continuous irradiation techniques have also been found in the low-energy irradiations of germanium.[15,16]

The existence of the levels close to the band edges has been investigated by Longo[17] using deuteron bombarded-samples. The energies reported for these levels are essentially the same as those found in this work. As in the present work, Longo found that the level close to the conduction band edge is introduced at nearly the same rate as the level close to the edge of the valence band.

The experiments of Longo and the present work yield different ratios of the experimental maximum removal rate to the calculated defect introduction rate. Assuming a threshold energy of 30 ev, one finds in the case of deuteron bombardment that the calculated defect introduction rate is about equal to the experimental maximum removal rate. For electron bombardment, the experimental maximum removal rate is nearly four times the calculated introduction rate for a 30-ev threshold. It is surprising that the theory gives poorer results for electron irradiations, since the damage is simpler in this case. Nevertheless, these results suggest that there is a real difference in the damage produced by deuteron and electron irradiation.

Thus we see, that there is some agreement from different sources on the existence of various levels and their energies. However, there exist rather serious discrepancies which are not understood at this time.

VI. ACKNOWLEDGMENTS

The author wishes to express his appreciation to Dr. K. Lark-Horovitz, Dr. H. M. James, Dr. J. W. MacKay, and Dr. E. E. Klontz for their advice and assistance during this work.

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[15] J. Kortright and J. MacKay, Bull. Am. Phys. Soc. Ser. II, **3**, 142 (1958).

[16] W. L. Brown and W. M. Augustyniak, Bull. Am. Phys. Soc. Ser. II, **2**, 156 (1957).

[17] T. A. Longo, Ph.D. thesis, Purdue University, 1957 (unpublished); see also T. A. Longo and K. Lark-Horovitz, Bull. Am. Phys. Soc. Ser. II, **2**, 157 (1957).

PHYSICAL REVIEW VOLUME 114, NUMBER 6 JUNE 15, 1959

Transitions from Ferromagnetism to Antiferromagnetism in Iron-Aluminum Alloys. Experimental Results*

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(Received October 16, 1958)

The magnetic behavior of iron aluminum alloys has been determined for compositions from 0 to 40 atomic percent aluminum in external fields to 12 kilo-oersteds from 4°K to 300°K. The alloys above 33 atomic percent exhibit antiferromagnetism. Those below 28 atomic percent are ferromagnetic. Between 28 and 33 atomic percent, the behavior is ferromagnetic at room temperature but shows a transition to antiferromagnetism on lowering the temperature. The effect of order on the magnetic properties has also been investigated. Antiferromagnetism is found only in the ordered structures.

INTRODUCTION

THE body-centered cubic phase of iron can be retained for additions of up to slightly more than one aluminum atom for every iron atom. The iron and aluminum atoms, however, do show strong tendencies toward ordering within this range of solid solution. The exact nature of the phase diagram is still under investigation.[1] The features of the phase diagram with which we are here concerned are illustrated by Fig. 1, where we represent the structures which result for particular heat treatments as a function of composition of the alloys. The two structures referred to as α' and α'' represent different types of ordering. The prototype of the α' structure is Fe_3Al and the prototype of the α'' structure is FeAl. The designation [Fe_3Al] will, there-

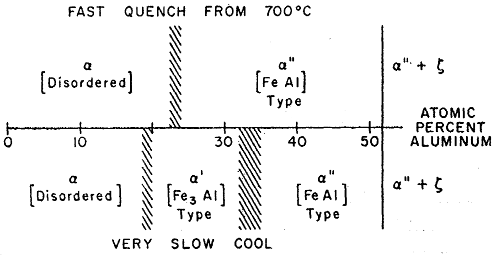

FIG. 1. Schematic diagram of structure resulting from different heat treatments as a function of atomic percent aluminum in iron.

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* A brief report of this and the following paper appeared in the Proceedings of the Conference on Magnetism and Magnetic Materials [H. Sato and A. Arrott, J. Appl. Phys. **29**, 515 (1958)].

