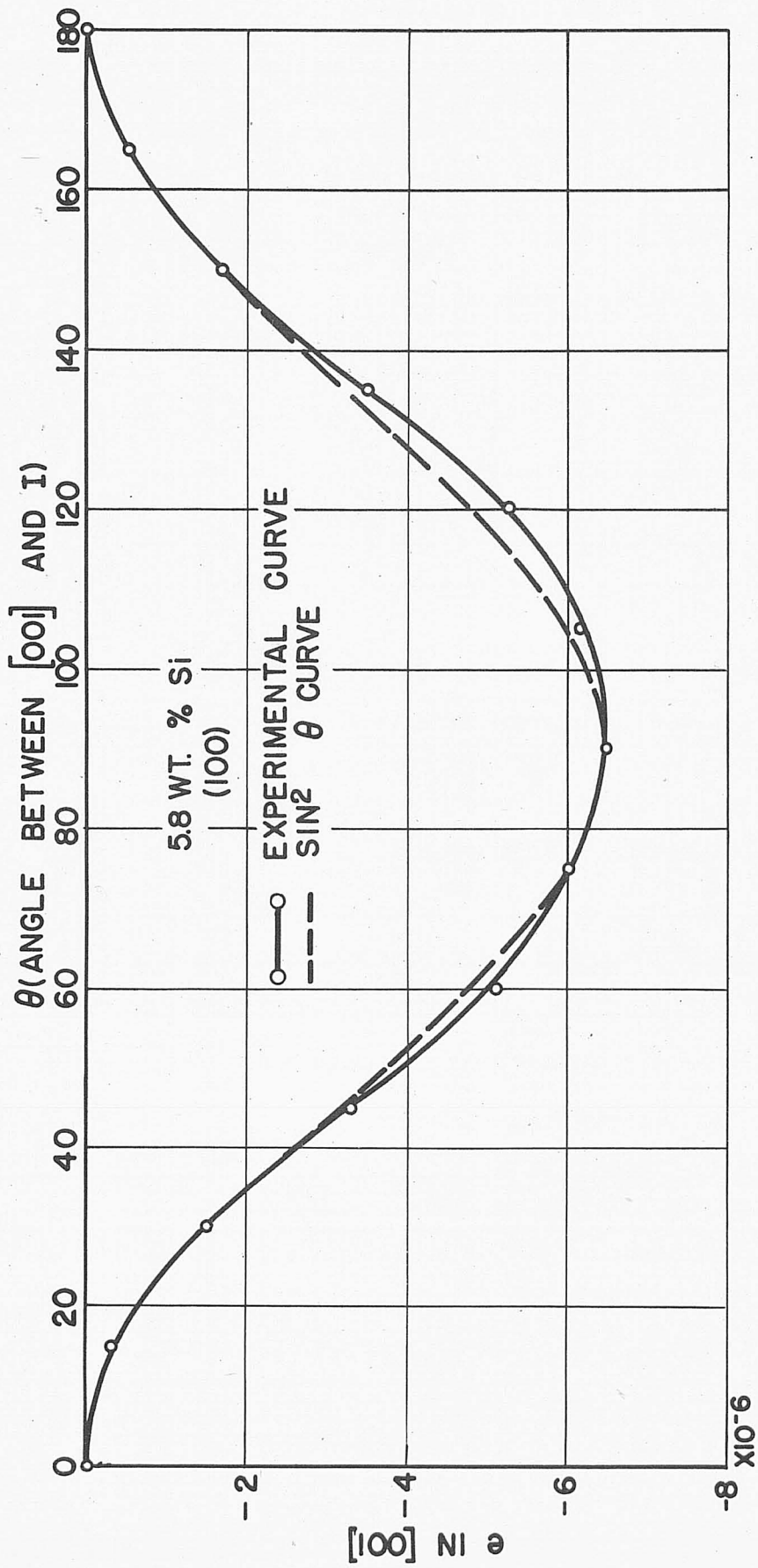


FIG. 22

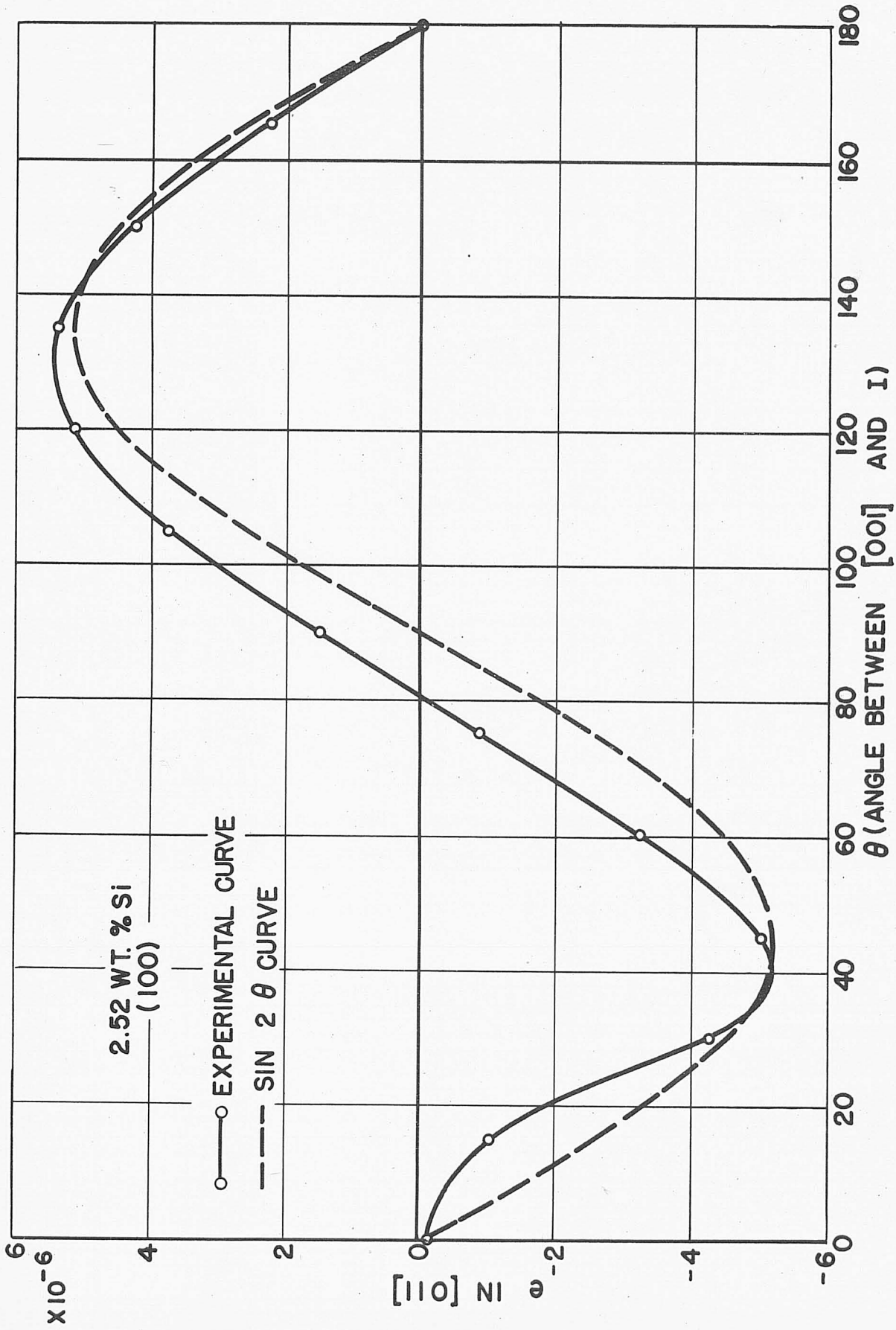


FIG. 23

