

Ferromagnetic Anisotropy of Iron Crystals at Various Temperatures*

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Magnetization curves in important crystallographic directions in small oblate spheroids cut from hydrogen-purified iron crystals show ferromagnetic anisotropy of the same sort as that previously reported for iron of less purity. At room temperature agreement with earlier work is good. At 296°C and at 500°C the principal (fourth-order) term in the anisotropy is larger, at 650°C and 696°C the same term is smaller than in earlier studies, anisotropy not being detected. The subsidiary (sixth-order) term is not accurately determined. The pendulum magnetometer method, which has certain advantages for such purposes over classical methods, was used and results for different spheroids agree well, especially at the higher temperatures.

INTRODUCTION

THE ferromagnetic anisotropy of iron crystals has been measured by Beck,¹ Webster,² Honda and Kaya,³ Honda, Masumoto and Kaya.⁴ The last of these studies covered the range in temperature from 5°C to 770°C, the others were confined to room temperature. Beck and Webster measured the torque on disks suspended in the air-gap of an electromagnet, Honda and Kaya measured the flux change through a search coil at the center of which an oblate spheroid was suddenly turned from one azimuth to another in the field of a magnetizing solenoid. Honda, Masumoto and Kaya used one of the same spheroids but changed the method of measurement by leaving the specimen in one azimuth during each cycle of heating and cooling, getting I - H curves at selected temperatures by reversing various currents in the solenoid.

All the data agree in fixing the tetragonal axes, of form $\langle 100 \rangle$, as the directions for easiest magnetization, the trigonal axes, of form $\langle 111 \rangle$, as the directions for most difficult magnetization. The digonal axes, of form $\langle 110 \rangle$, are intermediate between these extremes. The quantitative differences between results from different laboratories are larger than the errors in measurement can easily explain, so that they must be ascribed in part to differences in chemical purity,

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¹ K. Beck, *Dissertation* (Zürich, 1918); *Vierteljahrsschrift der naturforschenden Gesellschaft in Zürich* **63**, 116-186 (1918).

² W. L. Webster, *Proc. Roy. Soc.* **A107**, 496-509 (1925).

³ K. Honda, S. Kaya, *Sci. Rep. Tohoku Imp. Univ.* [1] **15**, 755-770 (1926).

⁴ K. Honda, H. Masumoto, S. Kaya, *Sci. Rep. Tohoku Imp. Univ.* [1] **17**, 111-130 (1928).

in mechanical and thermal treatment. Under these conditions it seemed worth while to re-measure the anisotropy in very pure and nearly unstrained iron by a method capable of fair precision and relatively free from systematic errors. Such a method is that developed in this laboratory⁵ and first applied here by J. W. Shih⁶ to iron-cobalt alloys. It uses an accurately made oblate spheroid carried on a light pendulum at the common center of a magnetizing solenoid and a pair of coils producing a weak but spatially uniform magnetic field gradient. These various windings have a common horizontal axis so that the resultant force on the specimen is also horizontal and can be measured by the deflection of the pendulum. The computed value of I then depends, except for a factor constant for each specimen, only upon the measured deflection of the pendulum and the measured current in the gradient coils. The absolute value of H within the specimen depends upon the measured current in the magnetizing solenoid and upon the value of I , but the difference between values of H along different diameters of the spheroid for the same value of I does not depend upon I , and it is this difference which is most important in the analysis of the results. Points on I - H curves near I_{sat} are particularly important in anisotropy analyses. These points are determined with greatest precision by the new method, with least precision by the older methods.

Several improvements in technique, recently described⁷ were devised and tested during the progress of the work reported herein.

⁵ L. W. McKeehan, *Rev. Sci. Inst.* **5**, 265-268 (1934).

⁶ J. W. Shih, *Phys. Rev.* **46**, 139-142 (1934).

⁷ L. W. McKeehan, R. G. Piety, J. D. Kleis, *Rev. Sci. Inst.* **7**, 494-497 (1936).