[1] A. J. Bradley and A. H. Jay, J. Iron Steel Inst. (London) **128**, 339 (1932); Proc. Roy. Soc. (London) **A136**, 210 (1932); A. Osawa and M. Murata, J. Japan Inst. Metals **5**, 259 (1942); H. Sato, Science Repts. Research Inst., Tôhoku Univ. **A3**, 13 (1951); A. Taylor and R. M. Jones, *Conference on Magnetism and Magnetic Materials* (American Institute of Electrical Engineers, New York, 1957), p. 246; J. Appl. Phys. **29**, 522 (1958); and private communication; Kayser, Showak, Heimerdinger, and Zackay (private communication).

fore, be used for the α' structure at nonstoichiometric compositions and the [FeAl] designation similarly denotes the α'' structure. If a body-centered cubic lattice is considered as two interlocking simple cubic sublattices each of which is further subdivided into two face-centered cubic sublattices, then FeAl has aluminum atoms on only one of the two simple cubic sublattices and Fe_3Al has aluminum atoms on only one of the four face-centered cubic sublattices. Table I gives the atom positions for the nonstoichiometric cases. Note that according to Fig. 1, for either heat treatment, an ordered state is obtained above 24 atomic percent Al. The only way so far determined to obtain a disordered structure above 24 atomic percent Al at room temperature is by means of severe cold working.[2]

Previous work[3] has shown that up to approximately 20 atomic percent aluminum, the magnetic saturation moment of iron-aluminum alloys decreases with increasing aluminum content at a rate comparable with what would be expected from simple dilution. Above this concentration, the moment falls sharply with increasing aluminum content.

The present work was motivated by a desire to investigate more fully the effects of order on the saturation magnetization and to find out the nature of the alloys with aluminum content greater than that for which previous results show by extrapolation the disappearance of ferromagnetism. The qualitative question was answered first by the authors' discovery of antiferromagnetism in alloys with greater than 33 atomic percent aluminum.[4] The subsequent measurements of the temperature dependence of magnetization of the "ferromagnetic" alloys, in order to investigate the role of order, showed the even more startling effect of the disappearance of ferromagnetism with decreasing temperature for the often-studied composition of 30.5 atomic percent.

TABLE I. Distribution of Fe and Al atoms on the simple cubic (sc) and face-centered cubic (fcc) sublattices of the body-centered cubic (bcc) lattice.

bcc					
sc		sc			
fcc	fcc	fcc	fcc	Name given structure	Fraction of Al atoms
Fe+Al	Fe+Al	Fe+Al	Fe+Al	[Disordered]	$c>0$
Fe	Fe	Al	Al	FeAl	$c=0.5$
Fe	Fe	Fe	Al	Fe_3Al	$c=0.25$
Fe	Fe	Fe+Al	Fe+Al	[FeAl] type	$c<0.5$
Fe	Fe	Fe+Al	Al	[Fe_3Al] type	$c>0.25$
Fe	Fe	Fe	Fe+Al	[Fe_3Al] type	$c<0.25$

TABLE II. Composition of alloys.

Sample number	Atomic %	Weight %	Analysis spread (weight %)
753	6.0	2.97	2.86– 2.97
715	11.7	6.05	6.02– 6.05
530	19.4	10.4	10.35–10.50
532	21.8	11.9	11.7 –12.0
746	24.4	13.5	13.3 –13.5
341	25.5	14.2	13.8 –14.6
786	25.5	14.2	14.2 –14.5
755	28.3	16.0	16.0
787	30.5	17.5	17.4 –17.6
956	33.8	19.8	19.8 –20.6
955	35.2	20.8	20.8 –21.7
847	36.3	21.6	21.6 –21.7
957	42.7	26.5	26.5

The investigation to date includes determinations of magnetic isotherms for annealed and quenched specimens at temperatures of 300°K, 77°K, and 4.2°K for applied fields up to 12 kilo-oersteds. The compositions investigated are, in atomic percent, 6.0, 11.7, 19.5, 22.0, 24.4, 25.5, 28.3, 30.4, 33.8, 35.2, 36.3, and 42.7. The last five compositions were investigated at a considerable number of temperatures between 4°K and 300°K.

EXPERIMENTAL PROCEDURES

Specimens were prepared by the staff of the Metallurgy Department of the Scientific Laboratory of the Ford Motor Company. They have developed means of minimizing oxygen as an impurity and are able to roll ingots with up to 31 atomic percent aluminum to a 90% reduction in area. Chemical and spectrographic analyses of compositions up to 31 atomic percent Al have been supplemented by density and resistivity measurements in order to determine chemical compositions.[5] The specimens with compositions above 33 atomic percent Al were taken from cast ingots. Table II gives the spread in chemical analyses as found from different laboratories together with what is considered the best value on the basis of chemical analysis, density, resistivity, and magnetic data. The specimens taken from the cast ingots showed the greatest variation in chemical analysis. This variation may well be a true reflection of the inhomogeneity of these specimens.[6] For this reason, the results presented here for the four alloys of higher aluminum

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[2] This effect is striking in that nonmagnetic materials become strongly magnetic, as has been noted by us and confirmed by other investigators.

