

## HALL EFFECT AND SPECIFIC RESISTANCE OF EVAPORATED FILMS OF SILVER, COPPER AND IRON.

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### ABSTRACT.

**Specific resistance and Hall coefficient of evaporated films, 20 to 200  $\mu\mu$  thick.**—The specific resistances of films of *silver, copper and iron* were found to be from 15 to 57 times the values for the bulk metals, being respectively 24.3, 29.7 and 506 instead of 1.6, 1.8 and 8.8. The Hall coefficients of films of silver and copper, two diamagnetic metals, were respectively 41 and 11 per cent. *less* than the bulk values, while for the iron films the coefficient was +42.5 as compared with +6.6 for bulk iron, an *increase* of over 500 per cent. For the silver and copper films the Hall effect is proportional to the field throughout, but for iron films the proportionality extends only to 10,000 gaussses, whereas the limit for bulk iron is 20,000 gaussses. This indicates that the *maximum intensity of magnetization in iron films* is only half as great as that attainable in pure bulk iron.

**Structure of evaporated metal films.**—X-ray analysis showed that films of silver and iron and probably other metals have the same crystal structure as the bulk metals, but the crystal grains are too fine to distinguish with a microscope.

**Effect of extreme fineness of crystal structure on certain physical properties of metals.**—The increase of *specific resistance* is probably due to the multiplication of the number of contacts between grains. The *Hall coefficient* is practically independent of the structure except as it affects the magnetic condition of the crystals. The *intensity of magnetization* in iron crystals seems to be limited by the demagnetizing action of the poles of the crystals, which is greater the smaller the crystal grains.

**Method of producing uniform metallic films by evaporation.**—This involves moving a heated filament uniformly over the surface to be coated, at a distance less than the mean free path in a high vacuum. The films are hard and bright and resist corrosion.

### INTRODUCTION.

THIS work extends an investigation on chemically deposited silver films by G. R. Wait<sup>1</sup> to films obtained by evaporation. For a discussion of the various film phenomena, and references of historical interest, the reader is referred to Wait's publication. Wait concludes that his results as well as the results of others are consistent with the conception that the film consists of granules, each granule having the properties of the bulk metal.

Evaporated films were employed in order to learn more of the nature of these granules. The Hall effect and the specific resistance were measured in films whose thicknesses varied from 20 to 200  $\mu\mu$ . The

<sup>1</sup>G. R. Wait, "Hall Effect and Specific Resistance of Silver Films," *PHYS. REV.*, 19, 615, 1922.

films were examined under a microscope for traces of structure, and also subjected to x-ray analysis.

#### EXPERIMENTAL METHOD.

The essential features of the evaporation method of obtaining a film consist in drawing an electrically heated filament of the metal back and forth over a microscope slide in vacuum. Both glass and quartz slides were used. The particles of metal given off by the hot filament adhere to the microscope slide, giving a uniform film.

The details of the evaporation apparatus are shown in Fig. 1. Two

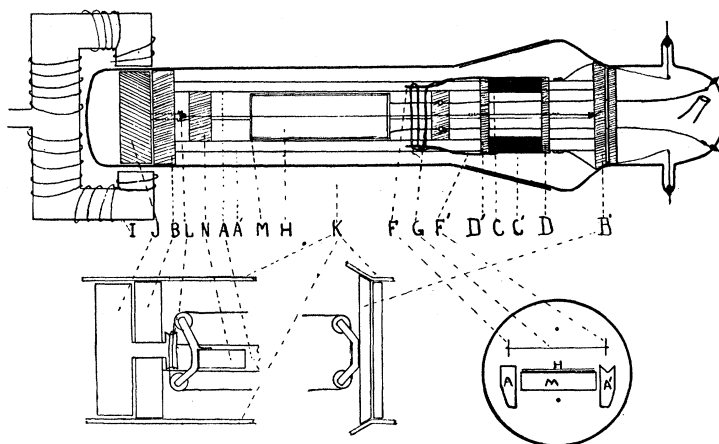


Fig. 1.

brass tracks  $AA'$  are fitted into the cylindrical fibre plugs  $BB'$ . Ground to fit the tracks are two brass blocks  $CC'$ , which, held rigid by the fibre straps  $DD'$ , constitute the carriage. The mica strips  $FF'$  serve to support the wire  $G$  connected across the blocks. The electro-magnet  $I$  turns around the pyrex tube  $K$ . Inside the tube, the iron core  $J$  turns with the magnet. By means of a small capstan  $L$ , the carriage is drawn along the tracks. The electro-magnet, which was designed by Dr. G. R. Wait and Mr. J. B. Dempster, is so constructed that it reverses its direction of rotation every three revolutions, and turns with constant angular velocity. Thus the carriage moves uniformly back and forth along the tracks.  $M$  is a small heating element to drive occluded vapors from the microscope slide  $H$ , prior to evaporation. Current is furnished the wire by means of leads connected to the brass tracks.

