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Determination of Ferromagnetic Anisotropy in Single Crystals and in Polycrystalline Sheets

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Following the work of Akulov and of Heisenberg on the magnetic anisotropy of cubic crystals, it is shown that by taking account of an additional term in the expression for the energy of magnetization the [110] direction may under certain conditions be the direction for easiest magnetization in a crystal, instead of [100] or [111] as given by previous theory. This is in accord with experiment. Magnetization curves for single crystals are calculated using the additional term and some peculiarities are recorded. The anisotropy constant appropriate for a single crystal (of iron) has been calculated from measurements on hard-rolled sheet in which there is preferred orientation of the crystals.

Introduction

It is now well established that single crystals of ferromagnetic materials are magnetically anisotropic even when the crystals have cubic symmetry. In a cubic crystal the energy of magnetization per unit volume in a uniform field H, may be expressed as a function of the direction cosines S_1, S_2 and S_3 of the magnetization I, referred to the crystallographic axes. In ascending powers of the direction cosines, the first three terms of the expression are:[1]

$$E=K_0+K_1(S_1^2S_2^2+S_2^2S_3^2+S_3^2S_1^2)+K_2(S_1^2S_2^2S_3^2). \quad (1)$$

When the magnetization is confined to a crystallographic plane (hkl), as for example in a highly oblate spheroid, the magnetic energy may be expressed as a function of the angle α, between any reference direction $[h_0k_0l_0]$ lying in the plane, and the magnetization. In a uniform magnetic field H, also in the plane (hkl), the crystal will be subject to a torque per unit volume

$$L=-dE/d\alpha. \quad (2)$$

Since α refers to the direction of magnetization, and not the direction of the field, this relation is not immediately useful in determining the constants K_1 and K_2 of Eq. (1). However, as H is increased indefinitely, the direction of I approaches the direction of H and α is then the angle between $[h_0k_0l_0]$ and H. Thus we have

$$L=-K_1(d/d\alpha)(S_1^2S_2^2+S_2^2S_3^2+S_3^2S_1^2)-K_2(d/d\alpha)(S_1^2S_2^2S_3^2) \quad (3)$$

where α is easily determined by experiment. Experimental methods of determining L and so the constant K_1 (and sometimes also K_2) have been given by a number of writers.[2]

It is the purpose of this paper (1) to show how the first anisotropy constant K_1 may be determined from the torque curves of hard-rolled polycrystalline sheet material; (2) to point out that when K_1 is small compared with K_2 and of opposite sign, the direction of easiest magnetization (that for which E is a minimum) may be [110] rather than the usual [100] (as in iron) or [111] (as in nickel); and (3) to calculate some magnetization curves where the K_2 term is important and to compare them with experiment.

Torque Curves for Single Crystals

When a disk or oblate spheroid is cut from a single crystal parallel to a crystallographic (hkl) plane, the torque in a high magnetic field which lies in the plane (hkl) and makes the angle α with $[h_0k_0l_0]$ can be calculated in the manner described in the appendix. Expressions for all of the simpler orientations are given specifically, and of these one may be taken for illustration. When $(hkl)=(110)$ and $[h_0k_0l_0]=[001]$,

$$L=-K_1(2\sin 2\alpha+3\sin 4\alpha)/8-K_2(\sin 2\alpha+4\sin 4\alpha-3\sin 6\alpha)/64. \quad (4)$$

When both K_1 and K_2 are positive, this expression is equal to zero for $\alpha=0$ ([001] direction) and for $\alpha=90°$ ([$1\bar{1}0$] direction), both being positions of stable equilibrium, and also for

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[1] N. S. Akulov, Zeits. f. Physik **67**, 794 (1931); R. Gans, Physik. Zeits. **33**, 924 (1932).

[2] E.g., see a paper by H. J. Williams recently submitted to Review of Scientific Instruments.