[3] M. Fallot, Ann. phys. **6**, 305 (1936); W. Sucksmith, Proc. Roy. Soc. (London) **A171**, 525 (1939); M. Yamamoto and S. Taniguchi, Science Repts. Research Inst. Tôhoku Univ. **A8**, 112 (1956); J. E. Goldman (private communication of unpublished results).

[4] A. Arrott and H. Sato, Bull. Am. Phys. Soc. Ser. II, **2**, 118 (1957).

[5] F. X. Kayser, Wright Air Development Center Technical Report WADC-57-298 (to be published).

[6] An indication of this is found in the preparation of specimens for neutron diffraction. These alloys are antiferromagnetic in the bulk, but on filing or crushing in a mortar and pestle they become strongly ferromagnetic. On subsequent heat treatment they become antiferromagnetic again, except for a small fraction which can be removed from the heat treated powder by magnetic separation. Analysis shows these more magnetic particles to be much more iron-rich than the bulk. In particular, a powdered and annealed 42.7 atomic percent specimen showed that the 1% which could be removed by a small Alnico permanent magnet has a mean chemical composition of 32 atmoic percent Al.

content are to be considered qualitative rather than to be interpreted in a quantitative sense. The work of preparing homogeneous high aluminum alloys and obtaining accurate analyses is in progress and a more detailed investigation of the alloys approaching stoichiometric FeAl will be reported on later.

The heat treatment given all but the four alloys of higher aluminum content consisted of hot-forging $2\frac{1}{2}$-in. ingots to $\frac{5}{8}$-in. square bars, turning 1-cm diameter spheres, vacuum-annealing in sealed Vycor at 900°C for two hours, followed by a three-hour cooling to 600°C, leaving at that temperature for two hours, cooling to 550°C in one hour, leaving at that temperature for one day, cooling by 20°C per day until 330°C was reached, and finally slow cooling to room temperature. Two specimens were prepared from each bar. These were compared magnetically before giving one of each pair a quench treatment consisting of heating to 750°C and dropping into water. The heat treatment given to the cast alloys was predicated on the nominal compositions as mixed in the melt. This supposed all compositions to be safely in the [FeAl] region. The chemical analyses show that three of the samples are in a range where the structure is uncertain. In addition, the inhomogeneity illustrated above indicates that the samples are in the region where the heat treatment used would certainly produce a mixture of [FeAl] and [Fe_3Al]. The heat treatment for the latter samples was a four-hour soaking in H_2 at 900°C followed by a five-hour furnace cooling to 200°C and removal from the furnace.

Magnetic moment measurements up to 31 atomic percent were carried out on spherical samples by use of a coil-motion technique. Magnetic moment measurements on the four specimens with higher aluminum content were made by means of a null-coil-and-sample-motion technique using short cylindrical samples. This technique was previously described.[7] Field measurements were with a Rawson rotating-coil gaussmeter. The field measurements may be in error by as much as 2%. The magnetic moment measurements, while capable of a precision of 0.1%, have an uncertainty of ±1% in absolute accuracy due to slight differences in sample size influencing sample placement and to the use of several designs of apparatus during the course of this work. The final design is described here.

The spherical sample is equivalent to a dipole of moment $\mu = MV$, where M is the magnetization of the sample and V is its volume. The dipole is effectively at the center of the sphere; thus any accurate measurement of M requires a means of accurately locating the center of the sphere. A detector coil, translated with respect to the dipole, senses the contribution of the dipole to the field but not the contributions of a superimposed uniform field. The change in field for a given translation of the coil, as seen by a fluxmeter galvanometer connected to the coil, corresponds to a given fraction of the dipole moment. Thus a fixed motion is calibrated by using a standard sphere, or can be self-calibrated as explained below. Fixing the ends of the motion is made less critical if one end is far from the sample and the other is directly over the sample. Sensitivity to position is reduced in the latter case as it corresponds to a maximum coupling of the coil and the dipole. The device shown in Fig. 2 minimizes the mass of the system so that it can be directly immersed in liquid helium from room temperature. Thus a series of samples can be measured at 4.2°K in a time little different than that required for room temperature measurements. The coupling of the detector coil with the applied field on rotation is used to obtain alignment in the magnetic field.[8]

Samples can be compared by placing the samples symmetrically in the specimen holder. Differences of 0.1% are detected and this accuracy is assured by interchanging samples. With the pairs of annealed samples, differences were less than 0.1% before quenching one of them.

