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鐵磁性多晶體的旋轉磁化

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一個鐵磁性試品受着磁場的作用而磁化的時候，如果它的磁化强度的增加是由於所包含的磁疇或晶粒內的‘飽和磁强向量’，從接近磁場的‘易磁化方向’，逐漸轉近磁場的方向而產生的，則這種磁化程序叫做‘旋轉磁化’。 一般認爲，曾經剔煉而內脅强很小的鐵磁性多晶體，在磁場强度約 10 至 500 奧斯特間，的磁化完全是反抗‘晶系力’（crystalline forces）的旋轉磁化。

多晶體既然不過是許多單晶體 — 晶粒 — 結合而成，而單晶體的旋轉磁化在理論上又已大體解決了，則多晶體在上述磁場範圍內的磁化曲線應可由平均單晶體，對磁場取各種不同方向時的磁化曲線而得。 在這種計算的企圖中，除了數學上的困難外，還有一個問題是：因各晶粒內的‘飽和磁强向量’的不同向，晶粒間界上有‘自由磁極’的分佈，而產生‘內磁場’。

從另一方面看來，如果上述的‘內磁場’實際上並不存在，則根據 Akulov 式的簡單理論可以預測：屬於同一晶系的，而主要的‘各向異磁性常數’又同號的，多晶體的磁化曲綫，當磁化强度和磁場强度都用適當的‘折合單位’表出時，應該互相重合。 換句話說，多晶體的旋轉磁化，在某些條件下，是有其‘對比態’的。

爲了檢查這一個預測的對否，作者曾測量過純鐵的和含有百分之十五鈷的鐵合金的多晶體，在各種溫度下，的‘正常磁化曲綫’。 結果是，每一多晶體的幾條正常磁化曲綫，用折合單位表出後，都互相重合。 這證明，從簡單理論所得到的推論倒是正確的。 因此可以結論：多晶體的旋轉磁化即使稍微受着‘內磁場’的影響，這影響也不會比單晶體中同樣的影響大。

從近幾年關於磁疇的理論的和實驗的新結果，人們對於單晶體內的磁疇和磁疇間界，在不同情況下，的構造和分佈比以前清楚得多了。 本篇在結束前，即根據這些新發展，對多晶體中並無廣大的‘內磁場’存在這一現象，提出了一些質的解釋。

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ROTATIONAL MAGNETIZATION IN POLYCRYSTALLINE FERROMAGNETICS*

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ABSTRACT

In the attempts to derive from theory the magnetization curve, in the region of rotational magnetization, for well annealed ferromagnetic polycrystals, a generally recognized complication is the possible existence of internal fields caused by the out of alignment of the magnetization vectors of the grains. However, if such internal fields do not really exist, a simple deduction by the usual formal treatment predicts that the magnetization curves of polycrystals, of the same crystalline structure and having the same sign for the values of the principal anisotropy constant, can be brought into coincidence by expressing the intensity of magnetization and the field in proper reduced units. This prediction is checked experimentally by measurements on iron and an iron alloy containing 15 percent cobalt by weight. In each case, it is found that the normal magnetization curves for various temperatures can be brought into coincidence in the manner as indicated. It is therefore concluded that the effect of internal fields on the process of magnetization in the case of polycrystals cannot be any greater than that in the case of single crystals. The absence of wide spread internal fields of magnetic origin in polycrystals during rotational magnetization is then discussed in the light of recent developments in the domain theory.

INTRODUCTION

The magnetization of a well annealed polycrystal of a ferromagnetic, in the field range 10–500 oersteds is presumably entirely rotational. Indeed, it has been generally recognized that the magnetization curve of such a polycrystal can be derived by averaging those of a collection of single crystals randomly oriented with respect to the applied field. On account of mathematical difficulties, such an averaging process cannot be carried through even under the most simplifying assumptions. For this reason, early theoretical works[1, 2, 3] have been confined to the region around the 'knee' of the magnetization curve and that near saturation. To complicate the problem further,

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1. N. Akulov, *Zeits. f. Phys.* **96** (1931), 822.
2. R. Gans, *Ann. der Phys.*, (5), **15** (1932), 28.
3. R. Becker and W. Döring, *Ferromagnetismus* (J. Springer, Berlin, 1939).