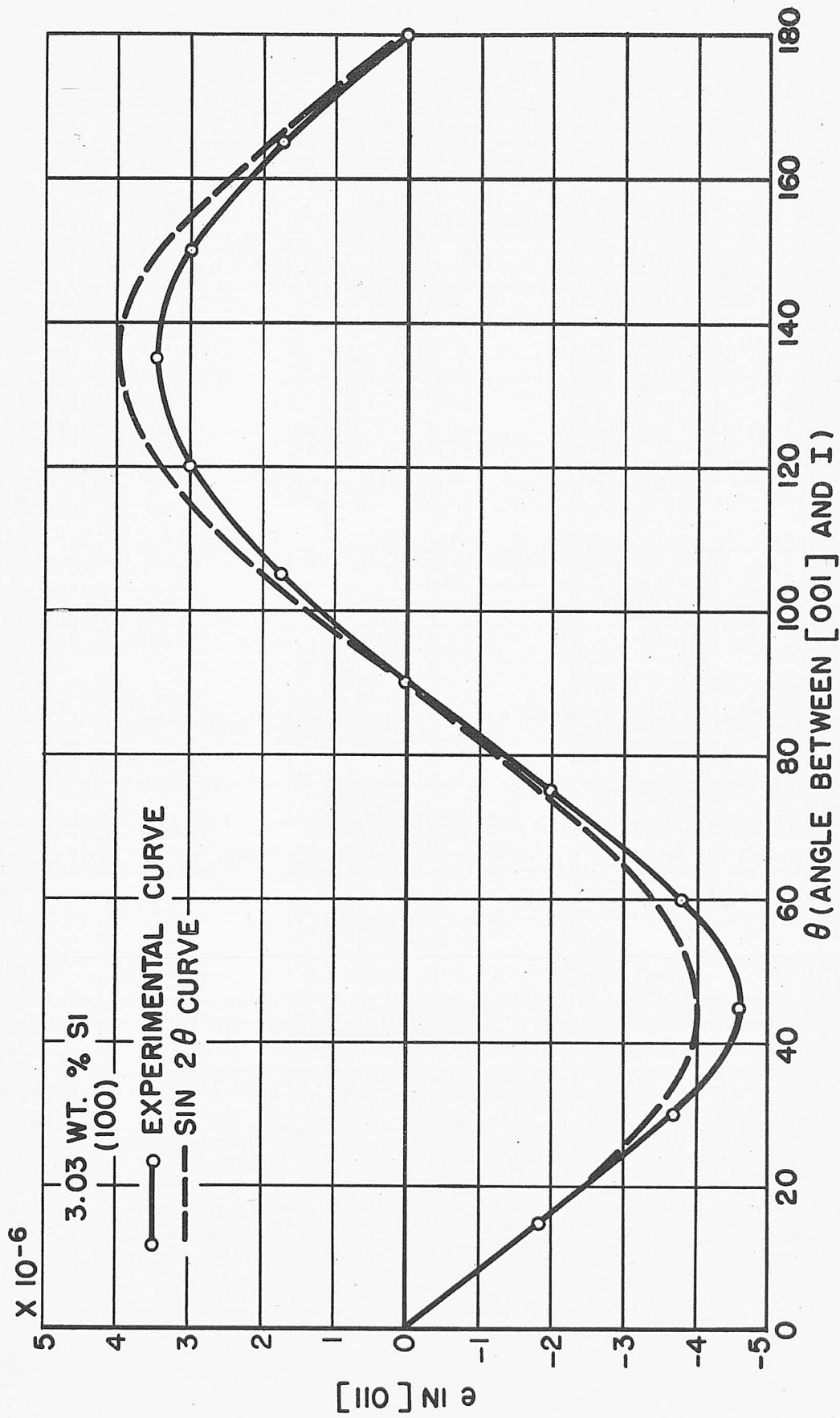


FIG. 24

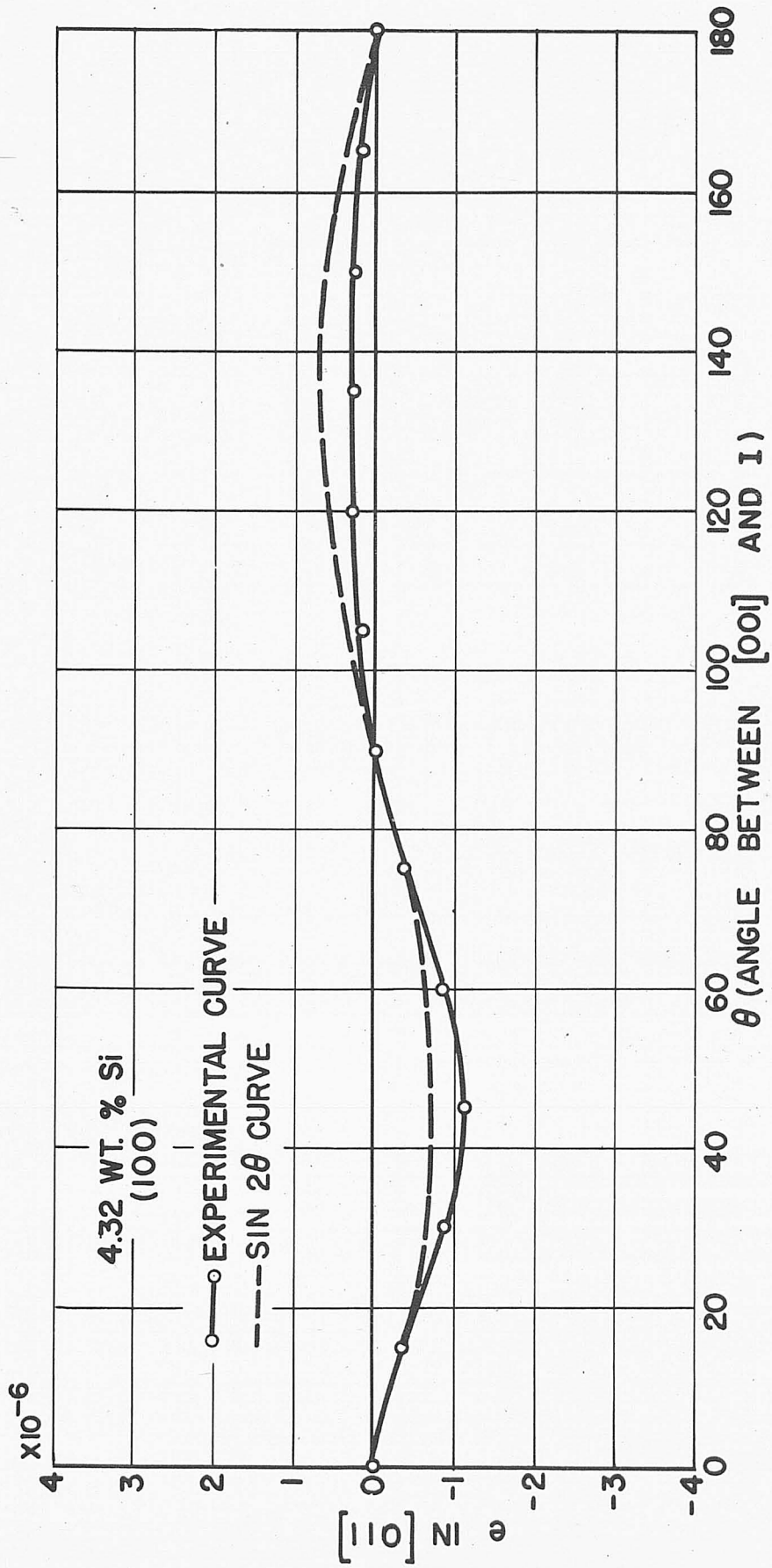


FIG. 25

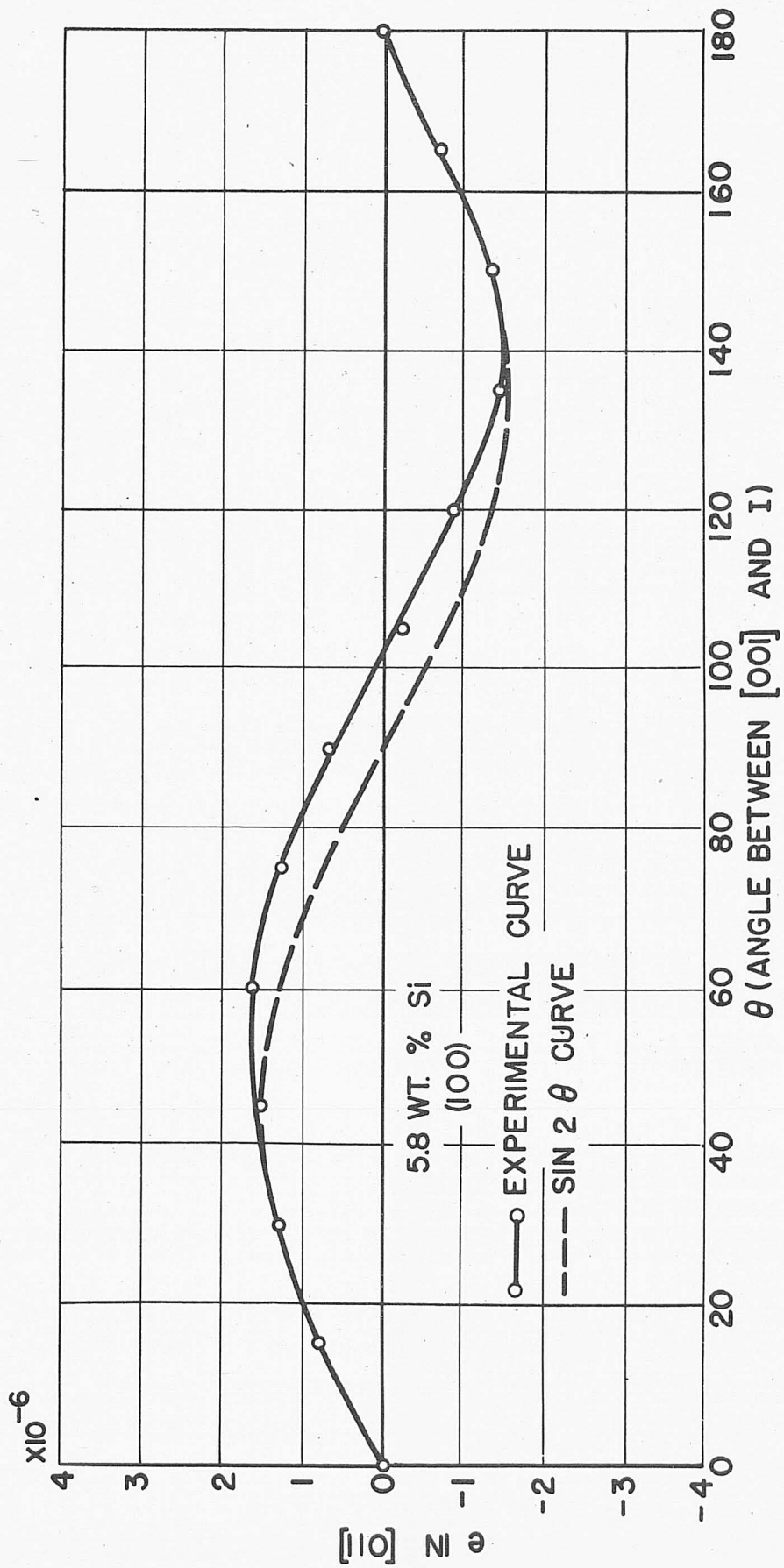