The tube was connected to a Knipp mercury vapor pump, with a Gaede mercury pump and a water aspirator as fore-pumps. The pumps which were very rapid were kept in operation during evaporation. Pre-

cautions were taken to insure the pressure being reduced to such a value that the maximum distance of the wire from the glass slide would be less than the mean free path of the gas molecules. The required pressure was .001 mm. mercury. As a rough indication, the pressure was reduced sufficiently to enable a spark to jump a one and one half inch air gap in parallel with the electrodes of an attached discharge tube.

It was found necessary to give a new wire a preliminary evaporation before desirable films could be obtained. In practice the wire was maintained at its evaporation temperature several hours before taking a film. When this precaution was not observed, the films obtained were found to be erratic in behavior and not constant with time. Etching the wire prior to evaporation did not eliminate the effect to any marked extent. Metals of very good purity were used, and the evaporated metals were probably very pure.

For microscopic examination a Bausch and Lomb metallurgical microscope was employed. X-ray analysis was made by means of a method described by Hull.<sup>1</sup> The apparatus used for the work was kindly loaned by Dr. C. W. Hewlett, and a description of its method of operation may be found in his publication.<sup>2</sup> The metals were evaporated under the same conditions as the films, and scraped from the surfaces upon which they were deposited. They were then finely powdered, and suitably mounted in a homogeneous beam of x-rays from a molybdenum target. A filter of zirconium powder .037 cm. thick reduced the spectrum to approximately a single line, of wave-length .712 A.U. The angular deviations of the lines were determined by observing the rate of discharge of an ionization chamber.

Hall effect and specific resistance were measured in a manner similar to that described by Wait.<sup>3</sup> Thickness was determined by weighing, assuming the density to be that of the bulk metal. This assumption is justified on the grounds that any sensible change in density would not account quantitatively for the magnitude of the observed effects.

#### RESULTS.

The Hall effect in a film is given by the relation,

$$E = \frac{RHI}{\epsilon},$$

where  $E$  is the Hall e.m.f.,  $R$ , the Hall coefficient,  $H$  the magnetic field,  $I$  the current through the film, and  $\epsilon$  the thickness of the film.  $E$ ,  $H$ ,  $I$ ,

<sup>1</sup> PHYS. REV., 10, p. 661, 1917.

<sup>2</sup> C. W. Hewlett, PHYS. REV., 20, 688, 1922.

<sup>3</sup> Wait, loc. cit.

and  $\epsilon$  are determined experimentally and  $R$  computed. The values given in Table I. were obtained during the investigation when not otherwise indicated.

TABLE I.

Metal.	$R \times 10^5$ (e.m.u.).	$\rho \times 10^{-3}$ (e.m.u.).
Taken from		
Bulk Silver (pure)—Smithsonian Tables.....	-88.0	1.6
Chem. Dep. Silver.....	-86.4	2.7
Evaporated Silver.....	-52.0	24.3
Bulk Copper (pure)—Smithsonian Tables.....	-56.0	1.8
Evaporated Copper.....	-50.0	29.7
Bulk Iron (pure)—A. W. Smith <sup>1</sup> .....	+ 660	8.8
Evaporated Iron.....	+4250	506.4

*Evaporated Silver Films.*

The Hall coefficient in evaporated silver is negative and independent of the thickness. The value given in Table I. is the average of the values obtained from a large number of films whose thicknesses varied from 20 to 200  $\mu\mu$ . In Fig. 2, experimental values of  $E/HI$  are plotted against  $\epsilon$ . The curve drawn through the points is a graph of the equation  $E/HI = R/\epsilon$ , where  $R$  has the average value obtained from the films.

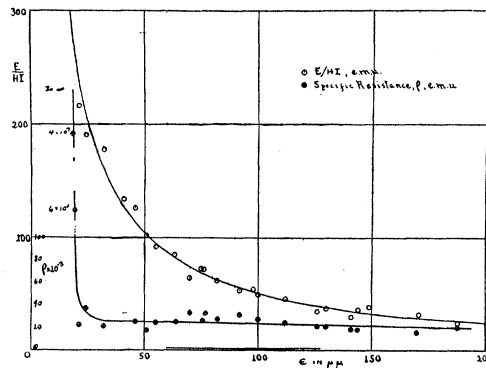


Fig. 2.