there is the question as to the existence of internal fields due to the incomplete alignment of the magnetization vectors of the grains, as it was first pointed out by Holstein and Primakoff[4] in 1941. Assuming the existence of such internal fields, the latter authors rederived the law of 'approach to saturation'. The main result was that, for moderately strong fields, the coefficient of the principal term in that law was reduced to half of what had been previously given. The same result was independently obtained by Néel.[5] Very recently, Stewart and Lawton[6] proceeded to derive a magnetization curve for polycrystals of iron, on the basis of their earlier work on single crystals.[7] On the assumption that the magnetization of each grain was equal to the macroscopic magnetization of the whole specimen, a mathod was developed in which averaging was actually done on the effective fields acting on the individual grains, so that the effect of internal fields was taken care of automatically. However, the agreement between theory and experiment did not come out as satisfactory as in the case of single crystals.

Approaching the same problems from another angle several years ago, the writer obtained some experimental evidence which indicated that no appreciable internal fields existed in polycrystals during rotational magnetization. Owing to war conditions, publication of this work has been long delayed. Now that the problem is again of current interest, it is thought that a presentation of the results might stimulate further investigations.

A Deduction from Simple Theory

Following Akulov's[8] formal treatment, we will investigate what functional relationship exists between the macroscopic magnetization of a specimen and the various energy parameters. Let us at first confine our attention to the magnetization of a single crystal of the cubic system. To proceed, take the three tetragonal axes of the crystal as the x,y,z axes, and, with the z axis as polar axis, form a system of ordinary spherical coordinates, using θ to denote the colatitude and φ the longitude measured from a {100} plane passing through the z axis. Then the part of the free energy density of the crystal which depends on the direction of its intrinsic magnetization vector, $\boldsymbol{I}_s$, is given by

$$U(\theta, \varphi) = \frac{K_1}{4}(\sin^2 2\theta + \sin^4\theta \sin^2 2\varphi) - I_s H\left\{\cos\theta\cos\theta' + \sin\theta\sin\theta'\cos(\varphi - \varphi')\right\} \quad (1)$$

4. T. Holstein and H. Primakoff, *Phys. Rev.* **59** (1941), 388.
5. L. Néel, *J. Phys. Radium* **9** (1948), 184.
6. H. Lawton and K. H. Stewart, *Proc. Phys. Soc.*, A, **63** (1950), 848.
7. H. Lawton and K. H. Stewart, *Proc. Roy. Soc.*, A., **193** (1948), 72.
8. For a general discussion see F. Bitter, *Introduction to Ferromagnetism*, Chap. VI, (McGraw-Hill, New York 1937). Also see reference 3, Chap. 10.

where K_1 is the first magnetocrystalline anisotropy constant,[9] H the external field, θ and φ are the direction parameters of $\boldsymbol{I}_s$, and θ' and φ' those of $\boldsymbol{H}$. The equilibrium orientation of $\boldsymbol{I}_s$ is in the direction of the absolute minimum of $U(\theta, \varphi)$. The conditions for the extreme values of $U(\theta, \varphi)$ are

$$\frac{K_1}{2 I_s H} = \frac{\cos\theta \sin\theta' \cos(\varphi - \varphi') - \sin\theta \cos\theta'}{\sin 4\theta + \sin^2\theta \sin 2\theta \sin^2 2\varphi}, \tag{2}$$

and

$$\frac{K_1}{2 I_s H} = \frac{\sin\theta \sin\theta' \sin(\varphi' - \varphi)}{\sin^4\theta \sin 4\varphi}. \tag{3}$$

The magnetization in the direction of the field, I, is given by

$$I = \frac{\boldsymbol{I}_s \cdot \boldsymbol{H}}{H} = I_s \left\{ \cos\theta \cos\theta' + \sin\theta \sin\theta' \cos(\varphi - \varphi') \right\}. \tag{4}$$

