MAGNETIC PROPERTIES OF THIN FILMS OF FERROMAGNETIC METALS PRODUCED BY THE EVAPORATION METHOD

By A. J. Sorensen

Abstract

Magnetic properties of thin films of iron, nickel and cobalt.—The films were deposited by the evaporation method on tin or aluminum foil, and the magnetic properties studied by an induction method for fields up to 139 gauss. The thickness of the films varied from 20 to 300 millimicrons (mμ). The results are as follows. (1) The maximum value of the intensity of magnetization is of the same order of magnitude as for bulk metal. In the case of cobalt, however, it is evident from the curve that with higher fields values higher than any heretofore attained might be secured. The magnetization is independent of the crystal size and the thickness of the film. (2) The remanence is high for iron and cobalt. For nickel a low value was obtained, perhaps due to oxidation. The remanence depends on the crystal size but not on the thickness. (3) The coercive force is high for all three metals and changes abruptly to lower values as the thickness is increased beyond a critical value, which for iron, cobalt and nickel is about 55, 70 and 200 mμ respectively. The high values of the coercive force may be due to the minute size of the crystals in the films. (4) The results combined with Steinberg's curves for the Hall effect in films, tend to contradict Kundt's conclusion that the Hall e.m.f. is proportional to the intensity of magnetization.

THE present work is an investigation of the magnetic properties of thin films of iron, nickel and cobalt, similar to those which were investigated in regard to Hall effect by J. C. Steinberg[1] and which in some respects gave remarkable results.

Method and Apparatus

The method used was similar in principle to one used by Kaufmann and Meier,[2] in which the intensity of magnetization was obtained by measuring the change of flux through a search coil, which was situated in a constant magnetic field, when the magnetized specimen was removed from the search coil. The arrangement of the instruments is shown in Fig. 1.

To obtain the sensitiveness required by the very small amount of metal available (one continuous operation of the evaporating device

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[1] Steinberg, Phys. Rev. **21,** 22 (1923)

[2] Kaufmann and Meier, Phys. Zeit. **12,** 513 (1911)

yielding only about one milligram of metal), the scheme illustrated in Fig. 2 was adopted.

In Fig. 2, the switches S_1 and S_2 are both in series with the galvanometer and search coil. The rubber band R is under considerable tension and can be held in the position shown in Fig. 2 by the rod A which has a little metal ring slipped loosely over it, through which R passes. Before a reading is to be taken, the switches are set as shown in the figure and R is connected by a string to the specimen in the search coil. Then the operator closes S_1, A slides ahead and releases the ring retaining R in place, and R moves back toward S_2 and pulls the specimen out of the search coil. When R reaches S_2 it catches the handle (not shown in the figure) and opens the switch so that the circuit is again open. The length

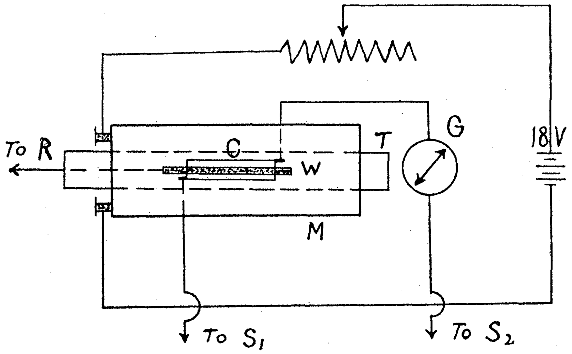

Fig. 1. Experimental arrangement.

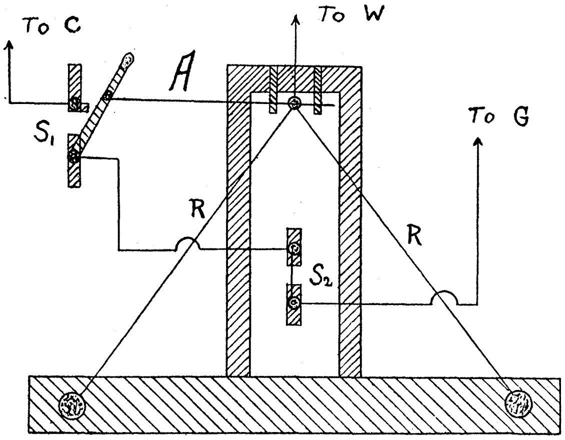

Fig. 2. Circuit controlling device.

of the time interval during which the circuit is closed can be varied by varying the position of S_2. Devices of a similar nature have been used by Wwedensky,[3] Gildemeister[4] and Wilson.[5]

In order to test the validity of the method and at the same time calibrate the set-up, the rod A in Fig. 2 was connected to the handle of a switch which was inserted in the circuit containing the magnetizing coil, so that small changes in the field through the search coil could be made and the corresponding deflections read. The deflections proved to be proportional to the deflecting currents and to be independent, within reasonable limits, of the tension on R and the position of S_2.