FIG.26

APPENDIX C

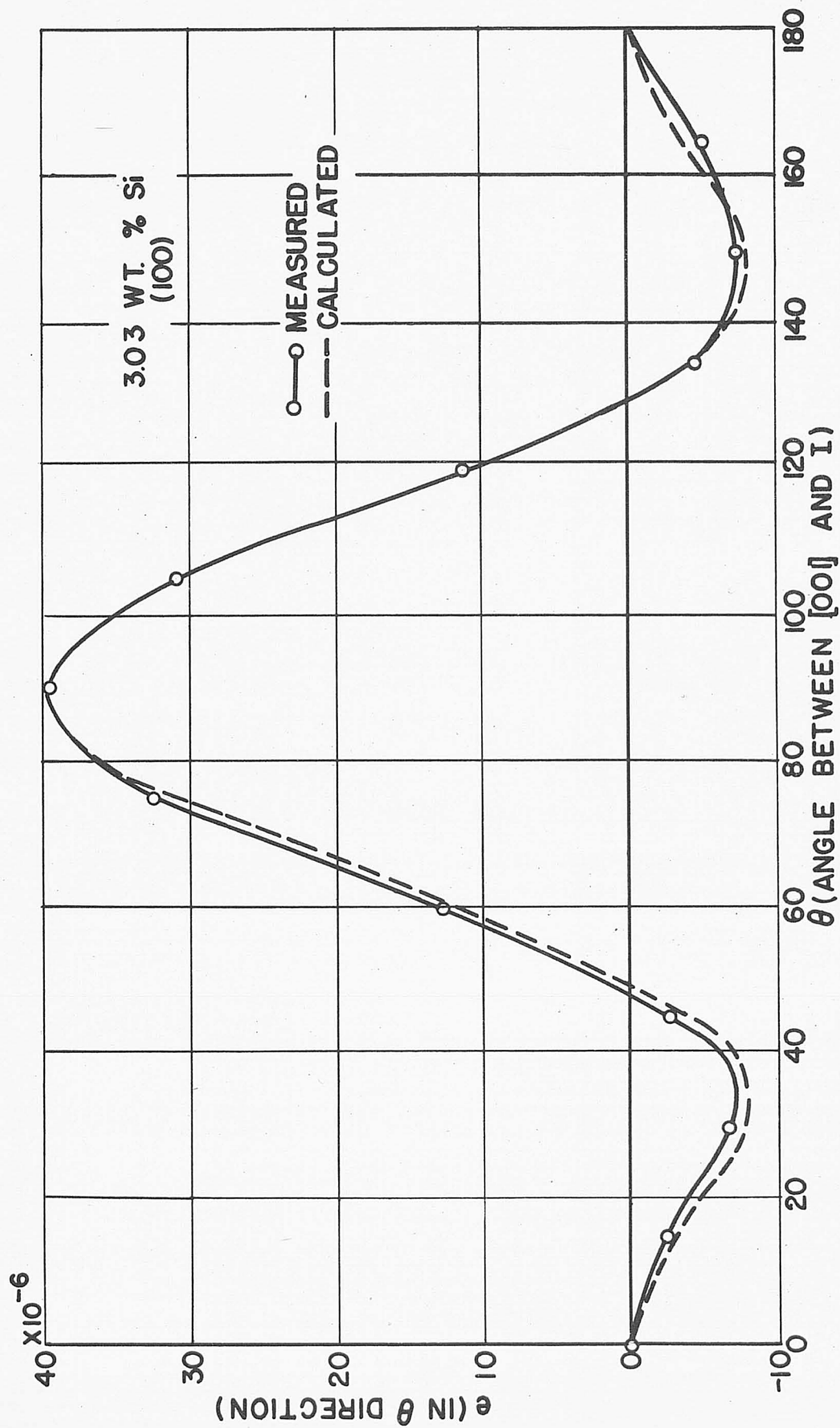


FIG. 27
LONGITUDINAL MAGNETOSTRICTION

