

## Ferromagnetic Anisotropy in Nickel-Cobalt-Iron Crystals at Various Temperatures

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Single crystals of ternary alloys of nickel, cobalt and iron with forty percent nickel or more have been examined, with a pendulum magnetometer, for ferromagnetic anisotropy. All alloys in this region have face-centered cubic structures, like that of nickel. A region of low anisotropy, for nearly equal cobalt and iron contents, appears to be connected with the points where binary alloys of nickel and cobalt on one side and of nickel and iron on the other side change their anisotropy types. Measurements were

made in all cases at room temperature, at about 200°C, and at about 400°C. At the higher temperatures anisotropy of either type generally diminishes and, in the region of low anisotropy, the principal anisotropy constant may change sign. The previously reported data for the binary alloys and single components in this system are recomputed and presented so as better to show the boundaries of the ternary field.

THE experimental work here reported may be considered as an extension of studies on binary alloys of nickel and cobalt<sup>1</sup> and of nickel and iron,<sup>2</sup> and is confined to the nickel-rich corner of the nickel-cobalt-iron triangular diagram. In this entire region there is only one solid-solution phase with the face-centered cubic structure characteristic of nickel at room temperature and of cobalt and iron at much higher temperatures. Whether or not there are any super-structures in this region has not yet been decided, and the results here presented do not

seem to furnish evidence for or against their existence.

G. W. Elmen and G. A. Kelsall, of the Bell Telephone Laboratories, were kind enough to let me select eight ternary alloys from their extensive series.<sup>3</sup> The compositions, given in Table I, are also shown by the dots inside the boundary in Fig. 1. All were obtained as polycrystalline rods 0.32 cm in diameter. They were remelted under a low pressure of hydrogen and progressively frozen to give castings about 0.6 cm in diameter from which single crystals of suitable

TABLE I. *Description of specimens.*

Ni-Co-Fe, NOMINAL	80-10-10	70-20-10	65-15-20	60-30-10	60-25-15	50-40-10	50-25-25	40-10-50
CASTING NUMBER	950	1092	898	979	899	926	1006	941
WEIGHT PERCENT Ni	79.29	69.38	64.56	61.19	60.05	50.79	50.26	40.19
Co	10.21	20.20	14.84	28.38	24.79	38.66	25.04	10.21
Fe	10.27	9.85	20.52	10.43	15.12	10.41	24.63	49.51
Mn	0.46	0.42	0.33	0.19	0.32	0.31	0.27	0.32
DENSITY, G.CM <sup>-3</sup>	8.796	8.793	8.633	8.748	8.703	8.741	8.574	8.309
SPHEROIDS (011̄)								
DIAMETER, 10 <sup>-4</sup> CM	3140	3103	3021	3013	2920	2810	3044	3168
THICKNESS, 10 <sup>-4</sup> CM	312	262	249	247	244	229	250	265
MASS, 10 <sup>-3</sup> G	14.4	11.6	10.4	10.5	9.68	8.58	10.5	11.45
VOLUME, 10 <sup>-6</sup> CM <sup>3</sup>	1637	1319	1205	1200	1112	982	1225	1378
DEMAGNETIZING FACTOR	0.860	0.740	0.720	0.738	0.739	0.720	0.739	0.730
ORIENTATION FACTORS								
[100] $f_1(0)^*$	0.0009	0.0077	0.0019	0.0116	0.0076	0.0191	0.0200	0.0016
$f_2(0)^*$	0	0	0	0	0	1	0	0
[011] $d_1(0.25)^*$	2496	2433	2509	2500	2500	2546	2573	2479
$d_2(0)^*$	1	18	5	5	0	45	42	2
[111] $t_1(0.3)^*$	3331	3247	3320	3277	3316	3196	3210	3308
$t_2(0.037)^*$	370	341	366	353	365	323	335	361

\* Ideal values, for explanation see text.

<sup>1</sup> J. W. Shih, Phys. Rev. 50, 376-379 (1936).

<sup>2</sup> J. D. Kleis, Phys. Rev. 50, 1178 (1936).