The deposits were made in a device described by Steinberg,[6] provided with a few minor changes, on tin or aluminum foil approximately 7.5 by 2.5 cm. The weights of the deposits were determined by weighing the foil before and after deposit, and the thicknesses were computed assuming the value of density in bulk. After this had been done, the foil was rolled upon a thin thread so as to form a uniform cylinder, 7.5 cm long and thin enough to be inserted in the search coil.

The iron used in this investigation was a sample of Armco iron, which, according to an analysis furnished by the makers (Page Steel and Wire Co., Bridgeport, Conn.) is 99.874 per cent pure. The nickel was furnished by Baker & Co. and contained, according to an accompanying analysis, 0.27 per cent iron, 0.05 per cent copper, and 0.004 per cent manganese. Cobalt wire was made by electrolytic deposition on a one mil tungsten wire by a method described by Langbein and Brannt in "Electrodeposition of Metals." These deposits presented a very bright surface but were exceedingly brittle so that great care had to be taken not to put any strain on them in inserting them in the evaporation device.

EXPERIMENTAL RESULTS

In Table I the results obtained for the three metals will be found. The following symbols are used:

δ = thickness of film in millimicrons ($m\mu$);
d_m = deflection for $H = 139$ gauss;
B_m = magnetic induction for $H = 139$ gauss;
B_r = remanence;
H_c = coercive force for a cycle in which the maximum value of the field was 139 gauss.

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[3] Wwedensky, Ann. der Phys. **66,** 110 (1921)

[4] Gildemeister, Ann. der Phys. **23,** 401 (1907)

[5] Wilson, Phys. Soc. Proc. **34,** 55 (1922)

[6] Steinberg, loc. cit.[1]

TABLE I. Experimental data.

Experimental data for iron, nickel and cobalt films.

	δ	d_m	B_m	B_r	H_c
Iron	14	16	16800	87 per cent	111
	26	34	19000	88	116
	32	45	20400	88	91
	39	66	23800	91	96
	40	63	22900		114
	55	78	22100	83	99
	59	108	26000	89	39
	74	125	24400	88	45
	98	148	22000	93	53
	150	222	21500	90	43
Nickel	51	6	1725	33 per cent	
	61	6	1460		105
	93	16	2530	55	77
	194	37	2710	54	75
	207	51	3280	59	56
	215	43	2830	53	61
	268	57	3070	56	62
	295	60	2890	58	55
Cobalt	18	22	16100	74 per cent	89
	22	32	19200	83	75
	25	33	17900	76	83
	36	40	14400	88	73
	63	78	16200	76	83
	77	91	16100	62	34

It will be seen that for each metal the films can be divided into two groups according to the coercive force. Fig. 3 represents the hysteresis curves obtained for one specimen from each group of iron films.

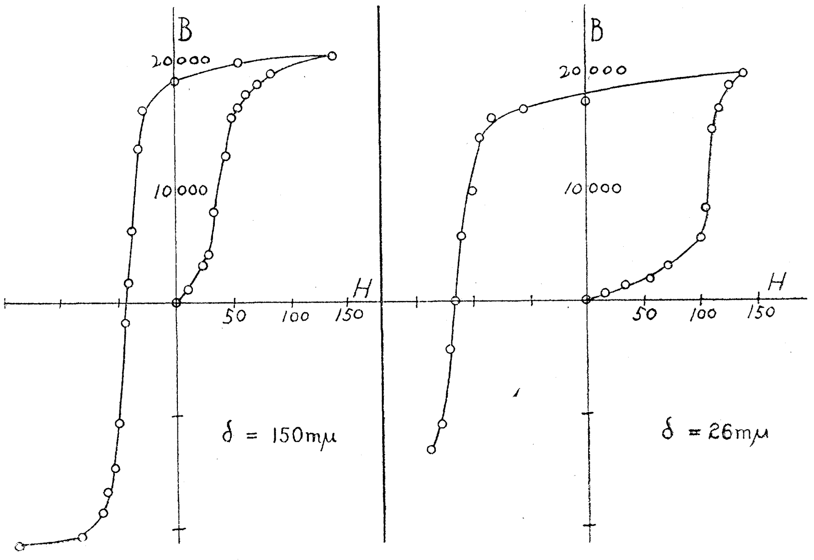

Fig. 3. Hysteresis curves for iron films.