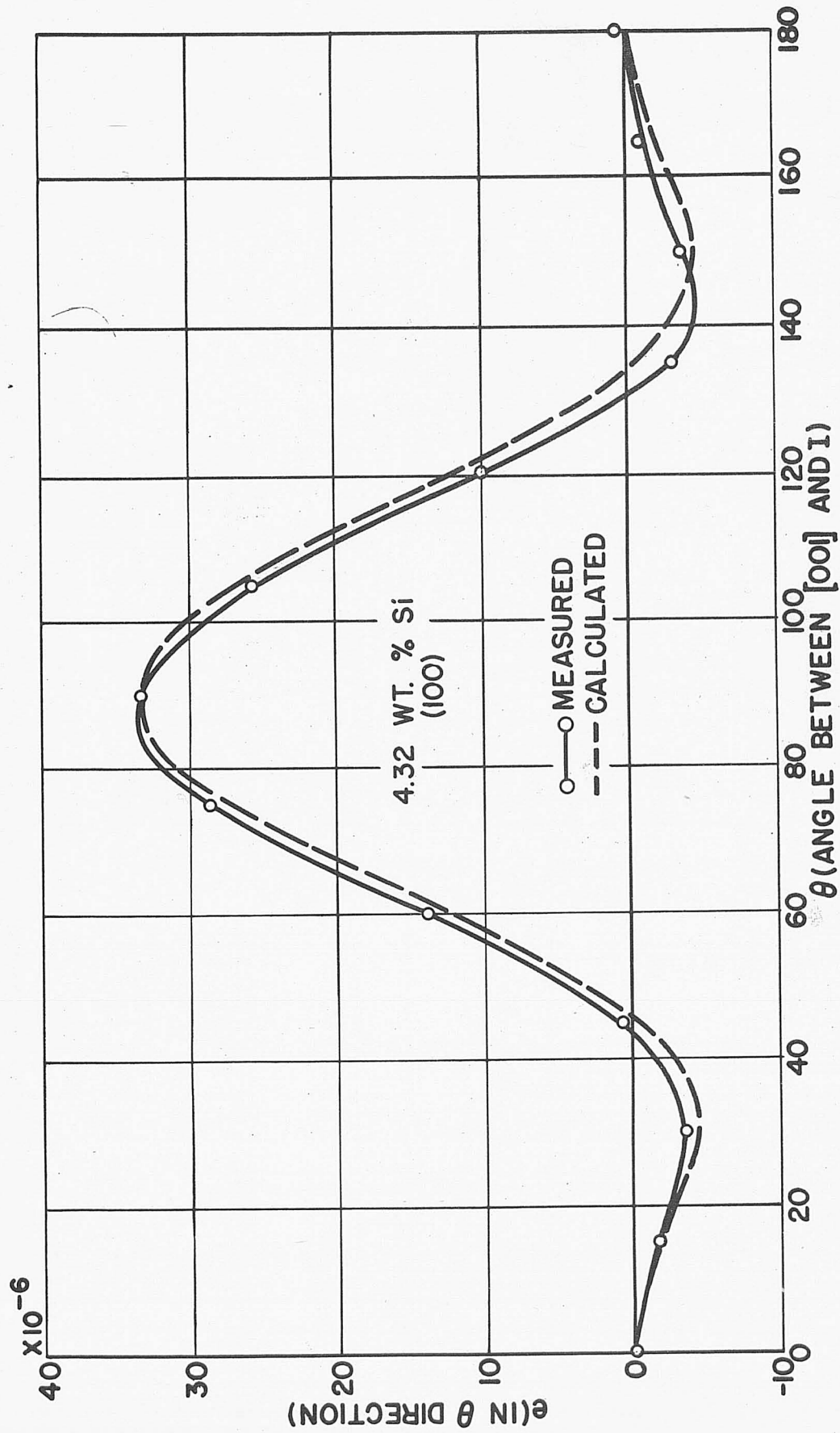


FIG. 28

LONGITUDINAL MAGNETOSTRICTION

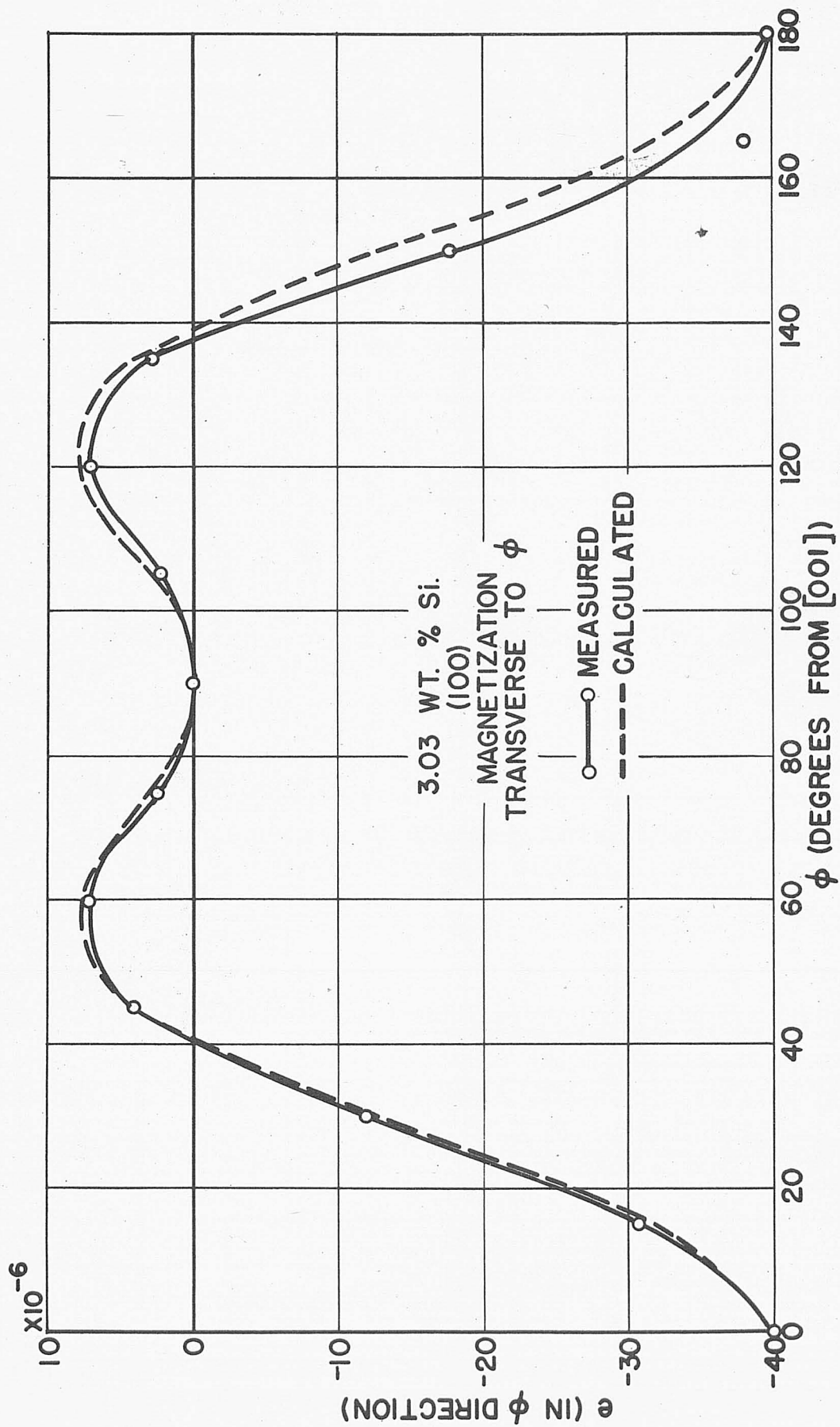


FIG. 29
