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Magnetic Anisotropy of Nickel-Cobalt Single Crystals

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The magnetic anisotropy of nickel-cobalt single crystals in the face-centered cubic range has been investigated with a pendulum magnetometer. The direction for easiest magnetization changes from <111> to <100> as the cobalt content increases from 3 to 10 percent and changes back to <111> when cobalt is more than 20 percent. Powell and Fowler's theory in regard to the relations between the direction of easiest magnetization and the crystalline structure, is again inadequate to explain these results, as in the case of iron-cobalt alloys. The anisotropy constants were calculated both from these monocrystalline specimens of Ni-Co alloys and from the polycrystalline specimens tested by H. Masumoto. The discrepancy between the two sets of values is too large to be accounted for by experimental errors in the present work.

In a previous investigation[1] on the magnetic properties of iron-cobalt crystals, it was found that the direction for the easiest magnetization in the body-centered cubic range of this system changes from the tetragonal axis at the iron end to the trigonal axis as the cobalt content increases. The transition occurs at about 42 percent cobalt. The results disagree with the theory of Powell and Fowler[2] concerning the deviation of the direction of magnetization from that of magnetizing field. In this paper, similar work on the nickel-cobalt system will be reported.

According to Powell and Fowler, in the case of a face-centered cubic lattice, the crystalline field, assumed to represent the interaction of the spin moment of electrons responsible for ferromagnetism and the orbital moment of the atom to which they belong, has a maximum lying in the tetragonal direction. With this assumption the trigonal axis becomes the direction for easiest magnetization. This has been found true for nickel.[3] It is of interest to see whether or not this idea holds for the nickel-cobalt system, which, from 0 to 70 percent[4] cobalt, has the same space-lattice as nickel.

Nickel was obtained from the International Nickel Company, New York. It contained 99 percent Ni, 0.3 percent Co and small quantities of iron, silicon and carbon. The cobalt used here was of the same kind as in the writer's previous work, namely, 99.3 percent pure. For the preparation of alloys and their monocrystals the slow solidification method was again adopted. The alloys were prepared, as before, in an induction furnace with moving coil. The alloy crystals of low cobalt content are more easily recrystallized in large sizes than those of high cobalt compositions. Ingots were first etched in nitric acid, of higher concentration for higher cobalt, and then in a concentrated solution of ferric chloride. Sometimes the mixture of these two solutions was more effective than either separately. The brightest etch planes were found to be of form {111}.

On account of the fact that these alloys were too soft to make exact oblate spheroids on a rapidly rotating shaft,[5] we modified the previous method for the preparation of specimens by sticking on the end of an iron rod of required diameter a thin plate which had been ground on an oil stone to a definite orientation and thickness, and filing it gently to shape without power drive. The resulting spheroid, which is not quite so perfect in shape as a lathe-filed specimen, was then slightly etched to remove the strained surface. After annealing in a hydrogen atmosphere at 900°C for three hours, the orientation was determined by the Laue method. From the shape of the Laue spots we know that the specimens obtained in this way are much more free from strain than those obtained by machining. Some constants of the specimens are given in Table I. Orientation is defined as before, a perfect $(01\bar{1})$ specimen having $\alpha=45°$, $\beta=0°$.

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

[2] F. C. Powell, Proc. Roy. Soc. **A130**, 167 (1930).

[3] S. Kaya, Sci. Rep. Tohoku Imp. Univ. [1] **17**, 639 (1928); W. Sucksmith, H. H. Potter and L. Broadway, Proc. Roy. Soc. **A117**, 471 (1928).

[4] H. Kühlewein, Physik. Zeits. **31**, 626 (1930); A. Osawa, Sci. Rep. Tohoku Imp. Univ. [1] **19**, 109 (1930).

[5] L. W. McKeehan, Rev. Sci. Inst. **5**, 265 (1934). We are informed that the original method has been modified so that softer crystals can now be handled by it, as will shortly be described by L. W. McKeehan, R. G. Piety and J. D. Kleis in this journal.

TABLE I. *Constants of crystal specimens.*

Percent Co by Synthesis	Density at room temp. g cm^{-3}	Volume cm^3	Thickness cm	Diameter cm	Demagnetizing factor N	$\alpha°$	$\beta°$
3	8.940	0.002972	0.0250	0.443	0.520	45	2
10	8.888	0.002778	0.0248	0.451	0.503	40	0
20	8.881	0.00166	0.0230	0.360*	0.580	45	0
40	8.785	0.00204	0.0210	0.376	0.515	45	10
50	8.790	0.00147	0.0203	0.339†	0.548	45	7
65	8.825	0.001873	0.0227	0.388	0.534	45	5

*For [111] and [011] only; along [100], dia. =0.358 cm and N =0.585.