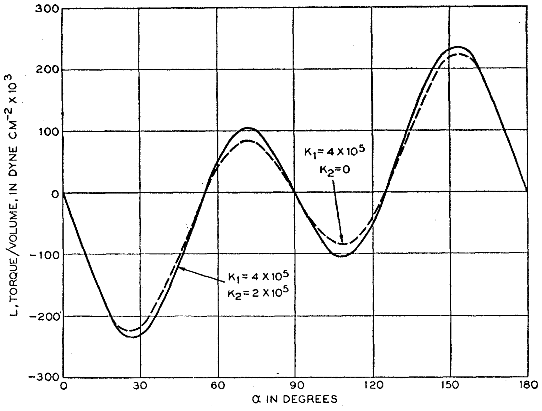

FIG. 1. L *vs.* α for the (110) plane, with (a) $K_1=4(10)^5$, $K_2=0$ (broken line); (b) $K_1=4(10)^5$, $K_2=2(10)^5$ (solid line).

$\cos\alpha=1/\sqrt{3}$ ([111] direction), a position of unstable equilibrium. L is plotted for this plane in Fig. 1 using $K_1=4(10)^5$, $K_2=2(10)^5$, values (in erg cm^{-3}) appropriate for iron. Here the dotted line is drawn for $K_1=4(10)^5$, $K_2=0$, to show the effect of K_2.

Torque data for single crystals have been reported repeatedly in the literature. New data for a crystal of an iron-silicon alloy, cut parallel to (110), are to be reported soon by H. J. Williams.[2]

TORQUE CURVES FOR ROLLED METAL

In fabricated material, the separate crystals of which the material is composed are usually oriented in a special way and as a result there is magnetic anisotropy which is calculable from the constants of the crystal and the distribution of the crystals among the various orientations. Fig. 2 shows the results of torque measurements of an iron disk 0.025 cm thick, cold-rolled from 0.63 cm, taken in a field of 1500 oersteds. The shape of this curve indicates that the coefficient of $\sin 4\alpha$ in the expression for the torque is large compared with the coefficient of $\sin 2\alpha$, and comparison with Table II in Appendix I shows that the crystals are oriented with (100) planes parallel to the sheet and either (1) [001] directions parallel to the direction in which it passed through the rolls, or (2) [011] parallel to the rolling direction. Alternative (1) is correct if K_1 is negative, (2) if K_1 is positive. Since it is well known that K_1 is positive for iron at room temperature, it may be concluded that orientation (2) is correct. This was confirmed by x-ray examination.

The results, and not the details, of the x-ray examination will be given here. It was found that although the mean orientation of all the crystals was such that the [100] direction was normal to the sheet, there were considerable variations from this orientation. The "spread" extends to about 45° rotation of [100] in each sense around the rolling direction, and about one-seventh or one-eighth of this amount around the cross direction. The distribution around the rolling direction is given in Fig. 3.

The effect of this spread in orientation may be taken into account, and K_1 for iron determined from the data obtained with the disk of hard-rolled material. Let the rolling plane (the plane of the disk) be $(hkl)=(100)$ and the direction of rolling be $[h_0k_0l_0]=[011]$, for those crystals lying in the typical position. For those turned from this position by the angle β around the rolling direction, the torque is given by

$$L=-K_1[(2-8\cos 2\beta+6\cos 4\beta)\sin 2\alpha \\ -(7+28\cos 2\beta-3\cos 4\beta)\sin 4\alpha]/64. \quad (6)$$

When $\beta=0$, this reduces to $L=-K_1(\sin 4\alpha)/2$, as given in Table II in Appendix I. Neglecting the effect of the relatively small spread due to rotations about the cross direction, the average value of L for the disk was calculated in terms of K_1 as follows. The coefficient of $\sin 2\alpha$ for a given value of β was multiplied by the fraction of the material having this same value of β, as read from Fig. 3. This product for a series of values of β was plotted against β, and the average value of the product was found between the actual limits $\beta=-45°$ and $\beta=45°$. A similar average was found for the coefficient of $\sin 4\alpha$. The result is:

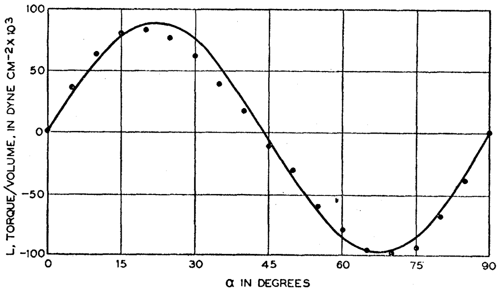

FIG. 2. L *vs.* α for a specimen of hard-rolled polycrystalline iron. The circles are observed points, the curve is calculated from the orientation distribution of crystals as determined by x-rays, with the one adjustable constant K_1 put equal to 415,000.