The equation of the magnetization curve is the solution of I from Eqs. (2), (3), and (4) as a function of H with θ', φ', I_s and K_1 as parameters. It is obvious that this solution must be of the form

$$\frac{I}{I_s} = \Psi\left(\frac{K_1}{I_s H}, \theta', \varphi'\right),$$

or

$$j = \Psi(h, \theta', \varphi'), \tag{5}$$

with

$$j = \frac{I}{I_s}, \quad h = \frac{K_1}{I_s H}.$$

Eq. (5) shows that, for a given crystallographic direction, the magnetization curves of single crystals having the same sign for K_1 can be made to coincide by plotting j vs h. This conclusion is reached on the tacit assumption that the crystal has no demagnetizing fields so that it consists of a single domain throughout the rotational magnetization process. In the case of an isolated single crystal, we know that this is not the case. However, if the crystal has a highly symmetrical shape, for example, a sphere, and the field is symmetrically oriented with respect to the directions of easy magnetization (for instance in a $\langle 110 \rangle$ or $\langle 111 \rangle$ direction in the case of iron), our conclusion will be valid, provided that H now denotes the effective field. Bitter[8]

9. The notations for the magnetocrystalline anisotropy constants used in this paper follow the convention of reference 3.

has verified this conclusion by bringing corresponding magnetization curves for various temperatures of an iron single crystal into coincidence.

Furthermore, if for some reason a grain in a polycrystal is not subject to the action of 'free poles', then the value of j for a polycrystal with random grain orientations will be given by the average of Ψ in (5) over all possible values of θ' and φ'. Obviously, such an averaging process will result in an expression of the form

$$j = \Phi(h). \tag{6}$$

Thus the conclusion reached above holds for the case of a polycrystal as well. Of course a real specimen cannot be devoid of surface 'free poles'. However, the effect of surface 'free poles' can be approximately accounted for by the demagnetization factor of the specimen so that if we take H to mean the effective field, which is uniform over the specimen, there will be no need to change our conclusion. Conversely, if the incomplete alignment of the magnetization vectors of the individual grains causes the occurrence of 'free poles' at the grain boundaries and, hence, internal fields, a relation of the type of Eq. (6) cannot be obtained. For, the contents of Eq. (6) mean that, if at a certain state of magnetization, one could, by some means, vary K_1 and I_s in the same proportion but keep the effective field constant, the equilibrium orientations of the individual magnetization vectors would not be disturbed. Now if internal fields due to 'free poles' are present, such fields will, to a first approximation, be proportional to I_s so that they will automatically change with a change in I_s, and, therefore, the orientations of the magnetization vectors cannot be in equilibrium any more.

In passing, some additional remarks seem necessary. First, we have neglected the second magnetocrystalline anisotropy constant K_2 in the above derivation. This may not be allowable. However, existing data give hardly any consistent values to K_2 for iron, while all sets of observations agree that K_2 vanishes for nickel.[3] For these reasons, the importance of K_2 in an actual case is rather doubtful. On the other hand if K_2 is directly proportional to K_1 its presence will not affect our conclusion. Secondly, the condition of random grain orientations is not necessary. All that is required is that comparison be made with polycrystals of the same distribution of grain orientations. Thirdly, our conclusion is valid as long as the magnetization vectors of the grains are rotating under the action of an effective field which is uniform over the whole specimen. It does not imply that the magnetization vector of an individual grain starts rotation from the direction of easy magnetization nearest to the field.

Experimental

If one checks the foregoing prediction by trying to bring the magnetization curves obtained from polycrystals of different substances into coincidence,

one is unlikely to succeed because ordinary polycrystals seldom have exactly the same distribution of grain orientations. Fortunately, both I_s and K_1 vary with temperature, but different rates. Therefore, one can check the prediction by using the magnetization curves obtained from the same specimen at various temperatures.

In the present undertaking, normal magnetization curves were measured on pure iron and an iron alloy containing 15 percent cobalt by weight, in the range from room temperatures to about 50°C below the Curie point. According to a phase diagram compiled by Hansen[10], this alloy is a b.c.c. solid solution below about 910°C. The transformation from the b.c.c. to the f.c.c. phase completes in a temperature range of a few degrees and the latter phase is nonmagnetic. The value of K_1 for the alloy at room temperature, as interpolated from Shih's[11] results, is about 2.7×10^5 ergs/cm^3.