SPECIMENS

Comparison of properties in different directions is safest when as many as possible of the important directions are accessible in one specimen. This condition is best met, as is well known, in an oblate spheroid with its equatorial plane of the crystallographic form $\{110\}$. If we choose to call this the $(01\bar{1})$ plane we have lying in it the following important axes: $[100]$, $[011]$, $[111]$, $[1\bar{1}\bar{1}]$. Principal effort was therefore expended on spheroids of this sort, $(01\bar{1})$ spheroids. As a check a (001) spheroid was also cut, containing the important axes: $[100]$, $[010]$, $[110]$, $[1\bar{1}0]$.

The iron was very kindly furnished by the Bell Telephone Laboratories in three flat pieces about 3 mm thick and several centimeters in diameter. These were parts of boxes in which P. P. Cioffi had purified small iron specimens by washing them with pure hydrogen at about 1450°C for 18 hours or more. His studies on this process,⁸ which produces extreme magnetic and mechanical softness, lead to the expectation that the only nonmetallic impurities remaining in these boxes in measurable amounts are nitrogen, sulphur and phosphorus. The first three of these together should not exceed 0.01 weight percent. Silicon may be in excess of this limit, especially in those parts of the box which are in contact with the alundum support, but these parts were not used here. A provisional limit of 0.03 weight percent silicon is suggested. Manganese and copper may amount to about 0.02 weight percent each. It can be stated with confidence that this iron is more nearly pure Fe than any previously used in anisotropy studies.

The crystals present in the pieces as furnished were of ample size for the pendulum magnetometer. The surface was first etched rather deeply with a mixture of water solutions of HNO_3 and HCl , restrained with $\text{C}_2\text{H}_5\text{OH}$. By observing reflections from etch planes—here of form $\{100\}$ —crystals of suitable size and orientation were selected. These were cut out by hand with a jeweler's saw and mounted on an optical goniometer of special design⁹ for more accurate observation. The orientation of the $\langle 100 \rangle$ axes

being thus determined with respect to the surface and edges of the fragment a rectangular iron bar was cut off at such an angle that when the fragment was cemented (with chaser's cement) on the cut in the proper azimuth, the axis of the spheroid to be cut was parallel to the axis of the bar. The blank was now faced off by extremely light slow cuts on a Brown and Sharpe surface grinder with a diamond-dressed wheel running at high speed. It was remounted on the squared end of a new holder and recut in the same way until the thickness was less than 0.5 mm but still somewhat greater than that of the proposed spheroid. It was now ready for an x-ray inspection of its orientation and freedom from distortion by the Laue method, using transmitted radiation so as to get a volume average. If the plane of the blank was found to be more than about 1° from the proposed crystallographic plane a new holder was cut and both sides were reground, otherwise only one side was reground. In either case the thickness was brought down to 0.006 cm over the proposed minor axis of the spheroid and the blank was filed, on its holder, to a circle a little larger than the proposed equatorial section. From this point on, the procedure has been described in the previous paper.⁷

The spheroids were finally given a second x-ray inspection by the Laue method to make sure that no serious distortion had resulted in the last stages of shaping. The criterion was that the Laue spots should be symmetrical ellipses without more than traces of radial asterism.

The applied field intensity, $H+NI$, is very much greater than the effective field intensity H in an oblate spheroid of practicable dimension-ratio (diameter/thickness). It therefore becomes very important to prove that the demagnetizing field, NI , is reasonably uniform in intensity throughout the volume, as the method of measurement supposes. Partly to test this and partly for other more obvious reasons two spheroids having the same orientation but differing in eccentricity were tested over the whole available range in temperature. The equatorial plane in each of these lay within the accuracy of the x-ray analysis (about 1°) parallel to the $(01\bar{1})$ plane. Their characteristics are given in Table I. Under N_{obs} is given the demagnetizing factor necessary to make the effective field sensibly zero up

⁸ P. P. Cioffi, Phys. Rev. 39, 363-367 (1932); 45, 742 (1934).