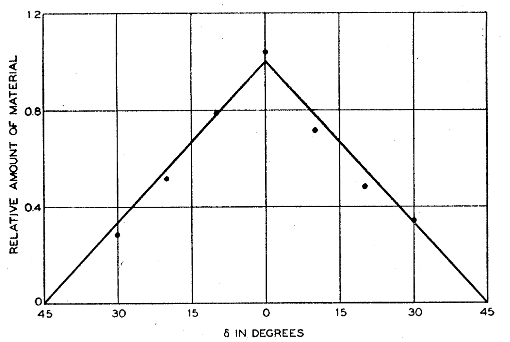

FIG. 3. The distribution of crystals about the rolling direction in the specimen of hard-rolled iron.

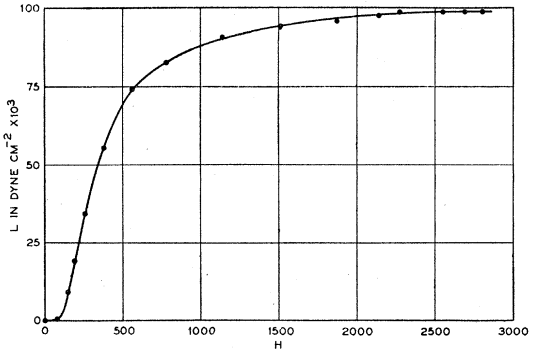

FIG. 4. Variation of torque with field strength for polycrystalline iron, showing finite limit.

$$L = -K_1(0.016 \sin 2\alpha - 0.22 \sin 4\alpha). \qquad (7)$$

The maximum of this curve occurs near $\alpha = 67°$, where $L = -0.23\ K_1$ and, from the measured value of L at this α, K_1 is found to be $4.15(10)^5$ ergs cm^{-3}, in good agreement with previous work on single crystals. Eq. (7), using this value of K_1, is represented by the curve in Fig. 2.

In Fig. 4 the maximum of the L curve is plotted against the field H, and is seen to attain a limiting value when H is about 2500 oersteds. The field of 1500 oersteds used when taking the L, α curve was sufficient to obtain about 95 percent of the maximum L.[3]

Thus the anisotropy constant can readily be determined from measurements on polycrystalline material in the highly strained cold-worked condition. A similar procedure can be applied to annealed material in which fiber structure is present, for example in most materials which have been hard rolled and then recrystallized.

EFFECT OF K_2 ON DIRECTION OF EASY MAGNETIZATION

Using only the K_1 term of Eq. (1), E is found to be a minimum for either [100] or [111], depending only on whether K_1 is positive or negative. When both the K_1 and the K_2 terms are important, E may also be a minimum for [110]. This can be seen from the three following expressions derived from Eq. (1).

$$\begin{aligned} E_{100} &= K_0, \\ E_{110} &= K_0 + K_1/4, \qquad (8) \\ E_{111} &= K_0 + K_1/3 + K_2/27, \end{aligned}$$

where the subscripts on the left-hand side refer to the direction of the magnetization. The directions of easiest, intermediate and hardest magnetization for all combinations of values of K_1 and K_2 are given in Table I. It is apparent that the principal directions of easiest, intermediate and hardest magnetization may have any permutation of the order [100], [110], [111]. The order of ease of magnetization: [100], [110], [111], occurs in iron and the reverse order in nickel. The other sequences may be looked for (1) in those alloy series for which K_1 changes sign, near the composition for which $K_1 = 0$, and (2) in iron or nickel or an alloy at elevated temperatures where K_1 is known to approach zero.[4]

In the iron-nickel series of alloys, K_1 has been reported to pass through zero at 66 to 76 percent nickel[5] and in the iron-cobalt series near 42 percent cobalt.[6] In nickel at 100°C, the data[7] give