† For [111] and [011] only; along [100], dia. =0.343 cm and N =0.543.

For magnetic measurement a pendulum magnetometer was used, of the same general design as before except for a few parts. The solenoid for producing uniform field was constructed with a length of about 25 cm instead of 20 cm. It produces a quite uniform field region sufficiently large so that the effect of its inhomogeneity on the displacement of specimen is inappreciable. The two coils for the nonuniform field were separated by $R\sqrt{3}$, where R is the mean radius of either, rather than by R as in the magnetometer first used. In order to eliminate external mechanical disturbances the moving part of the magnetometer on which the specimen rests was hung from a heavy and damped system suspended from a fixed support. With a field gradient of 2 oersteds, cm^{-1} and a magnification by the microprojector of about 740 times, a deflection of 20 cm or more may easily be obtained for almost all the specimens. The largest difference in deflection due to anisotropy is then about 6 cm. The probable error in the measurement is less than one percent, somewhat lower than in the previous work, the volume of the specimens being determined more accurately. The results are shown in Figs. 1 and 2. The saturation values of I here obtained are reasonably close to those previously reported by others.[6]

It is very interesting to note that the direction of easiest magnetization changes from <111> to <100> as the cobalt content increases from 3 to 10 percent, but when cobalt increases to more than 20 percent it changes back to <111>. The 20 percent alloy shows almost no difference in magnetization among the three principal axes. If its elementary crystals are easily magnetized in all directions, a polycrystalline material will be expected to have high permeability in low field. This is in agreement with Masumoto's findings.[6] By comparing the irreversible parts of the magnetization curves it is apparent that both the 40 and 50 percent Co alloys are magnetically harder than the others. This is probably due to the mechanical strain introduced in the process of preparation, since only these two specimens were made partly by machine and partly by hand. The strain is also evident from the shape of the Laue spots which are a little elongated in radial directions. In spite of this the I_p–H_e curves along the preferred direction show a rather sharp bend near saturation. This is the way a perfect crystal should behave according to the generally accepted view of the process of magnetization. For other directions we did not notice, beside the main bends, any small corners either on the uncorrected or on the corrected I–H curves as some investigators have reported both in pure ferromagnetic substances[7] and in alloys.[8]

The area enclosed between the magnetization curves for any two directions represents the energy difference of magnetization between these two directions. Due to the fact that the equatorial plane of the specimen is not in general

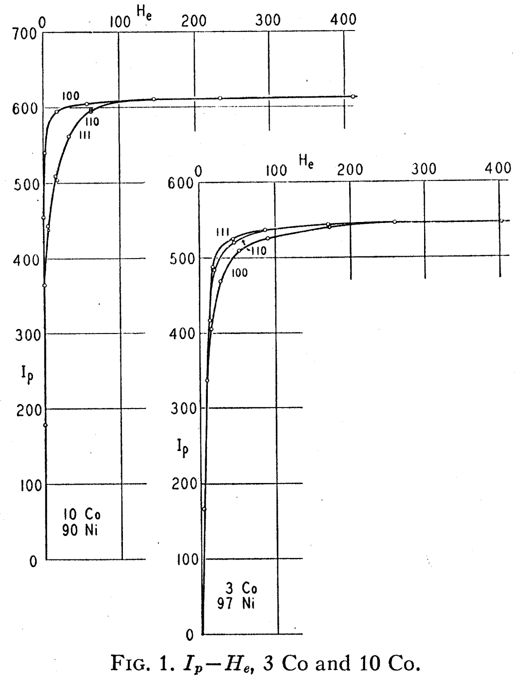

FIG. 1. $I_p - H_e$, 3 Co and 10 Co.

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[6] H. Masumoto. Sci. Rep. Tohoku Imp. Univ. [1] **16**, 321 (1927).

[7] G. J. Sizoo, Zeits. f. Physik **56**, 649; **57**, 106 (1992).

[8] F. Lichtenberger, Ann. d. Physik [5] **15**, 45 (1932).