⁹ L. W. McKeehan, H. J. Hoge, Zeits. f. Krist. 92, 476-478 (1935).

TABLE I. Data on spheroids (011̄).

Designation	Dimension Ratio	Major Axis (cm)	Minor Axis (cm)	Volume 10 ⁻³ cm ³	Mass (g)	N _{obs}	N _{calc}
P10	10/1	0.318	0.0318	1.768	0.0139	0.912	0.875
P12	12/1	0.307	0.0256	1.234	0.0097	0.710	0.743

to half-saturation in the actual measurements of I and H . This differs appreciably from N_{calc} , the demagnetizing factor for a perfect spheroid of the measured dimensions, but since N_{obs} was not dependent upon the direction along which H was applied it seems reasonable to suppose that the slight departures from ellipticity thus indicated are symmetrical with respect to the minor axis of each spheroid and that any resultant inhomogeneity in H and I throughout the volume is not so great as to make the measured averages misleading. It should be noticed that N_{obs} was greater than N_{calc} in one case, less in the other. A third spheroid cut with its equatorial plane parallel to (001) was tested less completely, but showed directional independence of N_{obs} and fair agreement between N_{obs} and N_{calc} .

Temperatures higher than 700°C could not, with available means, be kept constant enough to make I - H curves precise enough for anisotropy work. The difficulty is that temperature fluctuations change I_{sat} by relatively great amounts at temperatures near the Curie point so that it becomes very hard to take even one complete I - H curve under sufficiently constant conditions. So far as some unsatisfactory experiments indicated, however, there was no detectable anisotropy in magnetization above 700°C in this iron.

At 296°C and at 500°C the differences between I - H curves for the two (011̄) spheroids were so small that single curves were drawn to fit the combined data. At room temperature, 18°C, the data were treated separately.

Fig. 1 shows representative I - H curves and Table II gives areas included between such curves and the two lines $I=I_{\text{sat}}$ and $H=0$, together with derived quantities. All curves were carried to saturation, applied fields as high as 1800 oersted being used if necessary. Some of the areas differ slightly from those published in my dissertation. This occurs because all the areas have been redetermined by a more precise method.

TABLE II. Work needed to magnetize.

Specimens	°C	10 ⁴ erg. cm ⁻³				K ₂
		W ₁₁₁	W ₁₁₀	W ₁₀₀ =K ₀	K ₁	
P10	18	16.10	13.16	2.48	+42.7	-17
P12	18	16.59	12.38	1.33	+44.2	+14
P10, 12	296	8.08	6.45	0.65	+23.2	-8
P10, 12	500	2.56		0.21	+9.4	-21
P12	675	0.19			0	0
P12	696	0.31			0	0

ANALYSIS OF RESULTS

In the manner proposed by Gans¹⁰ we suppose that the work needed for isothermal magnetization to saturation, measured by the area defined above, is for any arbitrary direction in an unconstrained cubic crystal given by

$$W_{\alpha} = K_0 + K_1(\alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2 + \alpha_1^2\alpha_2^2) + K_2(\alpha_1^2\alpha_2^2\alpha_3^2). \quad (1)$$

In this equation α_1 , α_2 , α_3 are the direction cosines of the chosen direction referred to the three $\langle 100 \rangle$ axes, K_0 , K_1 and K_2 are not direction dependent and the only assumption made is that an equation of the sixth degree in the cosines is adequate.¹¹ The symmetry of the crystal annuls all other combinations of α_1 , α_2 , α_3 . For the important directions of form $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ Eq. (1) reduces to

$$W_{100} = K_0; \quad W_{110} = K_0 + K_1/4; \quad W_{111} = K_0 + K_1/3 + K_2/27. \quad (2)$$

These equations are not new. Some authors¹² use instead of K_1 a constant which in this notation is $2K_1$.

It is also possible, as Akulov¹² has shown, to compute I - H curves for particular values of K_1 , assuming K_2 to be zero. As might be expected the computed curves agree best with experiment at room temperature and are not good approximations at high temperatures.