The iron stock was supplied by the Westinghouse Laboratories. According to their information, this iron was quite pure and had very good magnetic properties. The alloy was made by melting weighed amounts of this iron and of a stock of high purity cobalt in vacuum by a high frequency induction furnace. Although no chemical analyses were available for the stock materials, the small coercive forces of the specimens (a few tenths of an oersted at room temperature for $B_{max}=1{,}000$ gauss) testified for the statements about purity. Both specimens were annealed at 900°C for 12 hours and slowly cooled before being measured. The sizes of the cylindrical specimens were:—

Iron: Length, 32.5 cm; diameter, 0.163 cm.

85 Fe—15 Co: Length, 20.0 cm; diameter, 0.252 cm.

The measurements were made by the usual ballistic method, using a sensitive ballistic galvanometer, an air-cored solenoid about 80 cm long, and a noninductively wound tube furnace which was 50 cm long and could be fitted coaxially into the solenoid. The solenoid supplied about 100 oersteds per ampere. The search coil covered a section of only about three centimeters at the middle of the specimen where the temperature and field were essentially uniform. The temperature was measured by a thermocouple placed close to the search coil. Both the intensity of magnetization and the field could be measured to an accuracy of about 0.5 percent. The demagnetizing fields of the specimens were evaluated according to Stäblein and Schlechtweg's[12] results.

Results

Figs. 1 and 2 show the normal magnetization curves of iron and 85 Fe-15 Co respectively, in the field range 5 to 500 oersteds. In order to show the

10. M. Hansen, *Aufbau der Zweistofflegierungen* (J.Springer, Berlin, 1936).
11. J. W. Shih, *Phys. Rev.* **46** (1934), 139.
12. F. Stäblein and H. Schlechtweg, *Zeits. f. Phys.* **95** (1935), 630.

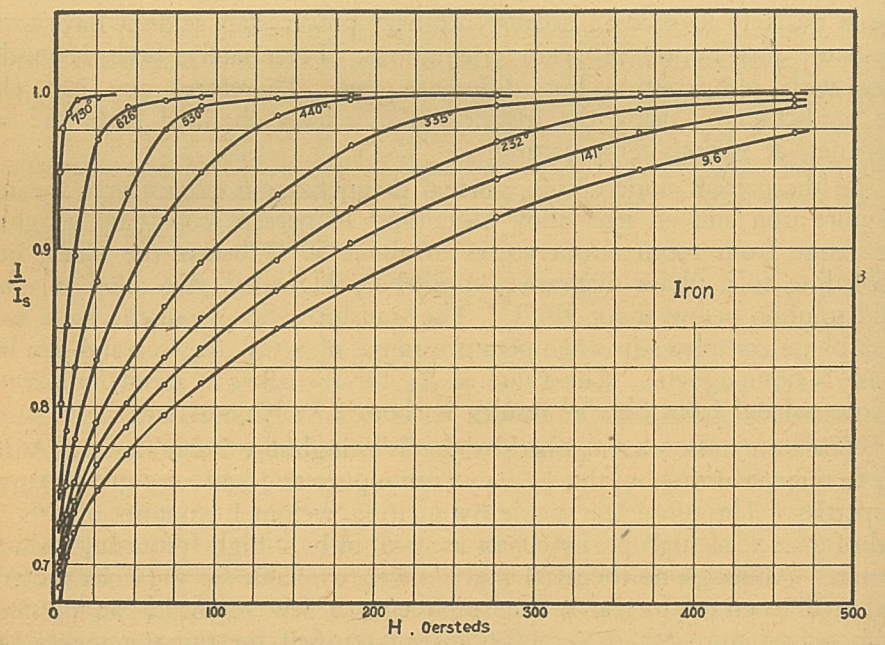

Fig. 1. Normal magnetization curves of pure iron at various temperatures, with the intensity of magnetization expressed in terms of the saturation magnetization.