TABLE I. *Directions of easiest, intermediate and hardest magnetization.*

K_1	$+$	$+$	$+$	$-$	$-$	$-$
K_2	$+\infty$ to $-9K_1/4$	$-9K_1/4$ to $-9K_1$	$-9K_1$ to $-\infty$	$-\infty$ to $9\|K_1\|/4$	$9\|K_1\|/4$ $9\|K_1\|$	$9\|K_1\|$ to $+\infty$
Easiest	[100]	[100]	[111]	[111]	[110]	[110]
Intermediate	[110]	[111]	[100]	[110]	[111]	[100]
Hardest	[111]	[110]	[110]	[100]	[100]	[111]

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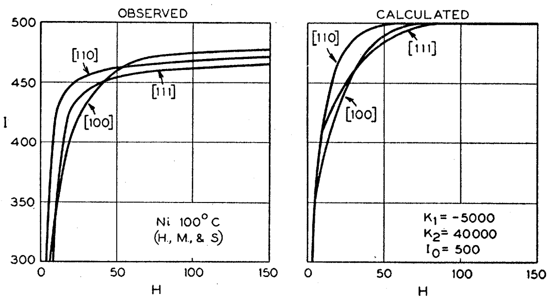
[3] See Appendix II.

[4] E.g., in iron at elevated temperatures the differences in the integrals $\int HdI$ for [110] and [100] seem to approach zero at about 500°C, according to the I, H curves of K. Honda, H. Masumoto and S. Kaya, Sci. Rep. Tohoku Imp. Univ. **17**, 111 (1928). A theory of the temperature dependence of K_1 has recently been given by Akulov, Zeits. f. Physik **100**, 202 (1936).

[5] F. Lichtenberger, Ann. d. Physik **15**, 45 (1932), 71 percent nickel; W. G. Burgers and J. L. Snoek, Zeits. f. Metallkunde **27**, 158 (1935), 66 percent; J. D. Kleis, Phys. Rev. (to be published), 76 percent.

[6] J. W. Shih, Phys. Rev. **46**, 139 (1934).

[7] As calculated from the magnetization curves given by K. Honda, H. Masumoto and Y. Shirakawa, Sci. Rep. Tohoku Imp. Univ. **24**, 391 (1935).



(a) (b)

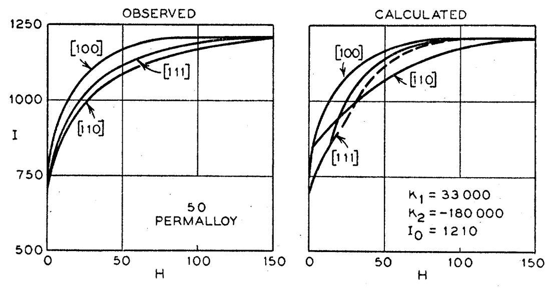
FIG. 5. (a) Experimental I, H curves for nickel at 100°C[7]; (b) I, H curves for [100] and [111] using $K_1=-5000$, $K_2=40{,}000$, $I_0=500$ with the K_0 term adjusted to the data. These show that [110] may be the direction of easy magnetization (a) by experiment and (b) according to theory.

some indication not only that K_1 is small but that it changes sign. In each case the values of K_1 and K_2 were obtained by integration of the I, H curves; that is $E_{hkl}=\int_0^{I_0} HdI$, when H and I are measured in the direction $[hkl]$. Use is then made of Eq. (8) put into the more convenient form:

$$\begin{aligned} K_0&=E_{100},\\ K_1&=4(E_{110}-E_{100}),\\ K_2&=27(E_{111}-E_{100})-36(E_{110}-E_{100}). \end{aligned} \tag{9}$$

The magnetization curves for nickel at 100°C are reproduced in Fig. 5 (a) to illustrate the sequence [110], [111], [100]. Fig. 6 (a) shows the magnetization curves for 50 permalloy, the iron-nickel alloy containing 50 percent nickel. These figures are to be compared with the theoretical magnetization curves discussed in the following section.

CALCULATION OF MAGNETIZATION CURVES



(a) (b)

FIG. 6. (a) Experimental I, H curves for 50 permalloy, according to J. D. Kleis; (b) Calculated I, H curves for $K_1=33{,}000$, $K_2=-180{,}000$, $I_0=1210$, the constants derived from $\int HdI$ for 50 permalloy.

