

THE MAGNETIC PROPERTIES OF EVAPORATED
NICKEL AND IRON FILMS

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ABSTRACT

Coercive force, retentivity, and hysteresis loop forms for evaporated films of Ni and Fe.—Films of nickel and iron, produced by evaporation at low pressure, were studied with the object of verifying the reported existence of an abrupt change of coercive force at a critical thickness, and also to discover the cause of certain peculiar hysteresis loop forms. *Critical thickness* was found for iron films at approximately $50\text{ m}\mu$, in satisfactory agreement with Sorensen. At this thickness the coercive force changes abruptly from the high value of approximately 100 for thinner films. The observations were made upon films deposited on a base of aluminum foil, .0025 cm in thickness. The foil was heated previous to and during deposit. No such critical thickness was observed in nickel. Peculiar *hysteresis loops* were found. Nickel films unheated have very narrow loops, with magnetic induction nearly proportional to the field up to 139 gauss, where the induction is one-third to one-half that of the metal in bulk. Films heated previous to and during deposit give a magnetic induction at 139 gauss that is approximately three times as great, or like that of metal in bulk. But the retentivity and coercive force increase five-fold and become less like those of the metal in bulk. In similarly heated films of iron, these last two magnetic properties are also much greater than with metal in bulk. The peculiarities of the films are probably caused by the nature of the crystalline state. To what extent the phenomena depend upon the presence of the aluminum base is yet to be ascertained. The present view is that the presence of gas alters the crystal growth. On the whole the experiments seem to emphasize the importance of the influence of the crystalline state upon magnetic properties.

THE investigation of thin films of the ferromagnetic elements has disclosed two peculiar properties,—a very large change in magnetic induction at a fairly well defined critical thickness, and under certain conditions broad, nearly rectangular, hysteresis loops. The earlier work¹ done in this field was on films electrolytically deposited, and the above effects were attributed² to the influence of various occluded gases present, especially hydrogen. If, however, these properties are inherent in the metals themselves, the establishment of this fact is of importance in the theory of magnetism.

In order to test the magnetic as well as other properties of the ferromagnetic elements, deposited in the absence of contaminating influences,

¹ Seckelson, Ann. d. Physik **67**, 37 (1899). Maurain, Jour. d. Physique, **10**, 123 (1901); **1**, 90 (1902). Schild, Ann. d. Physik, **25**, 586 (1908); **1**, 151 (1902). Gans, Phys. Zeits. **12**, 911 (1911); and others.

² Kaufman and Meier, Phys. Zeits. **12**, 513 (1911).

this laboratory has been making a study of films produced by evaporation. Sorensen³ found some evidence of a thickness critical for coercive force in evaporated films of nickel at about 200 $m\mu$ and in iron at 55 $m\mu$. The coercive force changed abruptly to lower values as the thickness was increased.

The first purpose of the present investigation was to check these critical thicknesses. If they exist, are they independent of the various conditions accompanying deposition, such as the heat-treatment of the film, the emission velocity from the evaporating filament, and the residual gas pressure?

The apparatus used in this study was not essentially different from that used by Sorensen, to whose paper the reader is referred for details. The evaporation occurred in a horizontal cylindrical glass vessel, containing two film carriers, one below the hot filament and one above. An electric heater was placed under the lower one. The carriers moved back and forth perpendicularly to the filament and in a manner insuring a uniform deposit. Later observations by Mr. K. J. Miller have shown that the variation in film thickness does not exceed 5 percent except at the extreme edge of the film. As a base for the films, aluminum foil one mil in thickness was employed. The lower film is the hotter as the upper one is not only further from the heater, but it is also shielded from it by the lower foil. Temperatures of the lower foil were measured for various heating currents by means of a thermocouple. Temperatures at the upper position though not determined were in all cases much lower.

To reduce the initial magnetization observed by Sorensen in some of his films, brass parts were substituted for iron in the evaporation apparatus, but as noted later, this substitution failed to accomplish its purpose.

RESULTS FOR NICKEL

In order to determine the critical thickness of nickel, if such existed, a large number of films were deposited and tested. It was found that the thickness affected the magnetic properties only slightly if at all, but variations of the heat-treatment during deposition had very considerable effects. The films are therefore classified according to their heat-treatments.