¹⁰ R. Gans, *Physik. Zeits.* **33**, 924-928 (1932).

¹¹ The analysis which follows differs in one important respect from that presented in my dissertation. R. M. Bozorth has pointed out in private discussion that the sixth degree term in Eq. (1) is capable of explaining anisotropy hitherto regarded as completely anomalous. I have therefore used his method of computing the coefficient of this term, and have included it in Tables II and III. I am much indebted to him for the opportunity of using his method of analysis prior to its publication.

¹² Especially N. S. Akulov, *Zeits. f. Physik* **57**, 249-256 (1929); **67**, 794-807; **69**, 78-99 (1931).

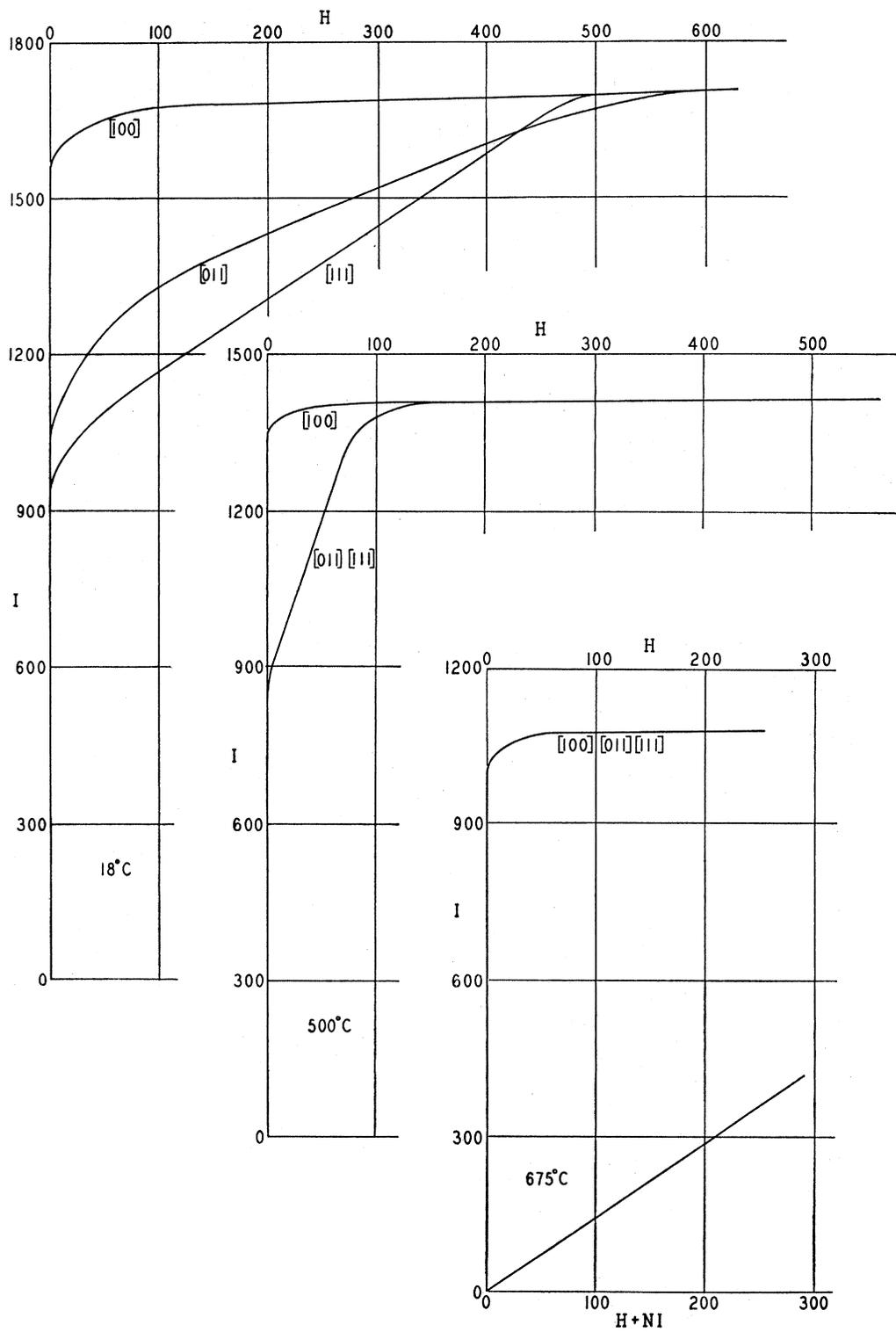


FIG. 1. Magnetization curves for specimen P12 at three temperatures, 18°C, 500°C and 675°C. In the last case the $I-(H+NI)$ curve is also shown (in part) to illustrate the importance of N .

It will be noticed that in Table II the constant K_2 varies widely in absolute value and even in sign. As far as these data go the necessity for the sixth degree correction is apparent only in the fact that W_{111} and W_{110} are indistinguishable at and above 500°. (Precise equality requires $K_2 = -9K_1/4$.) If the values of K_1 are plotted against temperature they fall on a straight line which intersects the line $K_1 = 0$ at 630°C. That K_1 does not actually become negative, however, is suggested by the tabulated values for 675° and 696°C. No permanent effect of heating and cooling the specimens during the measurement was detected at room temperature.

The agreement with the work of Honda, Masumoto and Kaya⁴ is very good at room temperature, not so good at the higher temperatures. For comparison, values of K_0 , K_1 and K_2 have been computed from their tables. In making this computation it has been assumed as in the present work that the correct demagnetizing factor to be used in getting H is that which makes the I - H curve rise vertically from the origin. This makes it necessary to decrease the areas derived from their tabulated H and I by an amount not exceeding 0.5×10^4 erg. cm⁻³ in any case, and