<sup>3</sup> G. W. Elmen, Bell Sys. Tech. J. 15, 113-135 (1936) gives references to earlier papers.

size were chosen for the preparation of oblate spheroids. The technique of this process, and the use of the pendulum magnetometer to find magnetization curves for important crystallographic directions lying in the equatorial plane of a spheroid, have been described in previous papers.<sup>4</sup> Table I gives the necessary information concerning the eight specimens used in measurements. Most of the entries are self-explanatory but the part of the table headed "Orientation Factors" requires some further remark.

Although efforts were made to cut each spheroid so that its equatorial plane should be the crystallographic plane (01 $\bar{1}$ ), and to mark a diameter parallel to the crystallographic direction [100], neither operation was precisely accomplished. Careful x-ray inspection, after all magnetic measurements had been made, gave the actual indices of the directions, nominally [100], [011] and [111], in which measurements had been made. The work per unit volume necessary to magnetize a crystal to saturation in a direction with direction cosines  $\alpha_1$ ,  $\alpha_2$ ,  $\alpha_3$  referred to the three cubic axes of the crystal can be approximated by the equation<sup>5</sup>

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

in which  $K_0$ ,  $K_1$  and  $K_2$  are energy-density coefficients independent of direction. For small angles of departure from a fourfold (cubic) axis we may write this

$$W_{100} = K_0 + f_1K_1 + f_2K_2, \quad (2)$$

where  $f_1$  and  $f_2$  are the appropriate functions of the direction cosines. For ideal cutting  $f_1=0$  and  $f_2=0$ . Similarly for a direction near a digonal axis,

$$W_{011} = K_0 + d_1K_1 + d_2K_2, \quad (3)$$

where the ideal values are  $d_1=1/4$ ,  $d_2=0$ . Finally, near a trigonal axis,

$$W_{111} = K_0 + t_1K_1 + t_2K_2, \quad (4)$$

where the ideal values are  $t_1=1/3$ ,  $t_2=1/27$ . The experimentally determined values of the six

<sup>4</sup> L. W. McKeehan, Rev. Sci. Inst. **5**, 265-268 (1934); L. W. McKeehan, R. G. Piety, J. D. Kleis, Rev. Sci. Inst. **7**, 494 (1936); R. G. Piety, Phys. Rev. **50**, 1173 (1936).

<sup>5</sup> R. Gans, Physik. Zeits. **33**, 924-928 (1932); R. M. Bozorth, Phys. Rev. **50**, 1077 (1936).

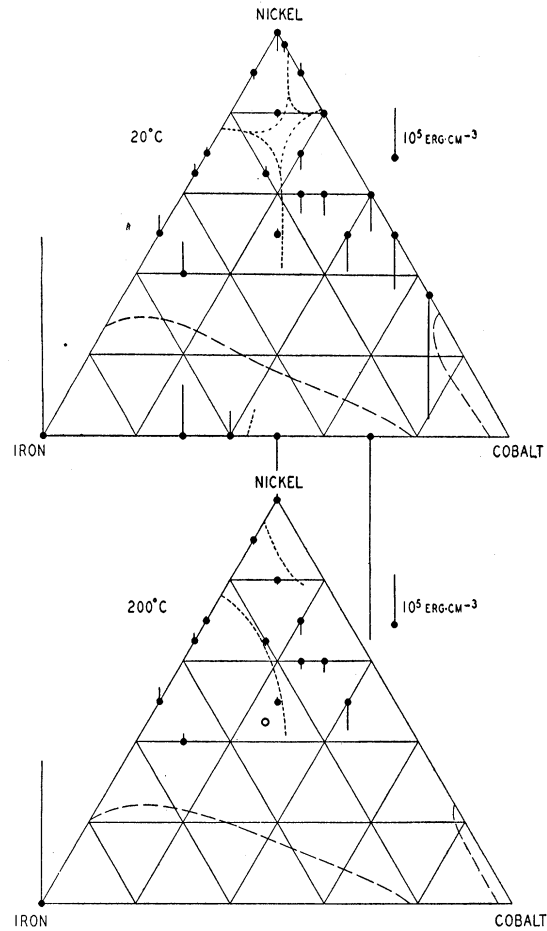


FIG. 1. The principal anisotropy constant ( $K_1$ ) as a function of composition in nickel-cobalt-iron alloys at 20°C (upper figure) and at 200°C (lower figure). Lines up or down from a dot indicate positive or negative  $K_1$  according to the scale at the right. Dashed lines show phase boundaries, dotted lines show changes in sign of  $K_1$ .