The results are presented in Table I. Subsequent to nickel film No. 6 all films deposited in the lower position are listed under an odd number, those deposited above, under an even. The only films discarded were those which had oxidized, or those deposited during improper function-

³Sorensen, *Phys. Rev.* **24**, 658 (1924).

ing of the evaporation apparatus. The graphs are plotted with ordinates 4π times the intensity of magnetization, rather than induction.

TABLE I

Nickel films having minimum heat treatment. δ is the film thickness in $m\mu$; β_{139} , the magnetic induction at 139 gauss; I_r , retentivity in percent; H_c , coercive force in gauss

Film No.	δ	β_{139}	I_r	H_c	Film No.	δ	β_{139}	I_r	H_c
8	23	1350	16	5	36	104	1940	26	24
2	39	1240	18	5	44	123	2100	32	32
5*	40	1330	30	11	13**	132	1680	31	35
40	67	2120	25	32	38	139	2100	28	29
14	75	1140	12	17	34	147	1520	12	10
32	77	2040	36	26	10	222	1300	25	28
20	90	1250	14	12	<i>Bulk nickel</i>				
22	96	1310	31	35	Annealed		4900	31	1.2
28	99	1430	32	35	Unannealed		3600	68	30

* 5 is an upper film.

**Heater not in action during deposition of 13.

Though most of the "unheated" films of Table I were subjected to the indirect heating effect of the heater in addition to that of the filament, the similarity of their loop forms would indicate that their temperatures did not differ widely. Films No. 34 and No. 10, Fig. 1, *a* and *b*, show the limits between which most of them lie. In Fig. 1, *c* is a hysteresis loop of a well-annealed specimen of the nickel wire (dimensional ratio 265) used for filaments, while *d* is a loop of an unannealed specimen of the same wire. All graphs of Fig. 1 are plotted on the same scale of magnetization and field strength.

The increasing retentivities and coercive forces with increasing thickness observed in Table I are probably due to the more prolonged heating which the thicker films necessarily received, since No. 34, $\delta=147$, for which the tube was water-cooled throughout, though a comparatively thick film, has the narrow form of loop shown by the thinner films. In those least heated, the coercive force and the area of the hysteresis loops are almost vanishingly small. In this respect they resemble well-annealed nickel. The dissimilar forms of their loops however make this similarity meaningless. The annealed bulk nickel is nearly saturated at a low field, while in the films, the magnetic induction is nearly proportional to the field up to the strongest fields that were available, yet at all points low compared with that of bulk metal.

All foils were heated in contact with a carbon lamp some twenty minutes before the pre-deposit weighing. Yet later work indicated that the occluded gas and water vapor still remaining on the foil largely affected

the magnetic properties of the film deposited thereon. To test this evidence, two films were deposited unheated on foils which had been heated

