THE MAGNETIC BEHAVIOR OF NICKEL AND IRON FILMS CONDENSED IN VACUUM UPON VARIOUS METAL BACKINGS*

By J. H. Howey
Physics Department, Yale University

(Received October 28, 1929)**

Abstract

The magnetic nature of evaporated iron and nickel films deposited on various kinds of backings was investigated to determine the effect of the two-dimensional strain which the backing must impose upon the film due to differential thermal contraction. The films were, of necessity, deposited at elevated temperatures and measured at room temperature. The temperature of deposit was varied. Nickel samples deposited on backings which place the film under tension were found to be extremely hard, magnetically, while compression made the film comparatively soft. The magnetic intensity of iron samples in general was not changed markedly by these stresses. All the samples were harder than bulk metal. The method used to describe the results shows that films deposited at higher temperatures are more like annealed bulk metal, rather than less so as others have concluded. Extreme precautions were taken to deposit the films under gas free conditions so that it was possible to be sure that the abnormal hardness of films deposited at ordinary temperatures was due to the different manner in which the metal condenses and not due to the presence of gas. A novel arrangement of the magnetizing coil of a sensitive astatic magnetometer used for the magnetic measurements is described.

Introduction

THE work of Edwards[1] and Miller[2] has shown that films of iron and nickel deposited upon aluminum foil by evaporation from a hot wire are very hard magnetically as compared to these metals in the usual form. The hardness was found to depend upon the temperature at which the films were deposited. Both of these workers interpreted their results to mean that the films deposited at the higher temperature were more abnormally hard, but it would be surprising if that were true in view of the usual effect of annealing on the magnetic behavior of bulk metals. All their films were deposited on aluminum backings at elevated temperatures (100° and 200°C) and were measured at room temperature. A simple calculation would lead one to expect that, under these circumstances, the nature of the backing might influence the magnetic behavior of the film by virtue of the strain in two dimensions which the backing imposes upon the film as the two contract at different rates in cooling from the temperature at which the deposit was made. The work here described was undertaken to determine to what extent the magnetic behavior of films deposited on various metals was affected

* Part of a dissertation presented for the degree of Doctor of Philosophy at Yale University.
** This paper was completed August 1, 1929.
[1] R. L. Edwards, Phys. Rev. **29,** 321–331 (1927).
[2] K. J. Miller, Phys. Rev. **32,** 689–690 (1928).

by these certainly existing strains. The temperature of deposit was extended over as wide a range as feasible and some of the samples were measured at a temperature of 100° C as well as at room temperature so that the temperature at the time of measurement would approach the temperature at which the film had been deposited.

Production of Films

The evaporating tube in which the films were deposited was designed to make it posssible to secure far better vacuum conditions than previous workers have obtained. This was desirable in order to be certain that the abnormal properties of the films could not be attributed to the presence of gas at the time of deposit. The tube was 4.5 cm in diameter and 25 cm long, and was made entirely of Pyrex glass without any waxed or greased joints. This not only eliminated undesirable vapors in the interior of the tube but also made it possible to bake out the entire tube at a temperature of 500° C by surrounding it with a furnace. A filament (iron or nickel wire) 12 cm long was stretched lengthwise through the center of the tube. Flat strips of various metals 1 cm wide and 6.5 cm long were arranged symmetrically about the filament so that the metal evaporating from the electrically heated filament would condense upon the inner face of the strips. The ends of the strips were covered so that the actual length of the ferromagnetic sample deposited upon the strip was 4.5 cm. Three samples were thus deposited simultaneously from one filament. A slotted metal cylinder surrounding the filament was provided to cover up the metal strips until conditons were suitable and steady for beginning the deposit at which time the shield was turned by an outside magnetic control so that the strips were exposed to the filament.