coefficients thus defined are given in Table I for each spheroid. It will be noticed that the deviations from the ideal values are not large in any case. Once the coefficients are determined the values of  $K_0$ ,  $K_1$  and  $K_2$  can easily be found from the experimental values of  $W_{100}$ ,  $W_{011}$  and  $W_{111}$  at any temperature.

The procedure just described has been used in less extended form by Lichtenberger<sup>6</sup> and by Shih,<sup>1</sup> who thus obtained the values of  $K_1$  (or rather of  $K_1/2$ ) on the assumption that  $K_2(\alpha_1^2\alpha_2^2\alpha_3^2)$  is negligible.

Magnetization curves were taken twice at

<sup>6</sup> F. Lichtenberger, Ann. d. Physik [5] **15**, 45-71 (1932).

each of three different temperatures in all cases, and in two cases single runs were taken at a still higher temperature. About sixteen points were obtained for each separate curve. Most of the curves are so nearly similar in appearance that graphical presentation on any reasonable scale would not be especially interesting. Instead of this the areas between the curves and the magnetization axis (up to  $I=I_{\text{sat}}$ ) have been carefully computed from faired curves drawn separately on a large scale (5 cm = 100 in  $I$  and in  $H$ ) and these results are collected in Table II. Saturation was easily attained in all cases and the applied field was generally carried about 300 oersted higher than necessary for saturation. The differences ( $W_{011}-W_{100}$ ) and ( $W_{111}-W_{100}$ ) which are used in computing  $K_1$  and  $K_2$  are believed to be correct to about  $\pm 0.02 \times (10)^4$  erg·cm<sup>-3</sup>. Inspection of the formulae used then shows that the corresponding uncertainty in  $K_1$  is about  $\pm 0.1 \times (10)^4$ , the uncertainty in  $K_2$  not less than  $\pm 1 \times (10)^4$ . The irregular variations of  $K_2$  with rise in temperature suggests that these limits are actually exceeded in some cases. Tabular values are carried to one more decimal place than their precision warrants.

As regards  $K_0$  the usual difficulty arises in estimating the demagnetizing factor,  $N$ , with the desired precision. It is encouraging to observe, however, that the same value of  $N$ , within a total range of about two percent, was obtained for each spheroid regardless of the direction chosen. A troublesome, though slight, variation of  $N$  with temperature, already noted by Kleis,<sup>2</sup> again appeared in these experiments, but there were now enough data to permit a decision as to its origin. There is little doubt that the apparent decrease in  $N$  as the temperature rises is due to a real decrease in the coercive force. Instead, therefore, of choosing  $N$  so that the whole magnetization curve up to  $I=I_{\text{sat}}/2$  nearly coincides with the line  $H=0$ , we must choose  $N$  so that the part of the magnetization curve just below  $I=I_{\text{sat}}/2$  is parallel to the line  $H=0$ . We then find this vertical part moving in toward the magnetization axis as the temperature rises, practically coinciding with it at temperatures just below the Curie point. The precision in measurement necessary to disclose this phenomenon in the present instance was

TABLE II. Results on ternary alloys.