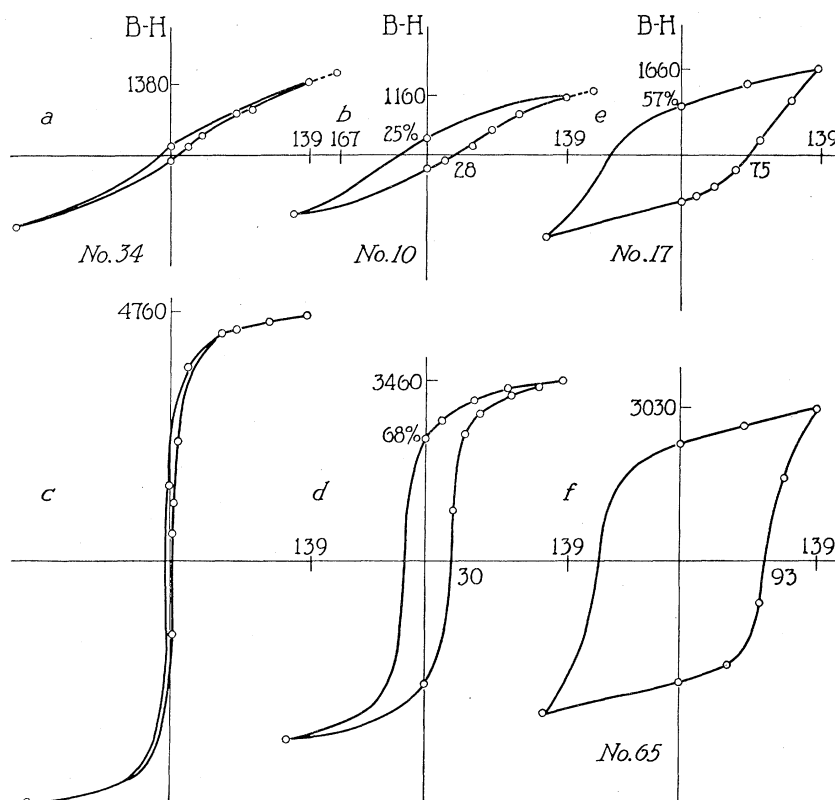


FIG. 1. Hysteresis curves for nickel films.

to 325° for forty minutes in the evaporation apparatus before depositing the films. The following results were obtained:

r	Film No.	δ	β_{139}	I_r	H_c
	72	83	2130	44	46.5
	71	161	2680	58	49

Changing the position of No. 72 in the evaporation apparatus during the interval between the heating and the evaporation process permitted its exposure to the atmosphere for several minutes and probably accounts for its lower induction, retentivity, and coercive force. Nevertheless both films show a decided increase in these properties over the films also deposited unheated but whose foils did not receive such a pre-deposit heat-

treatment. Their characteristics are very similar to those of the heated films of Table II following.

In order to determine the loss in weight of the foil due to extreme heating, two foils were heated to 325° for an hour in the evaporation apparatus and then reweighed. The loss in weight in neither case exceeded 0.05 mg, which corresponds to the weight of a nickel film of about 3m μ in thickness.

TABLE II

Nickel films heated during deposition, foils not previously baked. δ is the film thickness in m μ ; β_{139} and β_{167} , the magnetic induction at 139 and 167 gauss, respectively; I_r , retentivity in percent; H_c , coercive force in gauss

Film No.	δ	β_{139}	β_{167}	I_r	H_c	Film No.	δ	β_{139}	β_{167}	I_r	H_c
6	51	1700	1950	67	67	17	135	1800	2300	57	75
4	63	2300	2460	59	55	35	176	2550	2700	54	79
1	68	1740	1900	48	58	27	180	2430	2820	67	78
31	99	2510	3100	55	58	9	388	2160	2500	67	67
11	103	2010	2230	40	75						

Table II lists those films which were heated during deposition to a temperature of about 200°, but the foils of which were given no pre-deposit heating. Fig. 1 *e* shows the graph for a representative film of this group, No. 17, ($\delta = 135$). There is no evidence of any critical thickness shown in this table.

Table III presents the data for the films whose foils were heated for forty minutes at 200° previous to deposition, as well as heated to the same temperature during deposition. These show a further large increase in coercive force and retentivity. Fig. 1 *f* is for film No. 65, ($\delta = 257$), which

TABLE III

Films heated during deposition, foil previously well baked. δ is the film thickness in m μ ; β_{139} and β_{167} , the magnetic induction at 139 and 167 gauss, respectively; I_r , retentivity in percent; H_c , coercive force in gauss

Film No.	δ	β_{139}	β_{167}	I_r	H_c	Film No.	δ	β_{139}	β_{167}	I_r	H_c
51	50	1830	2800	59	81	73	179	2200	2650	80	96
63	63	1220	1900	72	104	53	182	2620	3100	75	87
47	76	2300	3600	81	99	69	183	2700	3200	76	96
67	78	1500	2100	58	87	87	219	2480	2850	70	88
41	94	2120	2800	72	104	59	224	2960	3350	73	84
45	97	2100	2680	62	94	43	230	2600	2780	67	81
39	104	2350	2850	65	104	37	244	3100	3500	78	93
61	107	1560	2200	69	104	65	257	3170	3520	78	93
21	123	2100	2600	71	104	77	259	2480	2950	68	89
15	161	2600	3000	76	83	55	276	2650	3000	74	78
81	168	2460	2820	71	90	89	304	2960	3300	75	89

may be taken as representative. These films were very hard both mechanically and magnetically. They were initially strongly magnetized, and the unsymmetrical hysteresis loops obtained for many would indicate

that demagnetization was imperfectly accomplished. No critical thickness is indicated.