TRANSVERSE MAGNETOSTRICTION

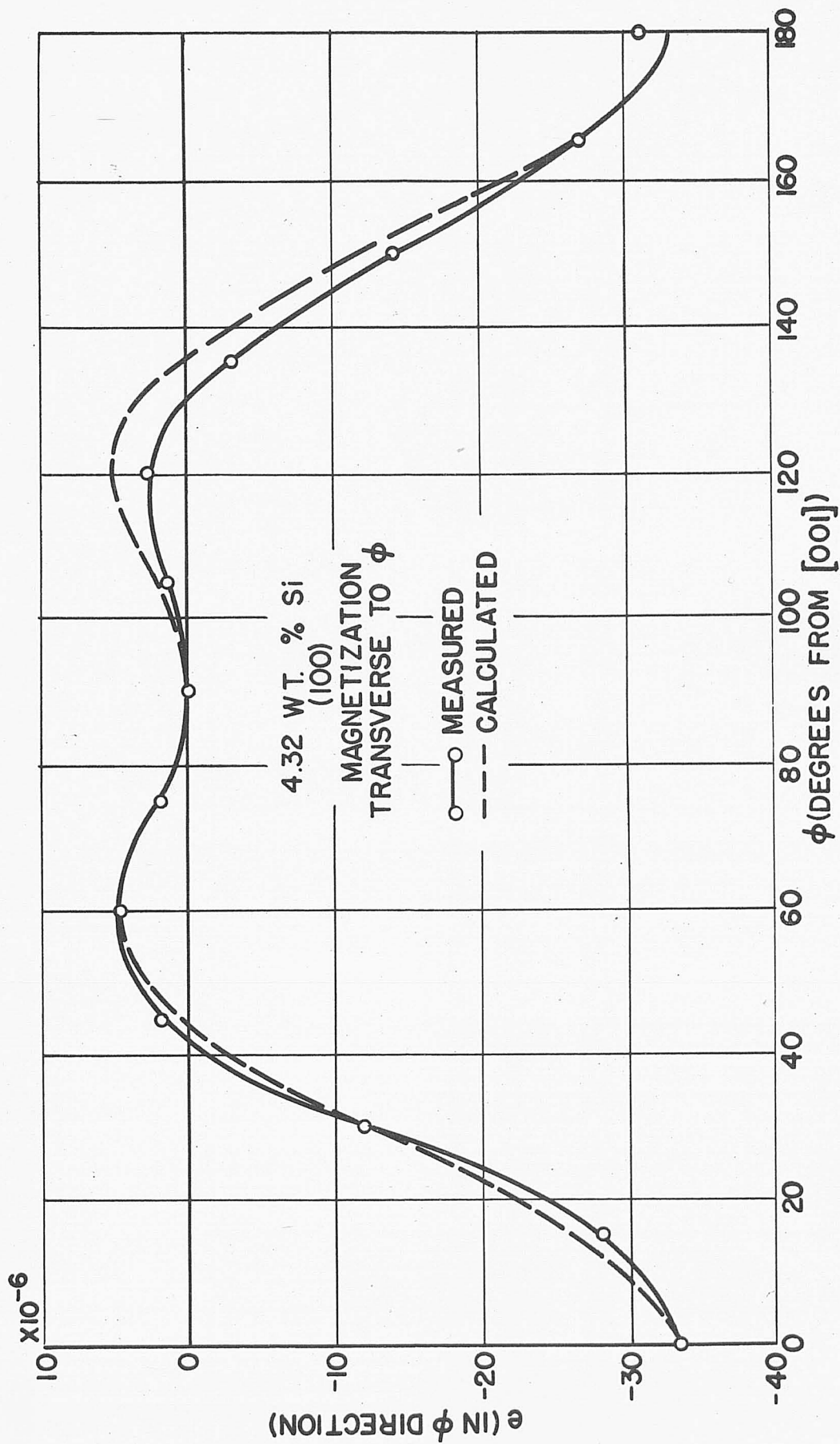


FIG. 30

TRANSVERSE MAGNETOSTRICTION