The specific resistance  $\rho$  as a function of thickness is also shown in Fig. 2. Its behavior is similar to that found in films obtained by other methods. The average value, taken from films whose thicknesses are greater than the critical thickness, *i.e.*, the thickness at which  $\rho$  becomes infinite, is given in Table I. It is to be noted that we have here a large increase in  $\rho$  as compared with bulk silver, about 1,000 per cent., and a relatively small change in the Hall coefficient. The straight line in

<sup>1</sup> PHYS. REV., 30, p. 1, 1910.

Fig. 2, near the axis of abscissas, indicates the value of  $\rho$  for bulk silver. The resistances of the films remained sensibly constant with time.

The x-ray analysis was only qualitative, it being desired merely to determine whether or not the metal was crystalline. From data published by Hull,<sup>1</sup> the angular deviations of the most intense lines were computed, and found to coincide within the error of the experiment, to the lines observed with evaporated silver. The existence of the lines in their computed positions was taken as sufficient evidence that the metal was crystalline, and of the type found in bulk silver, that is, with the face-centered cubic lattice, of elementary side 4.06 A.U.

The films obtained were hard and very uniform in thickness. They possessed an excellent reflecting surface, and did not deteriorate readily with age. Repeated attempts to observe granular structure under a microscope failed. The films appeared blue by transmitted light, which is the characteristic color of silver.

#### *Evaporated Copper Films.*

Evaporated copper was found to behave in a manner similar to that of evaporated silver. The average values of  $R$  and  $\rho$  are given in Table I. The values of  $E/HI$  against  $\epsilon$ , and  $\rho$  against  $\epsilon$ , are plotted in Fig. 3. As in the case of silver, evaporated copper has a large specific resist-

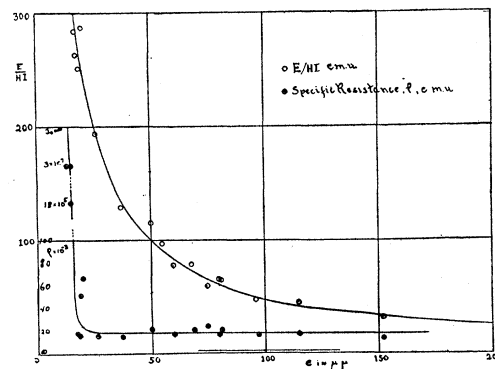


Fig. 3.

ance, although the Hall coefficient does not differ appreciably from the bulk value.

The copper films like silver were very good reflectors, although they reflect slightly more to the red than does silver. They appear green by transmitted light. Structure analysis was not made for the copper films.

<sup>1</sup> Hull, loc. cit.

*Evaporated Iron Films.*

Evaporated iron films behave in a manner similar to bulk iron in that they have a positive Hall coefficient which is not constant with magnetic field. Fig. 4 shows the relation between the Hall e.m.f. and the magnetic field for several films of different thicknesses. It will be seen that  $E$  is proportional to  $H$  up to fields of about 10,000 gauss. For bulk iron this proportionality holds for fields up to about 20,000 gauss. The bend in the curves is spoken of as the saturation of the Hall e.m.f. The average value of the Hall coefficient given in Table I., is computed for fields below 10,000 gauss by means of the relation

$$E = \frac{RHI}{\epsilon}$$

In contrast to its behavior in copper and silver, the Hall coefficient of evaporated iron shows an increase of about 600 per cent. over that of the bulk metal.

Wire containing 99.85 per cent. iron was used, so that the films obtained were relatively pure. The films appeared silver white by reflected light, and rather brownish by transmitted light. No trace of structure was visible under the microscope. The films reflect very well, were hard, and did not oxidize readily in air.

Evaporated iron was found to be crystalline, the lines occurring at the angular deviations computed from Hull's data.<sup>1</sup> As in the case of silver, it is inferred from the correspondence of these lines that the type of lattice is that found by Hull, which in iron is the centered cubic, of elementary side 2.86 A.U.

## DISCUSSION.

Let us consider in more detail the nature of evaporation in the sense that we use it. The wires were run at temperatures<sup>2</sup> that correspond to a vapor pressure of  $10^{-3}$  mm. of mercury. At this pressure, the distance of the wire from the microscope slide was less than the mean free path. It has been found that metallic vapors are monatomic.<sup>3</sup> Then if the

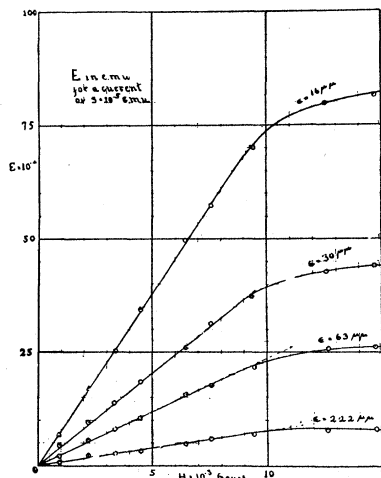


Fig. 4.