Oxidation invariably accompanies deposition at temperatures of 300° with the tube exhausted no better than was feasible in this work. The magnetic effects are greatly reduced retentivity and coercive force,—the former by a fourth or fifth, the latter by a half. There is no decrease in the calculated magnetic induction, though a part of the weight of the film from which the magnetic induction is calculated is nickel oxide. The induction is low however compared with that of bulk metal.

The brass surfaces in the evaporation apparatus adsorbed much gas and prevented the attaining of vacua much better than .001 mm and when the tube was heated during the deposition of a film, rapidly rising pressures invariably occurred. When pressures reached about .005 mm, the evaporation and heating processes were interrupted to permit reduction of the pressure. The most serious result was in the unequal heat-treatments which different films, supposedly treated alike, received, and it is to these variations rather than to the pressure variations that the comparatively wide range of results for heated nickel films is probably due. Later test cases showed the magnetic characteristics to be practically independent of pressure up to at least .015 mm, that is, the magnetic properties of such films were certainly no less uniform.

After some of the nickel films used in this investigation had stood in air for six weeks, they showed a magnetic induction lowered by about ten percent. There was however no change in the retentivity or coercive force. The slow change in properties of nickel films was also observed by Peacock⁴ in his work on the Hall effect, and by Manning in his yet unpublished work on optical effects.

RESULTS FOR IRON

The brass parts introduced in the evaporation apparatus to reduce the initial magnetization, not only failed to accomplish their purpose with the nickel films, but also owing to adsorbed gases introduced serious difficulties in the pressure control. The brass parts were accordingly replaced with iron for the work on iron films. Pressures through this portion of the investigation were maintained at less than .001 mm.

Preliminary tests indicated a broadening of the hysteresis loops with increasing heat-treatment as was the case with nickel. But in order to test for the presence of a critical thickness, most of the iron films were given the same type of heat-treatment,—heating the foil to 200° for an hour and then depositing the film at the same temperature. As in the case

⁴Peacock, *Phys. Rev.* **27**, 474 (1926).

of nickel, the iron films of Table IV are listed in order of their thickness as calculated from their weighings. There is striking evidence of a critical thickness shown by this table. Typical hysteresis loops (No. 5 and No. 9) below and above the critical thickness are shown in Fig. 2, *a* and *b*.

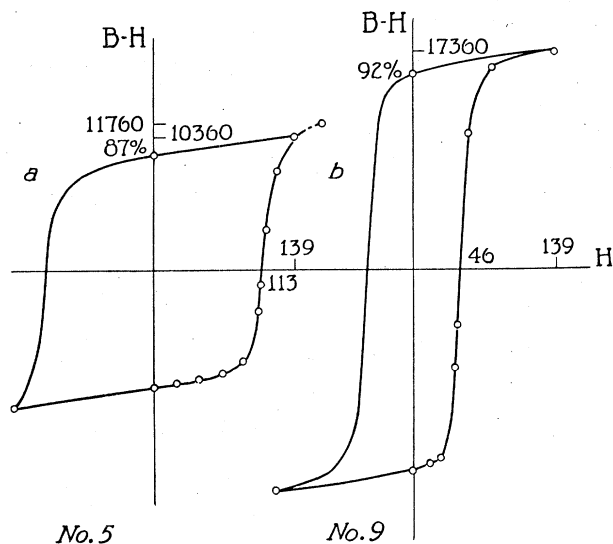


FIG. 2. Hysteresis curves for iron films.

The coercive forces of these films would indicate a thickness between 35 and 44 $m\mu$ as critical. However, the column of magnetic inductions

TABLE IV

Results for iron

δ is the film thickness in $m\mu$; β_{139} and β_{167} , the magnetic induction at 139 and 167 gauss, respectively; I_r , retentivity in percent; H_c , coercive force in gauss

Film No.	δ	β_{139}	β_{167}	I_r	H_c	Film No.	δ	β_{139}	β_{167}	I_r	H_c
45	22	15,450	16,100	88	89	9	68	17,500	17,800	92	46
5	28.4	10,500	11,900	87	113	35	69	17,000	17,400	90	45
29	30	13,800	16,600	89	110	15	69.5	16,300	16,400	92	43.5
19	34	8,600	10,200	77	99	43	71.5	16,300	16,600	92	39
49	35	15,300	16,200	93	95	11	79	15,400	16,000	90	46
39	44	19,250	19,600	91	49	3	85	18,100	18,200	90	35
41	54	16,000	16,600	90	52	7	141	15,200	15,400	83	23
21	54	16,100	16,200	91	48	Unannealed iron ⁵					
47	59	13,700	14,100	90	69			18,500	18,700	70	9

furnishes a valuable check on the correctness of the calculated thickness and indicates a slightly higher critical value. Film No. 39 is evidently of