Access to the interior of the tube was secured by cracking the glass completely around at one end of the tube. After new strips were fixed in place, the tube was fused together again, and the tube was then connected to a vacuum system having a two-stage mercury diffusion pump and a liquid air trap. By thoroughly baking the entire tube out before each deposit, it was possible to keep the pressure so low that it would not register on a McLeod gauge at the time of depositing the film, indicating a pressure of the order of 10^{-6} mm of mercury. The strips upon which the films were deposited were heated by radiation from the filament and the temperature at the time of deposit was varied by using filaments of different designs. The temperature of the strips for a given type of filament was determined by a thermocouple.

Films of nickel and iron were deposited upon backings of aluminum, copper, platinum, and molybdenum. These particular metals were chosen because of the range of the values of their coefficients of thermal expansion which are listed in the first column of Table I. In the second column are given the calculated values of the strain imposed upon the film by the backing per 100° C fall in temperature, assuming that the film was deposited upon the backing under no strain at a high temperature and allowed to cool. A negative sign indicates that the film will be subjected to a compressive

TABLE I

	Linear coefficient of expansion per 1°C	Strain per 100°C in nickel film	Stress to produce equal strain in nickel wire (dynes/cm²)
Ni	12.8×10^{-6}		
Fe	11.7		
Al	23.0	-10.2×10^{-4}	-22.6×10^{8}
Cu	16.6	-3.8×10^{-4}	-7.7×10^{8}
Pt	8.9	$+3.9\times10^{-4}$	$+7.9\times10^{8}$
Mo	4.0	$+8.8\times10^{-4}$	$+17.8\times10^{8}$
Tensile strength of Ni Wire 53.0×10^{8} dynes/cm²			

stress, while a positive sign indicates tension. The stresses required to produce equal linear strains in a nickel wire are given in the third column for comparison.

MAGNETIC MEASUREMENTS

Before measurement, the films were demagnetized by placing them in an alternating field which was decreased from a value of 300 gauss to zero. Magnetization curves were then obtained by a magnetometric method.

A high sensitivity magnetometer was constructed and used to make the magnetic measurements. It was of the null astatic type developed by Bozorth[3] and the reader is referred to his paper for an extended discussion of this type of magnetometer. As in the instrument described by him the specimen was placed beside the astatic pair, but a new arrangement of the magnetizing coil was used. The astatic pair and the specimen close beside it were arranged in the interior of a large vertical solenoid in such a way that the astatic pair was at the geometrical center of the solenoid while the suspension of the astatic pair coincided with the axis of the coil from the center up. Since the field at the center of the solenoid was uniform it exerted no torque on the astatic pair for any value from zero to the maximum field used to magnetize the specimen. This arrangement made it possible to place the specimen closer to the astatic pair which gave higher sensitivity and allowed an abundance of room for manipulating the specimens without deforming them in any way. It also served to decrease the zero shift which appears in this type of magnetometer due to the motion of the wires of the magnetizing coil expanding as the current heats them up, because the wires are farther removed from the astatic pair. Since the magnets of the astatic pair were to be used in the transverse field of the magnetizing solenoid, they were made of K. S. magnet steel and were strongly magnetized in a longitudinal field of 7000 gauss. They were matched by grinding on a fine carborundum wheel so that their longitudinal magnetic moments were equal after each had been placed in a transverse field of 200 gauss and subjected to enough reversals of the field to reach a steady state. In that condition, fifty reversals of the transverse field would produce no observable change in the remaining longitudinal moment, regardless of the final polarity of the transverse field.

In practice the zero position of the astatic pair was not entirely independent of the magnitude of the magnetizing field for fields above 100 gauss,

[3] R. M. Bozorth, Jour. Opt. Soc. Amer. **10**, 591–598 (1925).

so that a correction graph had to be plotted. The magnetometer gave a normal shaped curve for a sample of No. 40 nickel wire and from that curve the calibration constant of the magnetometer was determined. While such a determination was approximate and somewhat arbitrary, it was satisfactory since only comparative values were of importance in this work.

Most of the films measured were between 100 and 300mμ in thickness. The thickness was usually determined by weighing but in some cases the saturation value of the magnetic intensity was used to determine the thickness. The probable error in the determination of the thickness of such thin films was in the neighborhood of seven percent but this uncertainty was tolerable because conclusions could be drawn from comparisons of the shapes of the various curves rather than from numerical values of the intensity of magnetization at corresponding field intensities.