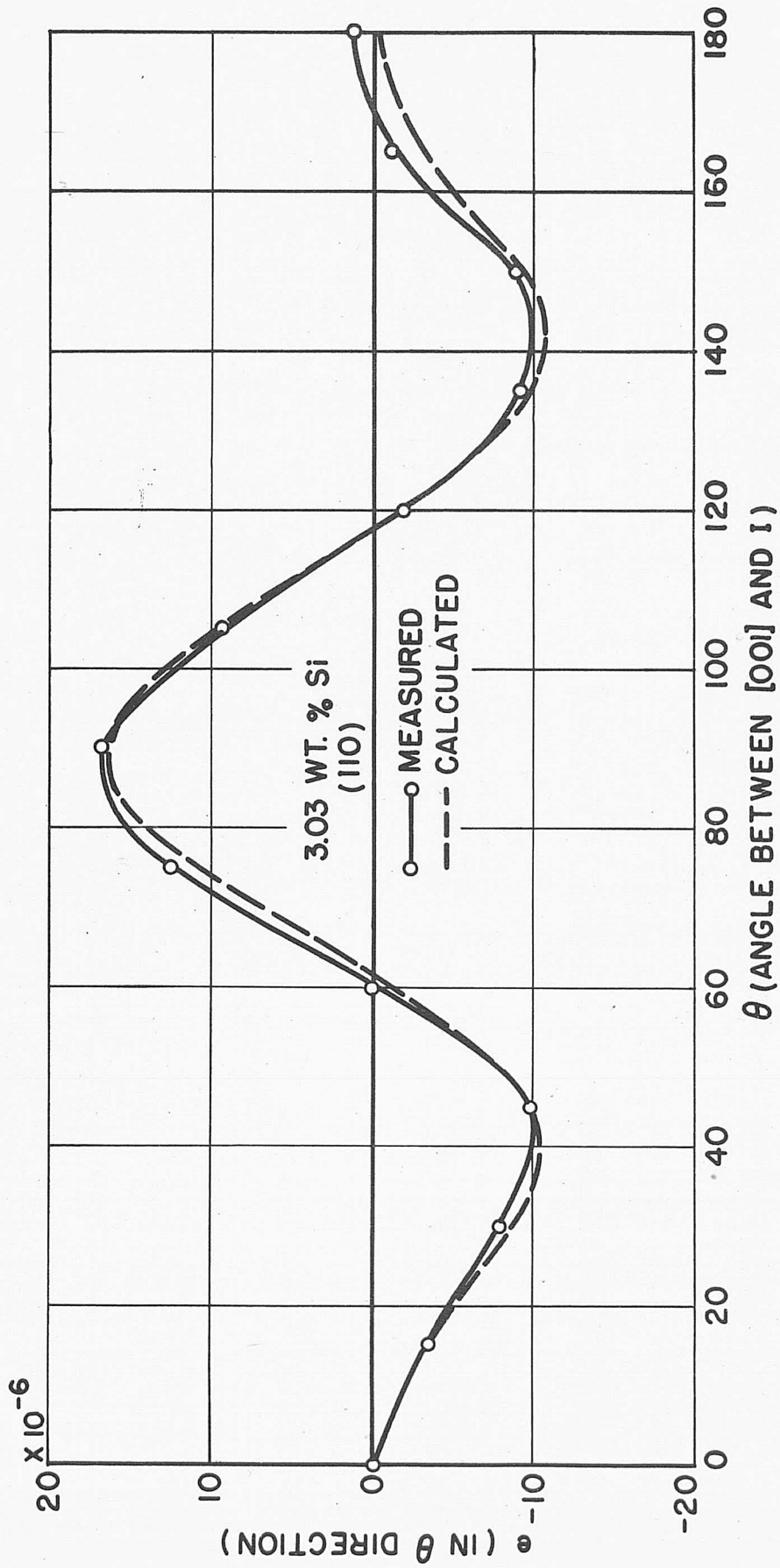


FIG.31
LONGITUDINAL MAGNETOSTRICTION

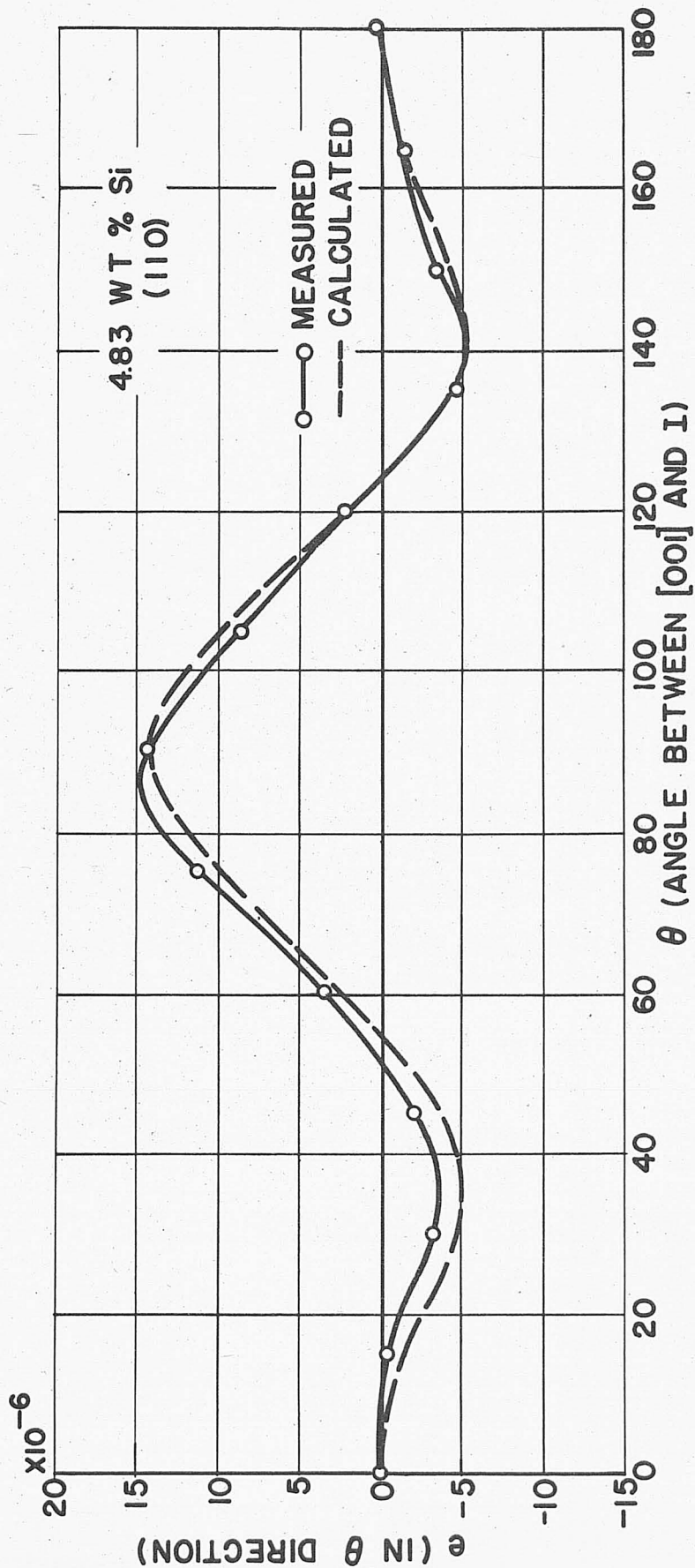


FIG. 32