Ni-Co-Fe	TEMP. °C	$I_{\text{sat}}$	ENERGY DENSITIES, erg. cm <sup>-3</sup> × (10) <sup>4</sup>					
			$W_{100}$	$W_{011}$	$W_{111}$	$K_0$	$K_1$	$K_2$
80-10-10	22	768	1.03	0.98	0.82	1.03	-0.20	-3.9
	200	710	0.65	0.60	0.51	0.65	-0.20	-2.0
	400	560	0.16	0.12	0.13	0.16	-0.16	0.6
70-20-10	516	359	0.14	0.06	0.11	0.14	-0.32	2.1
	22	872	2.32	1.64	1.46	2.34	-2.90	1.7
	200	822	1.69	1.11	1.13	1.71	-2.52	7.0
65-15-20	398	701	0.74	0.42	0.40	0.75	-1.38	2.9
	598	407	0.12	0.12	0.10	0.12	0.00	-0.6
	22	1005	1.29	1.51	1.18	1.29	0.91	-11.2
60-30-10	201	936	1.01	0.99	0.92	1.01	-0.08	-1.8
	398	808	0.30	0.22	0.20	0.30	-0.32	0.2
	22	974	3.29	2.38	1.80	3.33	-3.80	-8.2
60-25-15	203	926	2.03	1.61	1.30	2.05	-1.75	-5.0
	397	834	1.23	0.95	0.73	1.24	-1.17	-3.7
	22	1027	2.25	1.61	1.52	2.27	-2.64	3.4
50-40-10	201	971	1.55	1.30	1.05	1.56	-1.03	-4.5
	398	857	0.85	0.77	0.69	0.85	-0.33	-1.5
	22	1044	3.72	2.02	1.54	3.86	-7.21	-0.4
50-25-25	205	993	2.94	1.68	1.44	3.04	-5.43	4.1
	398	902	1.51	1.26	0.92	1.53	-0.87	-10.2
	22	1152	0.91	1.00	1.07	0.90	0.35	1.6
40-10-50	205	1083	0.60	0.69	0.72	0.59	0.38	0.2
	398	973	0.34	0.29	0.34	0.34	-0.25	2.2
	22	1325	1.30	2.80	2.72	1.29	6.10	-16.4
	200	1143	0.78	1.24	1.41	0.78	1.87	0.4
	398	682	0.36	0.54	0.39	0.36	0.74	-5.9

rather high, for the maximum coercive force thus disclosed was always less than 10 and the applied magnetizing field at half saturation was 300 or more. There is, accordingly, some variation in the apparent coercive force for different directions. Part of this may be systematic rather than accidental, but the precision of the data is hardly high enough to permit a decision. It may be mentioned that Sixtus<sup>7</sup> has reported higher coercive forces for directions of easier magnetization in silicon-iron crystals. No such correlation could be established in the present work.

Fig. 1 shows how the principal anisotropy constant,  $K_1$ , measured at room temperature, 20°C, varies over the range of compositions now available for comparison. This includes the ternary alloys here studied as well as the binary alloys of the same crystal structure dealt with by Shih<sup>1</sup> and Kleis.<sup>2</sup> The values for iron<sup>4</sup> and for iron-cobalt alloys<sup>8</sup> have been added for the sake of completeness. The crystal structure in this corner of the diagram is body-centered cubic. The cobalt corner has hexagonal structure. The boundaries of these regions, which are not well established, have been sketched in to agree with a diagram given by Kühlewein.<sup>9</sup> All points have

<sup>7</sup> K. J. Sixtus, Phys. Rev. 50, 395 (1936).

<sup>8</sup> J. W. Shih, Phys. Rev. 46, 139-142 (1934).

<sup>9</sup> H. Kühlewein, Wiss. Veröffentl. a.d. Siemens-Konzern 11 [1], 124-140 (1932).

TABLE III. Components and binary alloys. Computations from previous data.