It will be noticed that the thinner of the two films does not give a symmetric hysteresis curve. This is due to the fact that the maximum value of the field was not far enough beyond the coercive force to produce symmetry.

The lapse of time had very little influence on the magnetic properties of the films. While a slight decrease in permeability could be detected in the iron films four weeks after the first test was made, no change was detected in the remanence or coercive force of any of the three metals.

All of the cobalt films and some of the iron films acquired a magnetic moment during the process of deposition, which in some of the films amounted to as much as one-fourth of the moment due to the maximum field used in the tests, $H=139$ gauss.

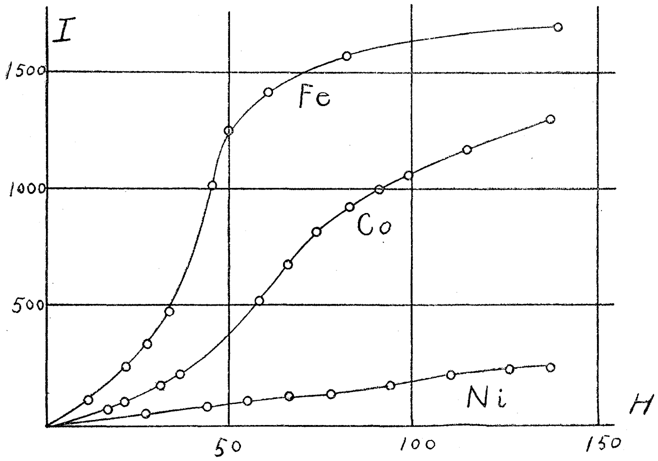

Fig. 4. Magnetization curves for films of iron, nickel and cobalt.

Discussion of the Results

In Table II and Fig. 4 are exhibited the most important results obtained.

Table II

Collected results for all metals.

	I_m	I_r	H_c
Fe......	1700	90 per cent	43
Ni......	234	56	62
Co......	1300	62	34

The values in the table are taken from the thicker groups of films in Table I. I_m and I_r are the maximum and the remanent values, respectively, of the intensity of magnetization.

In regard to the maximum value of I_m, the iron films appear not to be different from bulk iron. In the case of nickel I_m falls considerably below the accepted value for bulk metal (400, according to the Landolt-Börnstein tables). This is probably due, at least in part, to the very slow increase of I with the field. It may also be due, in part, to oxidation of the metal at the high temperature, which manifested itself during the process of deposition in a dull surface without metallic luster and which is largely responsible for the all too great irregularities in the B_m column of Table I.

In the cobalt films I_m reaches considerably higher values than are ordinarily given for bulk metal. Thus, Ewing[7] gives $I=800$ for $H=140$; Fleming, Ashton and Tomlinson[8] give 650 for $H=114$; Rowland[9] gives a still lower value. The samples used in these cases, however, contained considerable amounts of impurities. It therefore appears that cobalt is capable of a higher intensity of magnetization than has been heretofore attained. Moreover, the curve for cobalt in Fig. 4 indicates that still higher values might be obtained if stronger fields were applied.

In connection with the above results it should be noted that in work on thin films of electrolytic iron I_m has in all cases been found lower than for bulk iron.[10] Leick[10] investigated nickel and cobalt as well as iron and found for both approximately the same value as given for nickel in Table II. This low value for cobalt Leick explains as due to sponginess of the deposits.

The most remarkable result in Table II is the high value of the coercive force for all three metals. An explanation of this may perhaps be looked for in the crystalline structure of the films. It has been shown[11] that the size of the crystal granules in the metal has great influence on the specific resistance and the coercive force of iron but that the remanence and the maximum value of the magnetization are practically unaffected by changes in crystal size. Over a considerable range of crystal size the resistivity and the coercive force were both found to be linear functions of the number of crystals per unit length.

There is evidence[1] that the films are crystalline, but that the crystal granules are very small. Thus, Steinberg found evidence of crystal structure in iron films by an x-ray method, but failed to find any such

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[7] Ewing, Magnetic Induction in Iron and Other Metals.