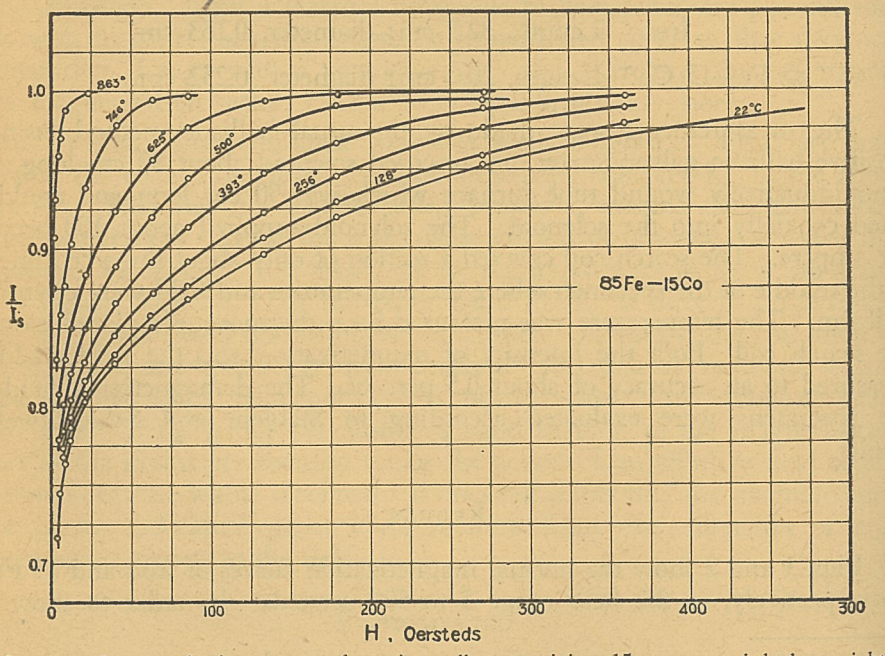

Fig. 2. Normal magnetization curves of an iron alloy containing 15 percent cobalt by weight at various temperatures, with the intensity of magnetization expressed in terms of the saturation magnetization.

effect of temperature more properly, the intensity of magnetization, I, has already been expressed in reduced units and plotted against the effective field H in these figures. Actual measurements were carried up to 1,000 oersteds. As the specimens were soft, the high field portions of the normal curves were practically straight lines with a slight slope. The straight portions of the curves were extended to the I axis and the intercepts thus obtained were taken as I_s. These values of I_s are believed to be within one percent of the true values.

For each specimen, an arbitrary value of I/I_s may be chosen and the ratios, C, of the corresponding value of H on the room-temperature curve to those on curves for elevated temperatures computed. In this way an average value of C can be obtained for each temperature from several I/I_s values. I/I_s can then be plotted against CH for all curves of each specimen. The results for iron and 85 Fe-15 Co are shown in Figs. 3 and 4 respectively. For convenience, most of the points plotted in Figs. 3 and 4 were arbitrarily taken from the smooth curves joining the measured points in Figs. 1 and 2. It is seen that, within the errors of measurements and graphical interpolations, all the points for each specimen lie on the same curve.