Results and Discussion

The results for nickel films will be considered first. In all figures T stands for the temperature of deposit, D for the thickness of the film, H for the magnetizing field in gauss, and I for the magnetic intensity in gauss. In Fig. 1 the full line curves are magnetization curves for three samples deposited upon backings of aluminum, copper, and molybdenum as indicated by

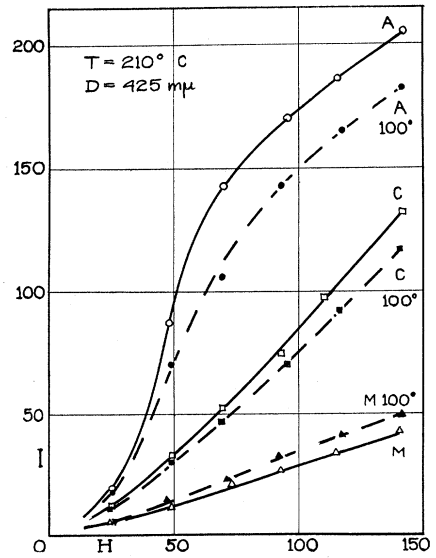

Fig. 1. Magnetization curves of nickel films on various backings measured at room temperature and at 100°C.

the letters accompanying the curves. They were deposited at a temperature of 210° C and measured at room temperature. If the effect of the stress on the magnetic nature of the sample increases in magnitude as the stress increases and depends in sign upon the sign of the stress as we may expect from the known properties of bulk nickel, then the curve for a sample which was under no stress due to differential thermal contraction should, according

to Table I, lie between the C and M curves. The dashed curves are the magnetization curves taken with the sample at a temperature near the boiling temperature of water, provision having been made to heat the sample by means of a flow of steam while it was in the magnetometer. At that temperature the stresses on the films should be partially relieved. The relative positions of the curves indicate that if the stresses were removed entirely the curves for all three samples would nearly conicide and lie between the C and M curves. The results for nickel films deposited at various other temperatures and measured at room temperature are shown in Fig. 2. The relative position of the curve P, Fig. 2c, which was obtained from a sample deposited upon a platinum backing gives further evidence that relative

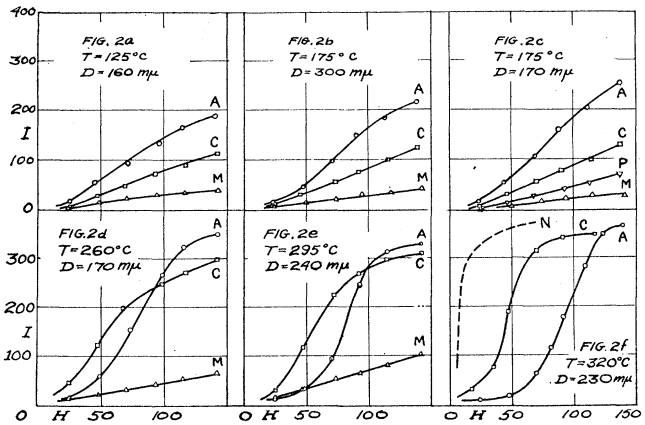

Fig. 2. Magnetization curves of nickel films deposited at various temperatures upon backings of aluminum, copper, platinum, and molybdenum.

thermal contraction is the determining factor in the influence of the backing. It can be seen that the relative effects of the backing upon the magnetic hardness of the film are in complete qualitative agreement with the relative stresses exerted by the various backings for all four materials used as backings.