It has been pointed out[8] that the magnetization curves of single crystals taken in different crystallographic directions may be calculated in terms of the anisotropy constant K_1. This is done by adding to E, which represents the magnetic energy of the crystal dependent upon the direction of magnetization relative to the crystal axes, the term $-HI_0\cos\theta$, which represents the energy it has by virtue of the field, and finding the value of θ for which the sum of these terms is a minimum, θ being the angle between the saturation (spontaneous) magnetization I_0 and the field H, as shown in Fig. 7. Thus, in the stable state we have

$$(d/d\alpha)(E-HI_0\cos\theta)=0 \tag{10}$$

or

$$H=\frac{-dE/d\alpha}{I_0\sin\theta d\theta/d\alpha},\quad I=I_0\cos\theta. \tag{11}$$

It is convenient to take $[h_0k_0l_0]$ as one of the directions of easy magnetization and to let α_0 be the angle between $[h_0k_0l_0]$ and H. Then $\theta=\alpha_0-\alpha$ and

$$H=\frac{K_1f_1+K_2f_2}{I_0\sin(\alpha_0-\alpha)},\quad I=I_0\cos(\alpha_0-\alpha), \tag{12}$$

where f_1 and f_2 are trigonometric functions of α, the coefficients of K_1 and K_2, respectively, in the expression for L given in Appendix I:

$$L=K_1f_1+K_2f_2. \tag{13}$$

In Fig. 5 (b) are shown curves calculated by this method for values of K_1 and K_2 consistent with $E_{110}<E_{111}<E_{100}$, namely, $K_1=-5000$ and $K_2=40{,}000$, with $I_0=500$. For calculating the magnetization curve when H is parallel to [100], use is made of the expression for L for $(hkl)=(100)$, $[h_0k_0l_0]=[011]$, $\alpha_0=45°$; and for the [111] magnetization curve are used $(hkl)=(110)$, $[h_0k_0l_0]=[1\bar{1}0]$, $\cos\alpha_0=\sqrt{6}/3$. In the figure the K_0 term has also been added in such a way that in the direction of easiest magnetization the I, H curve corresponds in general form to the experimental curves. Comparison of Fig. 5 (b) with the data for nickel at 100°C, given in Fig. 5 (a), shows that the general character of the curves is the same. The most important point to be noticed

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[8] N. S. Akulov, Zeits. f. Physik **67**, 794 (1931); W. Heisenberg, Zeits. f. Physik **69**, 287 (1931).

is that [110] is the direction of easiest magnetization at all flux densities (except at the very highest flux densities in the experimental curves). According to the ideas here presented, this can happen only when K_2 is important and has a sign opposite to K_1. It should be noticed also that both by calculation and by experiment the [100] and [111] curves cross at about nine-tenths of saturation, the [100] lying below at lower flux densities.

The data[9] and calculations for 50 permalloy are shown in Fig. 6. The constants derived from the data using Eq. (9) are $K_1=33{,}000$ and $K_2=-180{,}000$, consistent with $E_{100}<E_{111}<E_{110}$. The calculation of the magnetization curve for the [110] direction, using these constants and $I=1210$, is straightforward. For the [111] direction, however, an ambiguity appeared when it was found that the I, H curve calculated using Eq. (12), turned backwards toward the I axis at high values of I. In order to clarify this matter, E was plotted as a function of α for various values of H, as shown in Fig. 8. For $H=0$ there are two minima in E, the lower at $\alpha=0$, and the higher at 55°, parallel to H. As H increases, the first minimum moves to a higher α and the second minimum remains at 55°. The first minimum is the lower one until H is about 9.5 and $\alpha=10°$, when the second minimum becomes the lower and consequently the more stable position for the magnetization vector. Thus in the absence of hysteresis the ideal [111] magnetization curve jumps to saturation (I_0) from $I=I_0\cos(55°-10°)=0.71\ I_0=860$, at about $H=10$. In Fig. 6(b) the calculated [111] and [110] curves are shown with a K_0 term added so that the [100] curve is that obtained by experiment. The solid curve marked [111] is that calculated as just described, assuming no hysteresis. If hysteresis were present so that the magnetization continued to lie in the first minimum until that disappeared, the magnetization curve for [111] would be that shown by the dotted line. Simple application of Eq. (12) leads to a curve displaced still farther to the right.