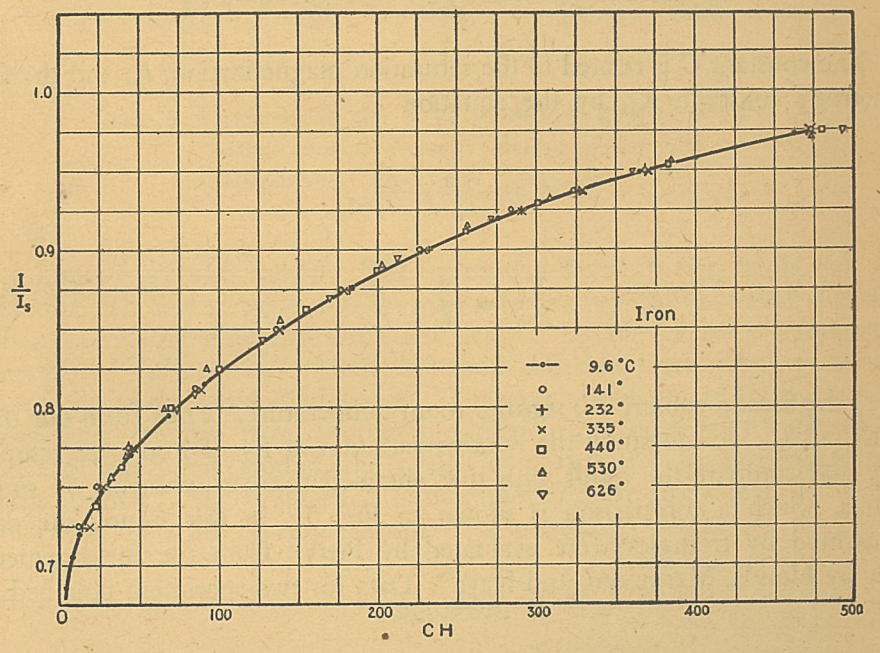

Fig. 3. Normal magnetization curves of pure iron at various temperatures, with both the intensity of magnetization and the field expressed in reduced units.

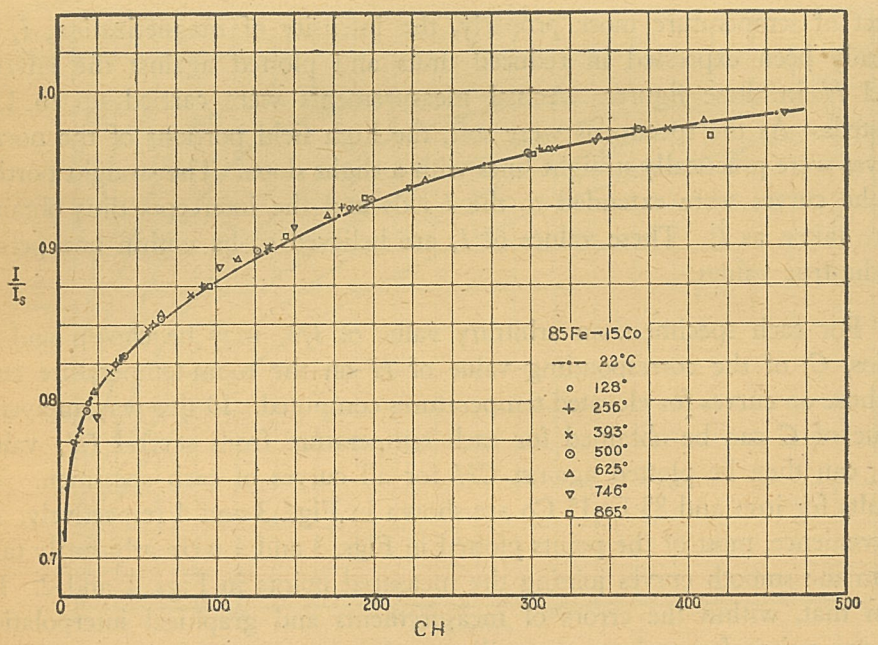

Fig. 4. Normal magnetization curves of the 85Fe-15Co alloy at various temperatures, with both the intensity of magnetization and the field expressed in reduced units.

The constant C is related to the saturation magnetization, I_s, and the first anisotropy constant, K_1, by the equation

$$C = \frac{K_{10}}{I_{s0}} \Big/ \frac{K_1}{I_s},$$

or

$$\frac{K_1}{K_{10}} = \frac{1}{C} \frac{I_s}{I_{s0}},$$

where the second subscript 0 signfies room temperature. Thus from our data for C and I_s we can study the relative variation of K_1 of iron with temperature and compare the result with that obtained from experiments on single crystals. Such a comparison is shown in Fig. 5. In this figure, the data represented by triangles were evaluated by Piety[13] from the measurements made by Honda, Masumoto, and Kaya[14]. Data for two specimens obtained by