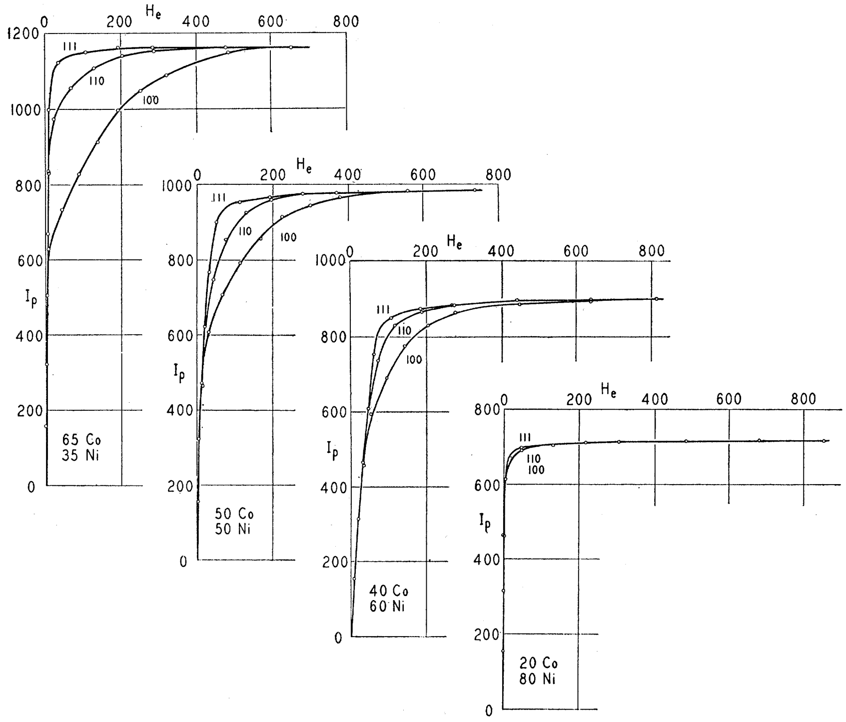

FIG. 2. I_p-H_e, 20 Co, 40 Co, 50 Co, 65 Co.

exactly parallel to the $(01\bar{1})$ plane the measured energy differences will generally be less than they should be. This nonparallelism can be corrected as we shall see later. The experimental and the corrected values of $W_{111}-W_{100}$ are given in Table II, while in Fig. 3 only the corrected values are plotted against the composition. It may be emphasized that the anisotropy constant for 65 percent Co has much the highest absolute value so far reported for face-centered cubic crystals. The value of $W_{111}-W_{100}$ for pure nickel in the first column was obtained by McKeehan[9] from Kaya's[3] measurements. From the figures we see that the transition of the easiest direction for magnetization from <111> to <100> probably occurs at about 5 percent and that from <100> to <111> at about 19.5 percent cobalt.

TABLE II. *Differences of energy of magnetization, orientation factors and anisotropy constant. Eq. (1)*

Percent Co	$W_{111}-W_{100}$ uncorrected 10^4 erg. cm^{-3}	Orientation factors $f_{111}-f_{100}$	Anisotropy constant K 10^4 erg. cm^{-3}	$W_{111}-W_{100}$ corrected 10^4 erg. cm^{-3}	K Polycrystals* 10^4 erg.cm^{-3}
Pure Ni	−1.67	—	−2.5	—	−5.5
3	−0.298	0.3325	−0.448	−0.298	—
10	+0.38	0.331	+0.574	+0.384	+4.4
20	$-0.087<x<0$	0.333	$-0.13<x<0$	$-0.089<x<0$	—
40	−2.08	0.299	−3.47	−2.32	−15.1
50	−3.54	0.314	−5.63	−3.75	−27.5
65	−7.80	0.320	−12.2	−8.14	—

* Data of H. Masumoto.[6]

Now let us consider whether Powell and Fowler's theory is valid in the light of these results. In general the direction dependent energy[10] of a cubic crystal may be expressed as

$$W=2K(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2)=2Kf, \qquad (1)$$

where the α's are the direction cosines of a magnetization vector with respect to the tetragonal axes, and K is the so-called anisotropy constant. In his notation, Powell's result may be reduced to the above form by letting

$$K=1/4NI\gamma, \qquad (2)$$

where

$$\left.\begin{aligned}\gamma&=5/2(G^4/N^4)\{1/2-5(p^4+q^4+r^4)/2\},\\ N&=3kT_c/(y+2)\mu_B,\quad p=\alpha_1,\ q=\alpha_2,\ r=\alpha_3.\end{aligned}\right\} \qquad (3)$$

The sign of K, and hence that of γ, depends upon the direction of easiest magnetization.