LONGITUDINAL MAGNETOSTRICTION

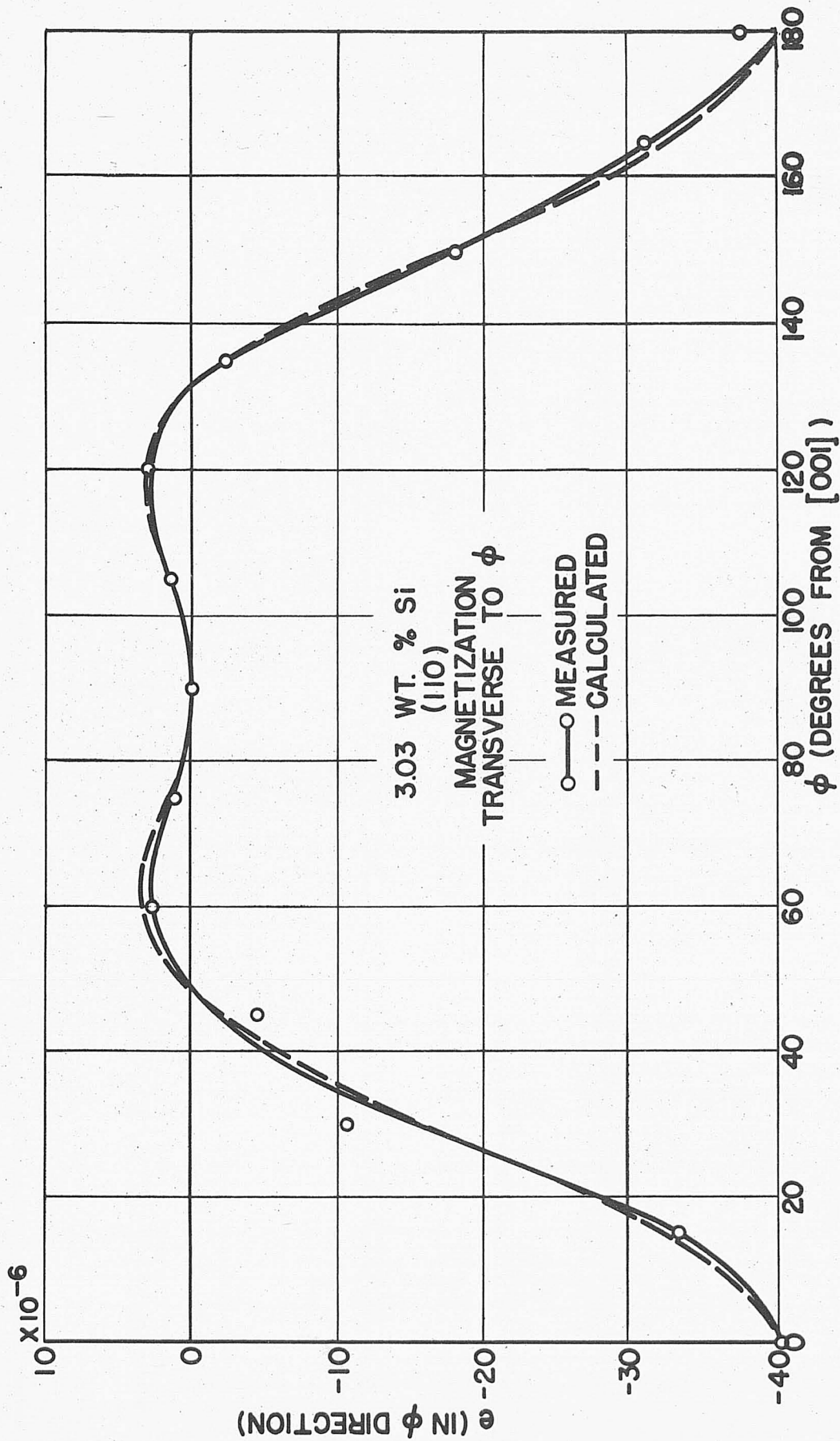


FIG. 33

TRANSVERSE MAGNETOSTRICTION