It will be observed in Figs. 2d, 2e, and 2f that, while the curve A for the sample on aluminum reaches saturation at a lower field than does the curve C for the corresponding sample on copper, in general agreement with the other samples, still the first part of the curve A lies decidedly underneath the curve C. The peculiar square shape of the curves for samples on aluminum which is found when the temperature of deposit is high is probably due to some result of the compression other than a simple uniform strain in the direction of the stress, such as a buckling of the film due to imperfect adhesion. In general one may speculate that any such effect would be greater the thinner the film, whereas the effect of a uniform strain in the direction of the stress, that is in the plane of the film, would not vary much with thickness. On this hypothesis, the curves shown in Fig. 3 may be taken to verify the fact that the films on copper are in a state of comparatively simple strain while those on aluminum, deposited at this high temperature, have developed some complexity in their strain, either due to the excessive compression to which they are subjected or to less perfect adhesion between the film and its

backing. Without digressing farther on this point we may at least conclude that for moderate temperatures of deposit, and therefore for moderate stresses, a film of nickel which is deposited on a backing which places the film under tension as it cools will be extremely hard magnetically, while one deposited upon a backing that places it under compression will be comparatively soft. This is analogous to the effect of a one-dimensional stress in the direction of the magnetic field in the case of bulk nickel. There tension makes the nickel much harder to magnetize while compression favors magnetization.

The influence of the temperature of deposit is obvious from a comparison of the curves for films deposited at various temperatures with the curve *N*, Figure 2f, which is given by Ewing[4] for bulk nickel. The samples deposi-

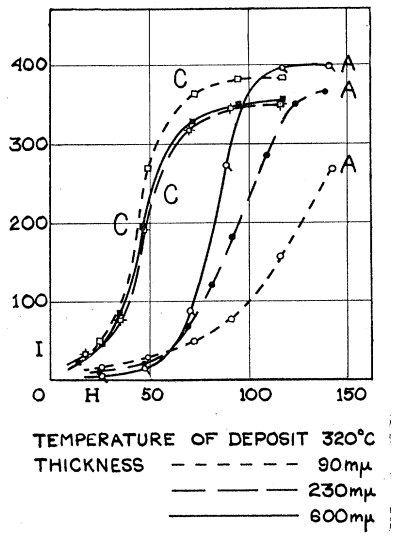

Fig. 3. Peculiar effect of thickness on the magnetic nature of nickel films deposited upon aluminum backings at a high temperature.

ted at higher temperatures are clearly more normal in their magnetic behavior, that is they are more like annealed bulk metal rather than less so as Edwards and Miller concluded. These previous investigators made use of hysteresis loops instead of magnetization curves to show the magnetic state of their samples, and based their conclusion upon the numerical values of the coercive forces and upon the areas of the loops. In so doing they overlooked the fact that the nature of the hysteresis loop may be taken as a distinguishing characteristic of a sample only when the maximum field applied is large enough to produce approximate saturation. Otherwise the nature of the loop depends markedly upon the maximum field applied. A comparison of the hysteresis loops given in Fig. 4 with the magnetization curves for the same three samples given in Fig. 1 shows that with a limited magnetizing field and hard samples there is no logical correlation between large coercive force and great magnetic hardness. The results of Edwards and Miller, interpreted with this in mind are not in disagreement with the

[4] J. A. Ewing, "Magnetic Induction in Iron and Other Metals," 3rd edition (1900), p. 87.

results found here where a wider range of temperatures of deposit has been used.

The results of the measurements made on iron films are shown in Fig. 5. In Fig. 5a the curves *F-h* and *F-s* are typical magnetization curves for hard and soft iron respectively, taken from Ewing.[5] The other symbols have

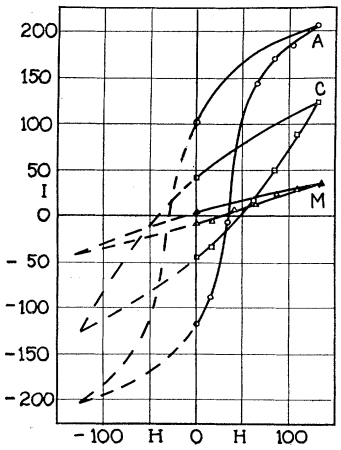

Fig. 4. Hysteresis loops corresponding to the magnetization curves given in Fig. 1.