⁵ Ewing, J. A., "Magnetic Induction in Iron and Other Metals."

a greater thickness than the value calculated from weighting. Taking $\beta_{139} = 16,000$ as the average magnetic induction of films of near the same thickness from which to calculate the thickness of No. 39, we obtain $\delta_{39} = 52$ instead of 44. Similarly calculating the thickness of No. 47, we obtain $\delta_{47} = 50$ instead of 59 $m\mu$, which would make 50 $m\mu$ appear as the best value of critical thickness. These corrections indicate that the error in the thickness determination may be as much as 15 percent. But Table IV gives unmistakable evidence of a thickness critical for coercive force, placing its value at approximately 50 $m\mu$.

The iron filament shows a strong tendency to oxidize even at pressures of .001 mm. The maintenance of evaporation then requires a progressively increased heating current, and at much higher pressures evaporation may not be accomplished. The study of the effect, if any, of pressure variation on the magnetic properties of the iron films would obviously require the use of oxygen-free gas. This was not attempted.

According to Ewing⁵, the magnetic induction of bulk iron for the maximum fields used in this work should be about 18,700—(there is little difference in magnetic induction for annealed and unannealed iron at this field strength.) The magnetic induction obtained by the writer for evaporated iron films runs about 15 percent below this value. This does not agree with Sorensen's determinations which were of the order of 15 percent higher than that of bulk iron. The lower value would seem to be more in accord with the predictions of the Hall-effect in evaporated iron films^{4,6} in that the low value of field required to saturate the Hall e.m.f. implies that a smaller intensity of magnetization is possible in such films than in bulk metal. It is possible, however, that the difference in magnetic inductions in the two investigations is due to dissimilar heat-treatments.

SUMMARY OF RESULTS

This investigation of the magnetic properties of evaporated nickel and iron films was undertaken with the hope of determining the existence of a thickness critical for abrupt change in the coercive force, and of discovering the cause of certain peculiar hysteresis loop forms. Though difficulties arising from occluded gases may have prevented the appearance of a critical thickness in nickel, surprising consequences of various types of heat-treatment are revealed. Nickel films deposited unheated have very narrow hysteresis loops, remarkable in that the magnetic induction is nearly proportional to the field up to 139 gauss. The magnetic induction at this field is from a third to a half that of unannealed nickel in bulk. If, however, the occluded gases are driven out by a pre-deposit heating, the

⁶ Steinberg, Phys. Rev. **21**, 22 (1923).

magnetic induction, retentivity and coercive force all increase very decidedly giving magnetic properties very similar to those obtained on heating only during the deposition of the film. But if these heat treatments are combined, a large further increase in these properties is obtained, in which case the films are very hard mechanically as well as magnetically, and though magnetic fields in the evaporation apparatus were neutralized as far as possible, these films were all initially rather strongly magnetized.

In iron there is observed a thickness of about $50 \text{ m}\mu$, which is critical for coercive force. This is approximately the value determined by Sorensen.⁴

The hysteresis loops for iron obtained in the two investigations both above and below the critical thickness are in agreement. Coercive forces and retentivities are numerically practically the same, but the present investigation gives values of magnetic induction which are about 30 percent lower than those of the earlier work.

DISCUSSION

There is a definite effect of heating the film, namely an abnormally large hysteresis loop, and, at large fields, a more nearly normal susceptibility. There is much evidence that these effects are in some way related to the crystal formation. That the deposition of a metallic film on a cold surface is amorphous was first stated by Langmuir.⁷ This theory has since had experimental verification with x-ray spectrograms⁸ and with the ultra-microscope⁹, while a post-deposit heating was found to induce a crystal growth. Ingersoll and DeVinney¹⁰ found that sputtered nickel films deposited at liquid air temperature were practically non-magnetic. After being heated, these films assumed their usual magnetic properties. Though in no case were the films of the present investigation deposited at a really low temperature, the magnetic induction of the least heated films is very small even in the strongest fields available, and there is reason to believe that if deposition had occurred at very low temperatures, the films might be non-magnetic. The temperature required to induce crystal growth during deposition, if this is the cause in the change in magnetic properties, is decidedly less than that required after deposition has occurred.