Uncertainties in absolute values as great as 1% were noted at liquid helium temperatures due to uncertainties in location of the center of the sample resulting from a combination of thermal contraction of the specimen holder and slight difference in specimen sizes. This has since been corrected by changing construction materials from nylon to linen micarta, but these results have not been repeated. Temperatures were determined from a copper resistance thermometer inserted in the specimen holder and are accurate to 0.5°K, with the possible exception of near 20°K where the errors may be as great as 2°K.

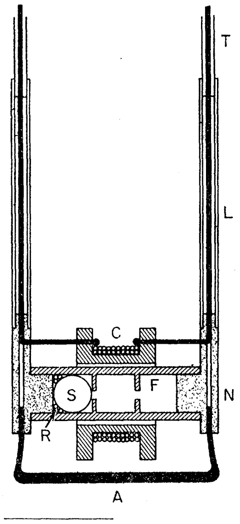

FIG. 2. Cross-sectional view of simple apparatus for measurement of magnetic moment at liquid helium temperatures. The coil C is moved by means of braided nylon strings passing through a nylon guide N. The linen micarta F holds the sample S (1.0 cm in diameter) and a resistance thermometer winding R. The samples are changed by removing the spring clip A. The thin-walled down tubes T are Monel except for a section L of length 12 cm which is linen micarta. The apparatus is directly immersed from room temperature into a 38-mm inside diameter metal Dewar containing liquid helium.

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[7] A. Arrott and J. E. Goldman, Rev. Sci. Instr. 28, 99 (1957).

[8] This has a slight inconvenience in that the initial part of a thermally demagnetized virgin magnetization curve can only be obtained by measuring up to a certain field with the apparatus aligned by eye, then aligning in a finite field, and using the value in fixed field before and after aligning to correct the unaligned values obtained at lower fields.

Measurements of magnetic moment can be made self-calibrating if the demagnetizing factor of the specimen is sufficiently large. This is because, well below saturation, the field inside the specimen is very small compared to the applied field. Thus the applied field is quite closely equal to the demagnetizing field which in turn is determined by the magnetization. Thus, if

$$H_0-H_d\equiv H_i\ll H_0,$$

where H_i is the internal field, H_0 the applied field, and H_d the demagnetizing field, we can write

$$H_0\approx H_d\equiv 4\pi DM,$$

where D is the demagnetizing factor. The reading of the fluxmeter galvanometer, which is proportional to magnetic moment, is proportional to the applied field and we can write

$$H_d=(H_0/\theta)_i\theta,$$

where $(H_0/\theta)_i$ is the ratio of applied field to galvanometer reading in those fields where H_i is negligible. Thus

$$M=(1/4\pi D)(H_0/\theta)_i\theta.$$

As an example, consider iron where values of $M\approx 1000$ emu can be obtained with internal fields less than 10 oersteds. The approximation consists in replacing $H_d=H_0-H_i$ by H_0. In this case $H_i=10$ and $H_d=4\pi D\times 1000$, and the error is $(1/4\pi D)\%$. For a sphere this is less than $\frac{1}{4}\%$. Actually the error can be made much less by taking the mean values between the increasing and the decreasing portions of the hysteresis loop. Because, in this work, the measurements of H_0 were less accurate than the reproducibility of the apparatus on replacing specimens, this expression was useful only as a check.[9]

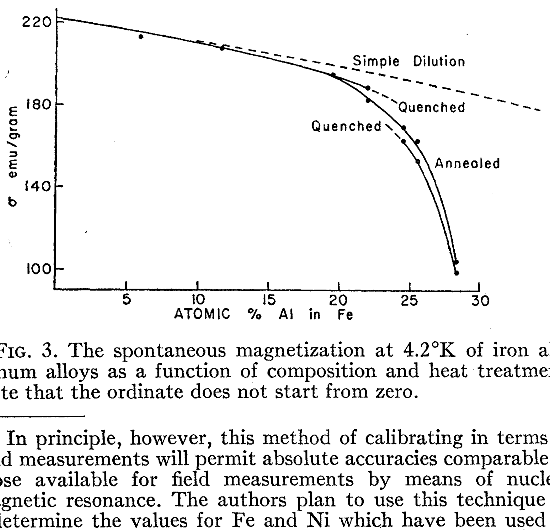

FIG. 3. The spontaneous magnetization at 4.2°K of iron aluminum alloys as a function of composition and heat treatment. Note that the ordinate does not start from zero.