<sup>1</sup> Hull, loc. cit.<sup>2</sup> Johnston, Journ. Ind. Eng. Chem., 9, p. 873, 1917.<sup>3</sup> Meyer, Ber., 12, p. 1426, 1879.

atoms are not reflected<sup>1</sup> upon striking the slide, the film must be deposited an atom at a time, the atoms reaching the slide without collision with each other. Since the evaporated metals are crystalline, some of the atoms evidently do not take random positions, but take definite places in a lattice, probably under the influence of the atoms already there. It is thought that rapid evaporation with a large temperature difference between the slide and the wire would tend to diminish crystalline growth.

The term crystalline grain is used in the sense employed in metallurgy,<sup>2</sup> that is, to designate a crystal of metal, in which the elementary crystals all have the same orientation. Its properties are supposed independent of the manner of obtaining it, as long as the same type of elementary crystal is obtained. Any specimen of metal is, in general, made up of an aggregate of crystalline grains separated by boundaries. The properties of the specimen are supposed to depend upon the size of the crystalline grains, and upon the contacts between them.

We conclude that in evaporated films, the crystalline grains are much smaller than in bulk metals, and that they are not in the intimate contact obtaining in bulk form. This view is supported by the fact that the metals have been found to be crystalline, that the crystalline grains were not visible under a microscope, that the surfaces of the films appear to have a very high polish, and that the essential nature of the process of obtaining them is not conducive to crystalline growth.

The large resistivity of evaporated metals leads us to believe that the resistivity of a specimen of metal will be smallest at a given temperature and pressure, when the specimen is a pure metallic crystal. The effect of the boundaries that separate adjacent crystalline grains is to increase the resistivity of the specimen. This increase is very pronounced when impurities are present that freeze out in the process of solidification. An idea of the effect upon the resistivity of a change in the size of the crystalline grains alone can be obtained by comparing the resistivity of two specimens of pure metal, the one obtained by sudden quenching, the other by slow cooling, during solidification.

The results on silver and copper indicate that the Hall coefficients of these substances do not change appreciably when the structure is changed. This might be expected, since the supposition that the impressed magnetic field produces the transverse e.m.f. accounts well for the observed effects. In the case of iron, however, for which the Hall coefficient is positive and therefore intimately associated with the molecular fields, one might expect structure to have a pronounced influence.

<sup>1</sup> Langmuir, *PHYS. REV.*, 8, p. 149, 1916.

<sup>2</sup> See Sauveur, "The Metallography of Iron and Steel," and Rosenhain, "The Study of Physical Metallurgy."

A. W. Smith<sup>1</sup> has found that as temperature is increased, the Hall coefficient of iron increases, and the value of the magnetic field at which the Hall e.m.f. becomes saturated shifts to smaller fields. He points out that the saturation of the Hall e.m.f. should occur when  $H = 4\pi I_m'$ , where  $I_m'$  is the maximum value of the intensity of magnetization, and that the decrease of  $I_m'$  as temperature is increased accounts for the observed shifts. Now our proportionality between  $E$  and  $H$  holds only up to one half the value of  $H$  for pure bulk iron. Furthermore the directional change in  $R$  is the same in the two cases. Thus it appears that if the Hall effect is essentially the same phenomenon in the two cases,  $I_m'$  for evaporated iron is less than that for bulk iron.

The results indicate, then, that the effect of decreasing the size of the crystalline grains has been to decrease the maximum value of the intensity of magnetization that can be attained in the iron. This is not so surprising if we suppose that the permeability of the boundaries is different from that of the crystalline grains. Then each crystalline grain would have poles at the boundaries, and as the grains become smaller, the poles would exert a greater demagnetizing force. It does not seem improbable that a perfect crystal of iron would have the greatest  $I_m'$ , and that the difference between  $I_m'$  for a perfect crystal and for a specimen of like size and shape, but made up of crystalline grains, would be a measure of the demagnetizing effect of the poles. Thus it appears that new importance is attached to the experiments in that they bear upon the nature of ferromagnetism.

In conclusion, I wish to express my appreciation to the members of the Department of Physics of the State University of Iowa for their assistance and interest, and especially to Dr. G. W. Stewart, under whose direction the work was carried on.

PHYSICAL LABORATORY,  
STATE UNIVERSITY OF IOWA,  
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<sup>1</sup> PHYS. REV., 30, p. 1, 1910.