There are some extraneous influences present whose effects might be considered as masking the intrinsic properties of the metal. That the

⁷ Langmuir, *Am. Chem. Soc. J.* **38**, 2221 (1916).

⁸ Kahler, *Phys. Rev.* **18**, 210 (1921).

⁹ Reinders and Hamburger, *K. Akad. Amsterdam Proc.* **19**, 958 (1917).

¹⁰ Ingersoll and DeVinney, *Phys. Rev.* **26**, 86 (1925).

effect of a pre-deposit heating of the foil is due to driving off the gas can scarcely be questioned. The fact that a film deposited *unheated* on a foil, which has been *previously heated*, has magnetic characteristics very similar to those of a film produced on a foil which is heated *during deposition only*, might infer that heating the foil either before or during deposition merely drives off the gas. But heating the film during its deposition produces the same change and of a similar magnitude whether or not the gas has been initially baked out of the foil. In either case the loop forms become more rather than less abnormal. It is evident, then, that the peculiar hysteresis loops cannot be due to the presence of gas. On the contrary, it seems that the gas, directly or indirectly, interferes with the phenomena.

We have assumed that the aluminum foil base on which deposition occurs does not affect the magnetic properties of the film. There are, however, two possible ways in which its influence might be present. The projected particles from the filament may alloy with the aluminum of the base. Were this true, the magnetic properties of the film would show decided change with thickness as the comparatively slow velocity with which the molecules emerge from the filament could not penetrate a depth of several hundred molecules of the aluminum. A second possible influence of the base is its interference with magnetostriction. Though this would not seem great enough to be the cause of the peculiar phenomena, experiments are now being devised in this laboratory in which all influences of the base will be removed.

One other operation deserves notice. When the film with its aluminum foil base is rolled for insertion into the magnetic test apparatus, a compression of the film must occur. For a film of thickness $100\text{ m}\mu$, on a foil of 1 mil thickness such as was used, rolled to a radius of curvature of 1mm which is approximately the minimum, this compression would amount to about one percent. Furthermore one side of the film is compressed slightly (about one percent) more than the other. Though the compression is much more than the elastic limit of the metals employed, since all films are similarly treated in this respect, it would not cause the remarkable variation observed in the film properties. However annealing after the rolling process should be tried.

It is well known that hysteresis decreases with increasing crystal size of metal in bulk. These crystals are probably very large as compared with those of evaporated films. It has been suggested that the function which determines hysteresis may be of a type that vanishes for either very large or very small crystals. Jensen¹¹ regards hysteresis as largely deter-

¹¹ Jensen, J. Am. Inst. Elec. Eng. **43**, 558 (1924).

mined by the amount of cementing material between the crystals which would ordinarily increase with decreasing size. It seems possible however that crystal strain or deformation may be a more important factor in determining hysteresis than mere crystal size. Perhaps perfect crystals are not formed by the evaporation process. The velocity of the particles on impinging may be too great for the molecular field properly to orient them. The observed film properties were not altered appreciably by changes in the velocity of emission, produced by variations of filament temperature. Welo and Baudisch¹² have produced crystals of magnetite by methods giving very different crystal sizes. Before they are annealed, their magnetic properties differ widely, but after annealing, even though crystal sizes remain dissimilar, their magnetic properties become similar. The necessity for annealing their crystals was probably occasioned by the rapidity of formation. It is possible that annealing the evaporated films would result in more nearly normal crystals.

McKeehan¹³ considers atomic magnetostriction as the prime cause of hysteresis, and in the experimental work on which his conclusions were based, this view would seem to be correct. It would be remarkable indeed if all hysteresis could be traced to this one cause. The evidence obtained by this study of thin films indicates that hysteresis is a crystalline rather than an atomic phenomenon.

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

HALL OF PHYSICS,
UNIVERSITY OF IOWA,
July 10, 1926.

¹² Welo and Baudisch in a private communication.

¹³ McKeehan, *Phys. Rev.* **26**, 274 (1925).