13. R. G. Piety, *Phys. Rev.* **50** (1936), 1173.

14. K. Honda, H. Masumoto, & S. Kaya, *Tohoku Imp. Univ. Sci. Rep.*, Series 1, **17** (1928), 111.

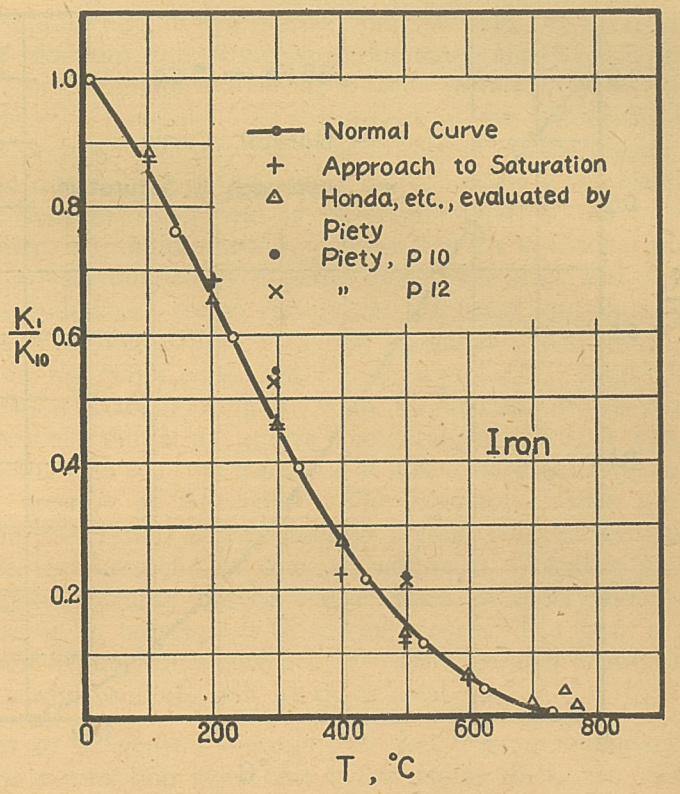

Fig. 5. Comparison of data on the fractional variation of the first magnetocrystalline anisotropy constant of pure iron with temperature.

Piety himself are also shown. The agreement between our result and that of Honda, etc., is very good, but the points for Piety's specimens are distinctly higher. There are differences of a few degrees among the room temperature values of the different sets of data, but these differences are not large enough to account for the latter discrepancy. The points represented by the upright crosses were obtained from another specimen of the same iron stock by 'approach to saturation' studies, the results of which will be reported shortly in this journal. Fig. 6 shows a similar plot for the iron-cobalt alloy, but, in this case, there are no single-crystal data to compare with.

Discussion

From the results just described, we may conclude that internal fields are not present in a polycrystal to an extent great enough to interfere with the

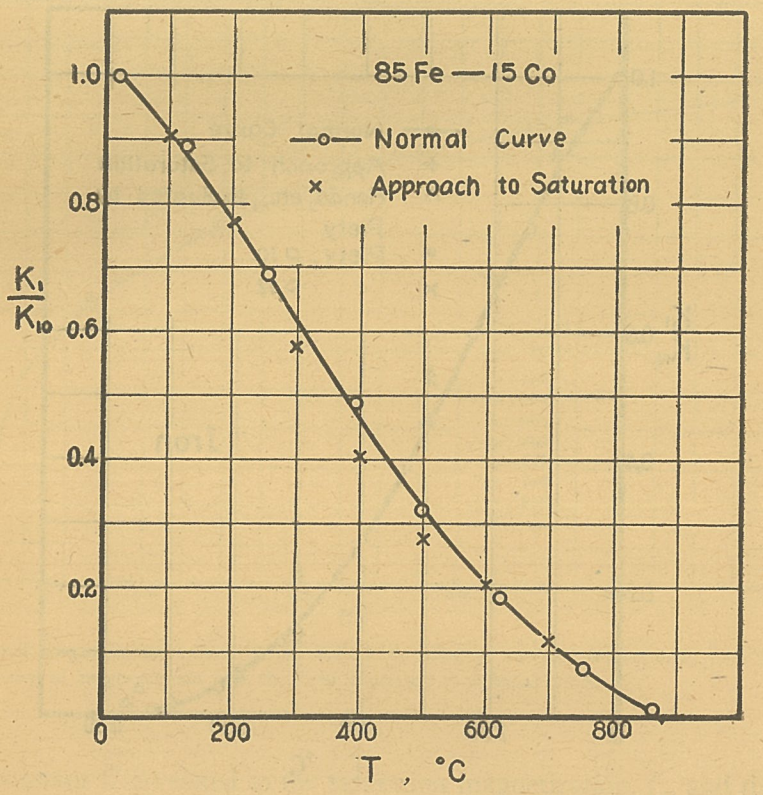

Fig. 6. The fractional variation of the first magnetocrystalline anisotropy constant of the 85Fe-15Co alloy with temperature.