usually much less. Where their maximum value of I does not occur at their maximum value of H it may be concluded that I has been over-corrected for flux through the air surrounding the specimen and the final drop in I has then been ignored in computing the area. Finally, small differences in I_{sat} have been corrected for by multiplying the computed areas by appropriate factors nearly equal to unity. The result of all this is to give a series of areas from which K_0 , K_1 and K_2 may be computed as already explained. Table III presents the results. The values of K_1 and K_2 , especially the latter, have been made to progress more smoothly up to 700°C by using faired curves for W_{100} , W_{110} and W_{111} to find the differences used in computation. Our values for

TABLE III. *Work needed to magnetize.* Derived from data of Honda, Masumoto and Kaya.

°C	10 ⁴ erg. cm ⁻³		
	K_0	K_1	K_2
5	0.60	+41.3	+14
100	0.27	+35.3	+20
200	0.26	+27.0	+22
300	0.38	+18.9	+19
400	0.015	+11.4	+11
500	0.014	+ 5.4	+ 9
600	0.065	+ 2.6	- 1
700	0.14	+ 1.0	- 2
750	0.21	+ 1.7	-12
770	0.33	+ 0.8	- 4

K_1 at 296°C and 500°C are considerably higher than those at 300°C and 500°C in Table III. It will be noticed that K_0 seems to rise a very little as the Curie point is approached in both sets of experiments. No one would be likely to suggest a linear drop in K_1 with increasing temperature from the values given in Table III. An exponential drop like that suggested for nickel by Akulov¹³ in a very recent paper is a fair approximation above 200°, but not below.

In conclusion it can be stated that the purest iron we could procure is both qualitatively and quantitatively much like the iron used by Honda and his co-workers. It seems, however, to retain its anisotropy better at 300°C and 500°C and to lose it more suddenly in the neighborhood of 600°C. The sixth-order term in the anisotropy equation is relatively unimportant except at temperatures above 500°C where the fourth-order term falls to negligible values.

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¹³ N. S. Akulov, *Zeits. f. Physik* **100**, 197-202 (1936).