[8] Fleming, Ashton and Tomlinson, Phil. Mag. **48**, 271 (1899)

[9] Rowland, Phil. Mag. **48**, 271 (1874)

[10] Kaufmann and Meier, loc. cit.[2];
Leick, Ann. der Phys. **58**, 691 (1896);
Schild, Ann. der Phys. **25**, 586 (1908)

[11] Thompson, Phil. Mag. **31**, 357 (1916)

structure under the microscope. This fact, together with the very high specific resistance of the films,[1] indicates that the crystal granules are very small. Therefore, according to the investigation of Thompson cited above, we should expect to find a high value for the coercive force.

The abrupt change in the coercive force at a certain thickness of film recalls the work of Maurain,[12] who found that when iron was deposited in a constant magnetic field, the magnetic moment of the deposit did not increase in proportion to the amount deposited until the thickness of the deposit was about 80 mμ. For nickel the same was found to be true, only in this case the critical thickness was 200 mμ. On the molecular field theory this is taken to mean that at those thicknesses the molecular field reaches a constant value, or, in other words, at those thicknesses the magnetic properties of the metal become definite. The present results may be taken to be in support of this theory in so far as a definite change is here found at a certain thickness, in one of the magnetic properties, namely, the coercivity. Since, however, the other magnetic properties do not change, the evidence is far from conclusive.

Assuming that the size of the crystals in the films is different from the size of those in bulk metal, the conclusions to be drawn are

(1) The maximum intensity of magnetization is independent of the size of the crystals and also of the thickness.

(2) The remanence depends on the crystal size but not on the thickness.

(3) The coercive force depends on the crystal size and also, in the way indicated above, on the thickness.

The above conclusions agree with the work of Thompson cited above, except in the case of the remanence, in which Thompson found no variation. This may have been due to the facts (1) that the crystals in his samples were very much larger than those in the films, and (2) that the range of sizes that he investigated was relatively small. It is interesting to note that the recent work on permalloy[13] and silicon steel[14] bears out the above conclusions.

On the basis of the results of an investigation[15] in which Kundt proved the Hall e.m.f. in ferromagnetic metals to be proportional to the rotation of the plane of polarization, and the results of an investigation by Du Bois,[16] in which the magnetic rotation of thin films of ferromagnetic metals was shown to be proportional to the intensity of magnetization

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[12] Maurain, Jour. de Phys. **10,** 123 (1901)

[13] Arnold and Elmen, Bell Technical Journal, July 1923.

[14] Yensen, Am. Inst. Elec. Eng., May and June, 1924.

[15] Kundt, Ann. der Phys. **49**, 257 (1893)

[16] Du Bois, Ann. der Phys. **31,** 941 (1887)

of the metals in bulk, Kundt drew the conclusion that the Hall e.m.f. is also proportional to the intensity of magnetization. If the Hall e.m.f. is plotted against the external field into which the experimental material is introduced, the curve rises rapidly at first as the field increases to a value roughly equal to 4π times the maximum value of the intensity of magnetization of the bulk metal. From this point on the curve rises only slightly as the field is further increased.

A. W. Smith[17] confirmed Kundt's results; however, in extending the investigation to higher temperatures he found the permeability and the Hall e.m.f. to vary in different ways with the temperature. Hence, the Hall e.m.f. cannot be expressed at various temperatures in terms of the permeability alone.

Steinberg[1] studied the Hall effect in thin films of evaporated iron and, in plotting the Hall e.m.f. against the external field, obtained a curve of the same general shape as those obtained by Kundt and Smith. However, the bend in the curve occurred when the external field had reached a value of 10,000 gauss, and the curve rose only slightly as the field was further increased. It was therefore inferred that the maximum value of the intensity of magnetization in the films was given, approximately, by

$$4\pi I_m = 10{,}000$$

so that I_m would be only about one-half of the value for bulk iron.

It will be seen that the present results do not support this conclusion. However, it must be borne in mind that in the study of the Hall effect the field is at right angles to the plane of the film, while the magnetic properties were measured in a direction parallel to it. Until the magnetic properties of the films have been studied at right angles to the film, the question can hardly be regarded as settled. However, until then the present results at least leave a doubt as to the proportionality between the Hall e.m.f. and the intensity of magnetization, even at room temperature.

In conclusion, the writer wishes to thank the members of the Department of Physics at the State University of Iowa for their assistance and interest, and especially Professor G. W. Stewart, under whose direction the work was carried on.

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[17] A. W. Smith, Phys. Rev. **30**, 1 (1910)