Ni-Co-Fe, nominal	20°C		200°C		NOTE
	$K_1$	$K_2$	$K_1$	$K_2$	
100-00-00	- 3.44	5.3	0.50	2.0	(a)
97-03-00	- 1.00	0.9			(b)
90-10-00	1.57	- 3.8			(b)
90-00-10	- 0.70	- 2.3	-0.20	- 1.0	(c)
80-20-00	- 0.35	0.8			(b)
70-00-30	0.68	- 1.7	0.15	-0.4	(c)
65-00-35	1.47	- 7.0	1.22	-4.2	(c)
60-40-00	- 7.39	3.7			(b)
50-50-00	-10.80	- 4.0			(b)
50-00-50	3.33	-18.1	2.52	-8.3	(c)
35-65-00	-25.83	15.5			(b)
00-70-30	-43.30	5.3			(d)
00-50-50	- 6.80	-38.7			(d)
00-40-60	4.54	-11.1			(d)
00-30-70	10.21	16.2			(d)
00-00-100	42.1	15	30.0	22	(e)

Notes. (a) Honda, Masumoto and Shirakawa.<sup>10</sup> From original curves. Variations of  $K_1$  and  $K_2$  over a wide range in temperature have been considered. (b) Shih.<sup>1</sup> From original curves and tabulated values of  $W_{111}-W_{100}$ . (c) Kleis.<sup>2</sup> Values at 200°C interpolated between values given for 150°C and 300°C. (d) Shih.<sup>8</sup> From tables of  $W_{111}-W_{100}$  and  $W_{011}-W_{100}$ . (e) Piety.<sup>4</sup> Values at 200°C interpolated between values given for 18°C and 296°C.

been plotted at their intended compositions. The sign and magnitude of  $K_1$  are indicated by the direction and length of the vertical line drawn from each dot. The data for nickel have been recomputed from the curves given by Honda, Masumoto and Shirakawa.<sup>10</sup> The plotted values for boundary points are given in Table III and have all been corrected for errors in cutting where these are known. The extension of the lines for  $K_1=0$  into the ternary field are made in a manner consistent with the measured values, but there are too few points to make the location of these transitions very precise. In particular it cannot be decided whether the region of positive  $K_1$  is separated into two parts by an isthmus of negative  $K_1$ , or *vice versa*. The evidence is regarded as favoring the first of these alternatives. It should be noticed that the ternary alloys contained an appreciable amount of manganese, the binary alloys did not.

Fig. 1 also shows the values of  $K_1$  at or near

<sup>10</sup> K. Honda, H. Masumoto, Y. Shirakawa, Sci. Rep. Tohoku Imp. Univ. [1] 24, 391-410 (1935).

200°C. It will be noticed that the lines  $K_1=0$  pass over only one of the selected compositions as the temperature rises from 20° to 200°C. At 400°C the left-hand transition line has moved still more toward the left but the absolute magnitude of  $K_1$  has fallen at all points. It is probably significant that the best perm-invars lie in the region in which the anisotropy becomes very small at or above 400°C. (An open circle in the figure shows a preferred composition.) Elmen<sup>3</sup> has shown that the best of these alloys are greatly altered by baking at 450°C. The crystalline anisotropy must be so low at this temperature that magnetostrictive strains should largely control the directions in which domains can remain saturated.<sup>11</sup> Local short-circuiting of magnetic flux should be especially easy in these alloys at such temperatures.

It is suggestive that the sign of  $K_1$  seems to depend more upon proximity to pure iron or pure cobalt than upon the particular cubic structure of a ternary or binary alloy. One is tempted to connect the lines  $K_1=0$  across the phase boundary in Fig. 1.

The values of  $K_2$  given in Tables II and III show no such regularity as the values of  $K_1$  and in view of the low precision with which  $K_2$  has generally been measured it seems premature to discuss its variations. It should however be noted that the maximum possible values of  $(t_1-f_1)$  and  $(t_2-f_2)$  are, respectively, 1/3 and 1/27 so that the absolute values of  $K_2$  may be much larger than those of  $K_1$  without having nearly as much effect upon magnetic anisotropy.

In conclusion I wish to express my particular indebtedness to J. D. Kleis who postponed his own affairs to complete the necessary measurements, which he had undertaken as my research assistant. I am also obliged to J. W. Shih who sent me his paper on nickel-cobalt alloys<sup>1</sup> prior to its publication and thus led me to undertake this special study of the nickel-rich alloys of the ternary system.

<sup>11</sup> R. M. Bozorth, J. F. Dillinger, Physics 6, 285-291 (1935).