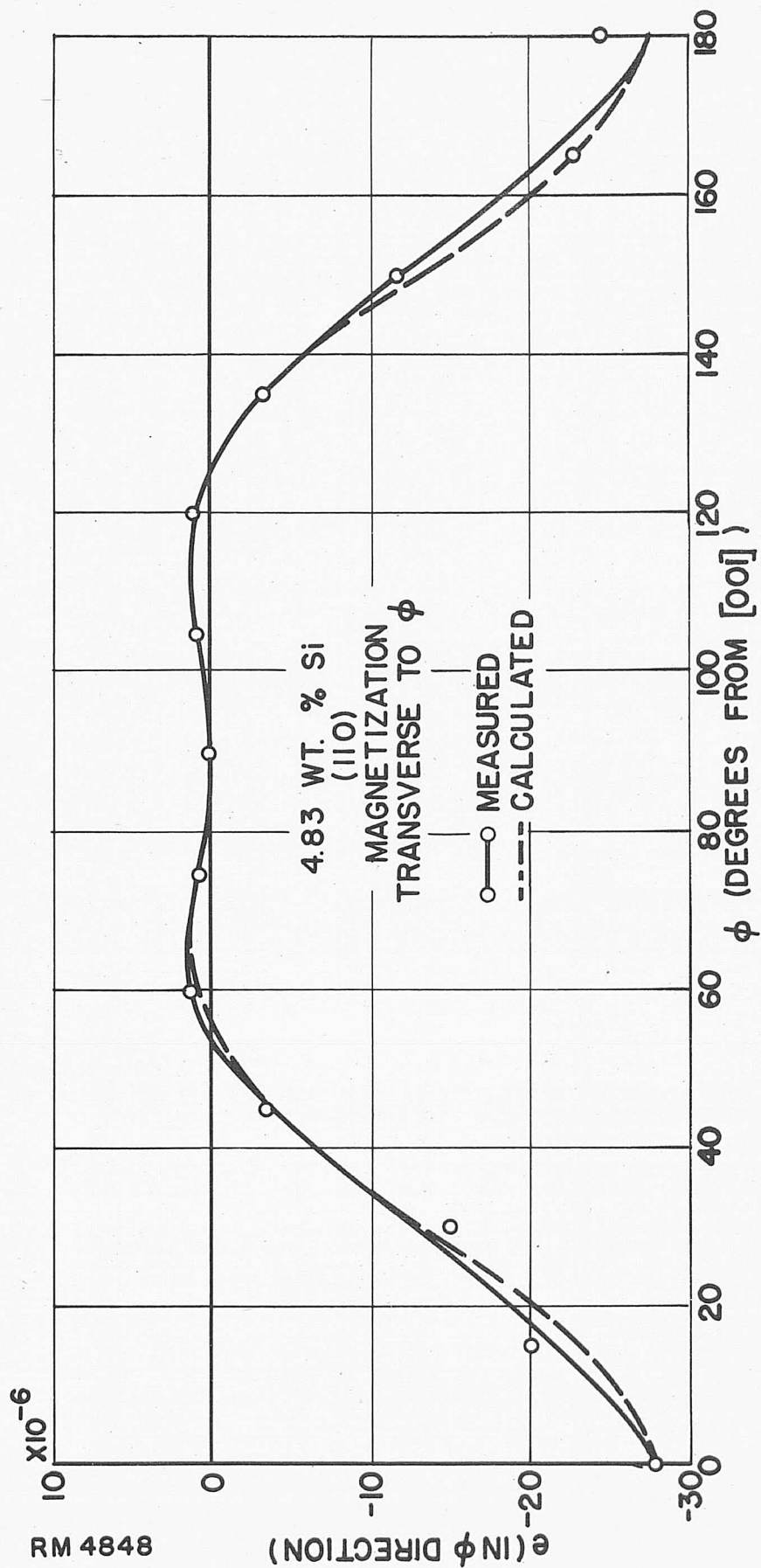


FIG. 34

TRANSVERSE MAGNETOSTRICTION

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