rotation of the magnetization vectors of the grains by the over-all effective field. Therefore, an average grain in a polycrystal behaves like a single crystal without demagnetizing fields of its own. This conclusion seems at first sight rather surprising. However, viewed in the light of the results of recent theoretical and experimental studies on the formation of domains in single crystals by Lifshitz[15], Néel[16], and Williams, Bozorth, and Shockley[17], this situation seems qualitatively quite understandable. These authors have shown that owing to the effect of the demagnetization factor, a single crystal of common shape is divided into domains even during rotational magnetization; that the domain boundaries must be plane and must so orient themselves that 'free poles' are eliminated; that the major domain formation do not extend to the surface of the crystal where 'free poles' would develop to

15. E. Lifshitz, *J. Phys. U.S.S.R.* **8** (1944), 337.
16. L. Néel, *J. Phys. Radium* **5** (1944), 265.
17. H. J. Williams, R. M. Bozorth, and W. Shockley, *Phys. Rev.* **75** (1949), 155-183.

disturb the equilibrium of the domain magnetization vectors, and therefore 'flux closure' domains and, sometimes, domain branching are formed. On the basis of these fundamental ideas, a crude picture of what happens in a polycrystal may be imagined. At the surface of the polycrystal, the domain formation is probably not very different from that on the surface of a single crystal. The distribution of 'free poles' is reduced to a minimum, with the macroscopic demagnetizing field as the resultant effect. As for the interior of the specimen, it is important to remember that the individual grains are not isolated but are surrounded by material of its own kind, and, since domains and domain boundaries are so readily created, large concentrations of 'free poles' at the grain boundaries can hardly be stable. Thus, during rotational magnetization, the major portion of a grain is probably a single domain with its magnetization vector rotating out from the direction of easy magnetization nearest to the field, but, at the grain's boundaries there will be a minor formation of domains such that the transport of flux from grain to grain is affected without the creation of 'free poles'. The existence of such minor domain formations in polycrystals as well as single crystals will not show up conspicuously in flux measurements of ordinary accuracy. Whether such a picture is any thing near reality can be best checked by observation of magnetic powder patterns on polycrystals of special texture. For iron, a rolled sheet in which most of the grains have a {100} plane parallel to the rolling plane, but are otherwise random, will be quite ideal.

It seems to be generally recognized that a grain boundary in a pure polycrystal is a transition layer only a few atoms thick. In it, the ordered arrangement of atoms is more or less broken up but the short range forces between atoms are still effective so that a Bloch wall can extend through a grain boundary. It can then be alternatively suggested that a grain boundary is probably the seat of a domain boundary which completely envelops the former. Such a domain boundary will so adjust its orientation that the usual function of transporting flux without the creation of 'free poles' is achieved, but will be different from those of the usual kind in that it is not affected by a field as far as body displacement is concerned. In this connection, a situation that is conceivable but not so easy to justify is that, at the early stages of rotational magnetization, not all the grains may have their magnetization vectors rotating away from the direction of easy magnetization nearest to the field. For, a given grain boundary may be so oriented with respect to the directions of easy magnetization nearest to the field in the grains on both sides of it that the creation of a domain boundary to cover it would be energetically unfavorable. In such a case, the magnetization vector in one of the grains may start out from a direction of easy magnetization which is not nearest to the field. The occurrence of such a situation can probably be detected in the macroscopic remanence but not by the type of experimental observations as we have made.

The problem of calculating the macroscopic magnetization of a specimen

is a search for the state of minimum free energy. Not being able to solve it by taking all the energy terms into account at once, previous investigations have considered the various energy terms more or less separately, approaching thereby an integrated picture. Whatever the final picture will be, experimental results seem to show that the wide spread occurrence of 'free pole' concentrations in a specimen is precluded.

Acknowledgement

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