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[9] L. W. McKeehan, Tech. Publication No. 554 of the Am. Inst. Min. and Met. Eng., August 1934; Trans. A. I. M. E. **111**, 11 (1934).

[10] N. S. Akulov, Zeits. f. Physik **67**, 794 (1931).

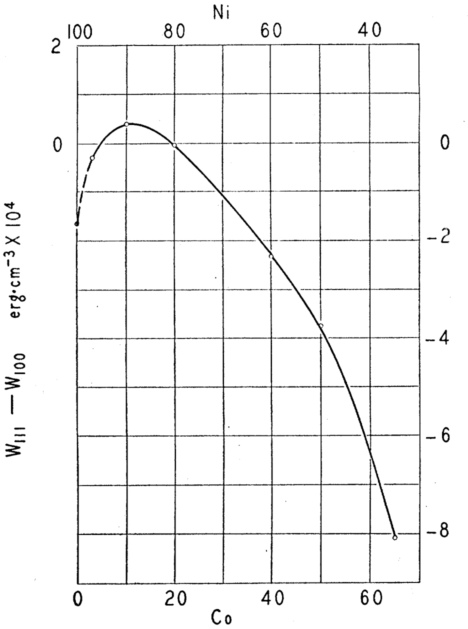

FIG. 3. $W_{111}-W_{100}$, nickel-cobalt single crystals.

If this direction is <111>, K is negative; if <100>, K is positive. This means whenever the direction for easiest magnetization changes, the sign of γ will reverse. In order to fit the experimental findings of the nickel-cobalt alloys the direction of the crystalline field, G, or rather the directions of its crystallographically equivalent maxima, must change twice, once at 5 percent Co and again at 19.5 percent Co. Suppose that the interaction between the electrons and the atoms (and hence the direction of maximum G) depends more or less upon the electron configuration inside the atoms of an elementary domain. It is hard to see why the electron pattern, which has been changed by the addition of a few cobalt atoms, should be restored to the original configuration when more cobalt is added.

Furthermore, for the alloys of the transition composition, the constant K must pass through zero. We do not know much about the nature of K. If it is represented by Eq. (2), then, when K becomes zero, either N or γ is equal to zero. Since N, which corresponds to Weiss's molecular field, seems necessarily to exist in all ferromagnetics and we cannot take special values for α_1, α_2, α_3, such as 1, 0, 0, or $1/\sqrt{2}$, $1/\sqrt{2}$, 0, to make the factor f in Eq. (1) equal to zero, G has to be zero. In other words the interaction of the orbital and the spin moments ceases in the transition alloys. The reason for this is again hardly conceivable. In brief, Powell and Fowler's theory is inadequate to explain the fact that the easiest direction for magnetization of a ferromagnetic changes in one and the same lattice structure.

As we have mentioned above, the energy difference of magnetization may be corrected for the imperfect cutting of specimens. This can be done by finding the orientation factors, f, of the component of magnetization, I_p, parallel to the direction under consideration. Having f_{111}-f_{100} and W_{111}-W_{100}, K may be computed. From the value of K, the true energy difference of magnetization for perfect cutting was calculated. These are given in the fifth column of Table II. The anisotropy constant may also be obtained from the I–H curve for polycrystalline substances by the following formula[11]

$$K=5A/2, \tag{4}$$

where

$$A=\int_0^{I_\infty} HdI. \tag{5}$$

From Masumoto's[6] data, K for pure nickel and for the 10, 40 and 50 percent Co alloys was thus evaluated. Evidently the discrepancies between these values and those from single crystal data are much too large to be ascribed to the experimental errors in the present work. It is to be noted that small differences in the demagnetizing factors of the rods used by Masumoto would account for large differences in A. Furthermore, no correction for the width of the hysteresis loop, which may be considerable for polycrystalline specimens with high cobalt content, has been made in reducing his data. The sign of this correction is such that it would reduce the absolute (negative) values of K for 40 and 50 percent Co. It should also be remembered that Messkin and Somin[11] assumed random orientation of microcrystals in deriving their formula for K, and this condition may not exist in actual rods.

In conclusion the author wishes to express his appreciation to Mr. S. T. Pan for valuable aid throughout the work.

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[11] W. S. Messkin and B. F. Somin, Zeits. f. Physik **98**, 610 (1936).