the same significance as in the previous figures. It is obvious from these curves that the iron films which were deposited at higher temperatures were softer and more like bulk metal in their magnetic behavior as was the case with nickel, but here there is no positive consistent evidence as to the way

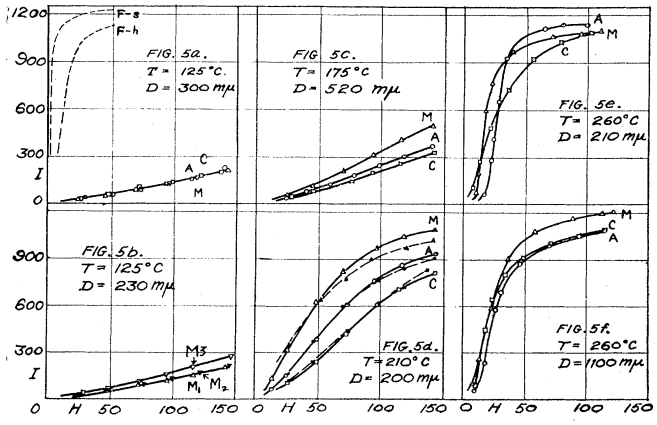

Fig. 5. Magnetization curves of iron films deposited at different temperatures on various metal backings.

in which the nature of the film depends ugon the material of the backing. The broken line curves of Fig. 5d are the magnetization curves of the same three samples for which the full lines obtain at room temperature, taken with the samples at a temperature of approximately 100°C. Contrasting the effect

[5] Reference 4, p. 81.

of stress on the magnetic properties of bulk iron with corresponding effect for nickel we see that this result is not surprising. For iron, the magnitude of the effect is less and there exists the Villari reversal point somewhere along the curve where the effect changes sign. This point may lie almost anywhere along the curve depending upon the amount of stress applied and upon the hardness of the unstressed sample. The samples here compared varied widely in either one or both of these respects.

The difference between the results for the two metals makes it impossible to attribute the decided influence of the material of the backing observed in the case of nickel to the chemical nature of the backing metal. Such chemical differences as exist would presumably affect films of both metals in the same way since they are chemically much alike. It is also impossible to note any correlation between these results and the relative lattice spacings in crystals of the metals involved. It seems logical to conclude that the differences observed with different backings, large in nickel films and small in iron films, are almost exclusively due to differential thermal expansions and that the general hardening as compared with bulk metal in both nickel and iron is due to something else.

It can be seen from these measurements that unless films of iron and nickel are deposited at temperatures considerably above 100° C they are so abnormally hard that the field strengths ordinarily available in magnetometers will not permit tracing much of their magnetization curves. To make certain that this abnormality in films deposited at low temperatures was not due to a surface layer of gas which persisted through the ordinary baking out, the evaporation tube was altered so that one of the strips could be brought to a red heat by electronic bombardment in addition to the usual baking out of the entire tube. It can be seen that the sample of iron deposited immediately thereafter upon this backing (curve *M*3, Fig. 5b) does not differ much from other samples deposited at the same temperature showing that all the backings were so well baked out that what gas remained there had little effect upon the results. As a further test for influence of surface conditions film *M*2 was deposited upon a backing of molybdenum with an etched instead of the usual polished surface.

Without going far afield to discuss the possible mechanism of the influence of the temperature of deposit upon general magnetic softness, which is undoubtedly connected with the relative perfection of crystal structure, as others have suggested, it is of importance to note that this range of depositing temperatures, which was wide enough to cause extreme difference in the magnetic behavior of the samples and presumably in their crystal structure, was secured merely by regulating the energy input of the filament, and without any special provision for heating or cooling the samples. It seems likely that some of the apparent disagreement as to whether films condensed from vapor are well crystallized or not[6] would disappear if the temperature of the surface at the time of their deposit had been recorded.

The writer is deeply indebted to the faculty of the Physics department of Yale University, particularly to Professor L. W. McKeehan who suggested the problem.

[6] Sophie Dembińska, Zeits. f. Physik **54,** 46–52 (1929).