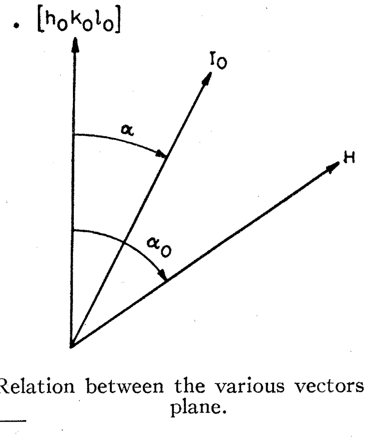

FIG. 7. Relation between the various vectors in an (*hkl*) plane.

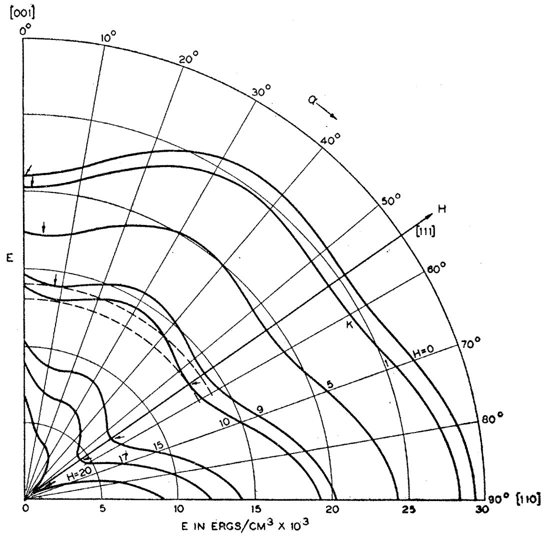

FIG. 8. The energy of a magnetic material as a function of the strength of the field and the direction of the magnetization, in the (110) plane, for $K_0=21{,}000$, $K_1=33{,}000$, $K_2=-180{,}000$.

Comparison of the calculated with the observed curves for 50 permalloy indicates a similarity in general character but none in detail. It does show that the integrals $\int HdI$ increase in the order $E_{100}<E_{111}<E_{110}$ instead of the order $E_{100}<E_{110}<E_{111}$ which would exist if the K_2 term were not taken into account.

No consideration has been given to inclusion of a K_3 term of higher order.

I am indebted to F. E. Haworth of these Laboratories for the measurements of the intensities of the x-ray reflections used in plotting Fig. 3; to L. A. MacColl for checking the various formulas and supervising the calculations; and to H. J. Williams for the measurements of Fig. 2 and for many discussions.

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[9] I am indebted to L. W. McKeehan and J. D. Kleis of Yale University for the opportunity of seeing Mr. Kleis' data before publication.

APPENDIX I

When E is given by Eq. (1), $L=-dE/d\alpha$ may be expressed for any plane (hkl) as a function of α, the angle in the plane between the magnetization and the direction $[h_0k_0l_0]$. The values of the squared cosines to be used in Eq. (1) are the following:

$$S_1^2=A_1+B_1\cos 2\alpha+C_1\sin 2\alpha,$$
$$S_2^2=A_2+B_2\cos 2\alpha+C_2\sin 2\alpha,$$
$$S_3^2=A_3+B_3\cos 2\alpha+C_3\sin 2\alpha.$$

Where

$$A_1=h_0^2/(2H_0^2)+(kl_0-k_0l)^2/(2H^2H_0^2),$$
$$B_1=h_0^2/(2H_0^2)-(kl_0-k_0l)^2/(2H^2H_0^2),$$
$$C_1=h_0(kl_0-k_0l)/(HH_0^2),$$
$$A_2=k_0^2/(2H_0^2)+(lh_0-l_0h)^2/(2H^2H_0^2),$$
$$A_3=l_0^2/(2H_0^2)+(hk_0-h_0k)^2/(2H^2H_0^2),$$

and B_2, B_3, C_2 and C_3 are similar expressions and

$$H^2=h^2+k^2+l^2,$$
$$H_0^2=h_0^2+k_0^2+l_0^2.$$

When $K_2=0$, L is given as follows:

$$\begin{aligned}L/K_1=&+2[A_1(B_2+B_3)+A_2(B_3+B_1)+A_3(B_1+B_2)]\sin 2\alpha\\&-2[A_1(C_2+C_3)+A_2(C_3+C_1)+A_3(C_1+C_2)]\cos 2\alpha\\&+2[(B_1B_2+B_2B_3+B_3B_1)-(C_1C_2+C_2C_3+C_3C_1)]\sin 4\alpha\\&-2[B_1(C_2+C_3)+B_2(C_3+C_1)+B_3(C_1+C_2)]\cos 4\alpha.\end{aligned}$$

TABLE II. *Values of L.*

(hkl)	$[h_0k_0l_0]$	L
100	001	$K_1(-\sin 4\alpha)/2$
100	011	$K_1(\sin 4\alpha)/2$
110	001	$K_1(-2\sin 2\alpha-3\sin 4\alpha)/8+K_2(-\sin 2\alpha-4\sin 4\alpha+3\sin 6\alpha)/64$
110	$1\bar{1}0$	$K_1(2\sin 2\alpha-3\sin 4\alpha)/8+K_2(\sin 2\alpha-4\sin 4\alpha-3\sin 6\alpha)/64$
110	$1\bar{1}1$	$K_1(2\sin 2\alpha+7\sin 4\alpha)/24+K_1(-\cos 2\alpha+\cos 4\alpha)/(3\sqrt{2})+K_2(3\sin 2\alpha+28\sin 4\alpha+23\sin 6\alpha)/576+K_2(-3\cos 2\alpha+8\cos 4\alpha-5\cos 6\alpha)/(144\sqrt{2})$
111	$1\bar{1}0$	$K_2(-\sin 6\alpha)/18$
111	$11\bar{2}$	$K_2(\sin 6\alpha)/18$
211	$01\bar{1}$	$K_1(-2\sin 2\alpha+7\sin 4\alpha)/24+K_2(13\sin 2\alpha-20\sin 4\alpha+25\sin 6\alpha)/576$
211	$1\bar{1}\bar{1}$	$K_1(2\sin 2\alpha+7\sin 4\alpha)/24+K_2(-13\sin 2\alpha-20\sin 4\alpha-25\sin 6\alpha)/576$

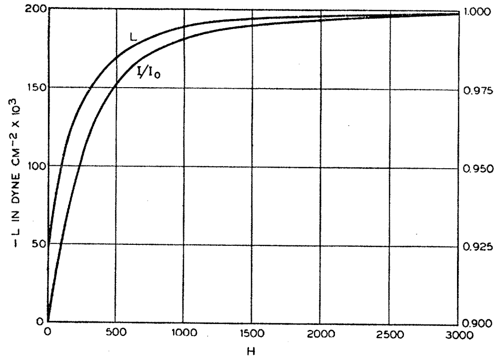

FIG. 9. Calculated L, H and I, H curves for iron with H directed 22.5° from [100] in (001), using the constants $K_1=4(10)^5$, $I_0=1760$.

L has been calculated, with the K_2 term included, for the following combinations of (hkl) and $[h_0k_0l_0]$, given in Table II.

APPENDIX II

For a single crystal—see Fig. 7 and Eq. (12)—

$$H=\frac{K_1f_1+K_2f_2}{I_0\sin(\alpha_0-\alpha)},\quad I=I_0\cos(\alpha_0-\alpha),\tag{14}$$

and from Eq. (13)

$$L=HI_0\sin(\alpha_0-\alpha).$$

Thus, for specific values of (hkl), $[h_0k_0l_0]$, K_1, K_2, I_0 and α_0, both H and L may be calculated as functions of α, and so L as a function of H. Also I_0 is given for any value of L (or H). In Fig. 9, L and I/I_0 are plotted against H for (100), [001], $K_1=4(10)^5$, $I_0=1760$ (constants for iron), $\alpha_0=22.5°$. The curves show, for example, that when $L/L_{\max}=0.9$, $I/I_0=0.994$; in other words, saturation in I is much more complete than saturation in L, in any given field.

Eq. (13) shows why L approaches a finite limit as H becomes indefinitely large. As H increases, α approaches α_0 and L becomes $K_1f_1(\alpha_0)+K_2f_2(\alpha_0)$, which for the constants of the preceding paragraph becomes $2(10)^5\sin 4\alpha_0=2(10)^5$.