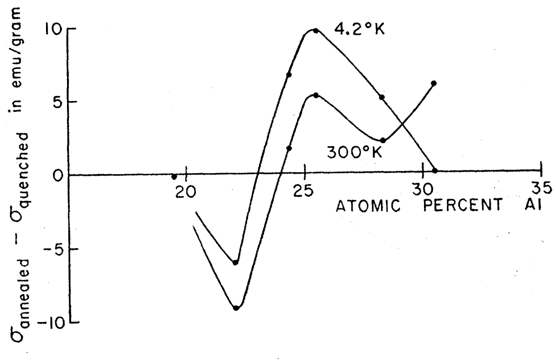

FIG. 4. The differences of spontaneous magnetization between quenched and annealed specimens at 300°K and 4.2°K as a function of aluminum content. Note that the zero value for the 30.5 atomic percent samples result from neither being assumed to have spontaneous magnetization.

RESULTS

The spontaneous magnetization at 4.2°K is shown in Fig. 3 as a function of aluminum content. The value taken for spontaneous magnetization is some sort of fit to the expression,

$$\sigma_{\mathrm{obs}}=\sigma_{\mathrm{spon}}\{1-a/H_i-b/H_i^2\}+\chi_P H_i,$$

where a/H_i represents a "magnetic hardness" associated with structure defects, b/H_i^2 represents the anisotropy of domain rotation, and χ_P is a parasitic paramagnetism which reflects the increase of magnetization within a domain due to the internal field H_i. The effect of heat treatment is made more apparent in Fig. 4 where the differences in magnetization between annealed and quenched samples at room temperature and liquid helium temperature are plotted as a function of aluminum content. The measurements at room temperature are the result of first comparing like pairs of well-annealed specimens by a difference measurement and then quenching one and recomparing it to its unchanged mate. The difference at 4.2°K are between separate determinations of the magnetization.

From this magnetic behavior, the alloys fall into the following groups:

1. The 6.0, 11.7, and 19.5 atomic percent alloys show a decrease in magnetization with increasing aluminum content slightly more rapid than simple atomic dilution of a magnetic matrix by nonmagnetic atoms. Any effect of heat treatment on spontaneous magnetization does not fall outside of the experimental error of 0.1%. These alloys saturate easily and are to be classified as typical ferromagnetic materials.
2. The alloys from 22 to 31 atomic percent show a more rapid decrease in magnetization with increasing aluminum content. These alloys are affected strongly by heat treatment. As is clear from Fig. 3 and 4, the saturation-concentration curves for annealed and quenched specimens cross at about 23 atomic percent Al. This effect has been reported by several workers.[3]

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[9] In principle, however, this method of calibrating in terms of field measurements will permit absolute accuracies comparable to those available for field measurements by means of nuclear magnetic resonance. The authors plan to use this technique to redetermine the values for Fe and Ni which have been used as standards in past work. The best values found in the literature for Fe and Ni at room temperature are inconsistent with the ratio found using a single apparatus, whether that apparatus is a force-measuring or induction-measuring device.

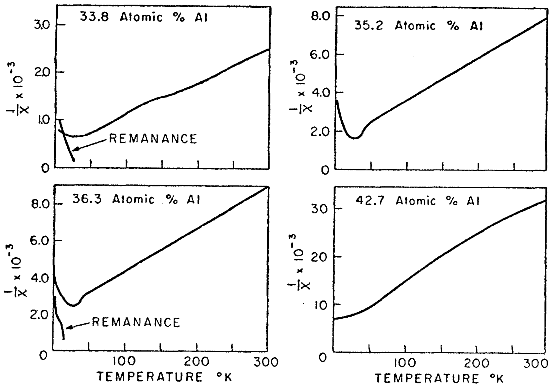

FIG. 5. The inverse susceptibilities of the four cast alloys with higher aluminum contents as functions of temperature. The remanence refers to the magnetization in zero field after magnetizing in an applied field of 12 kilo-oersteds.

Reference to the phase diagram, Fig. 1, leads us to conclude that the saturation value for the disordered state is the highest, that for the [FeAl] state is the lowest, and that for the [Fe_3Al] state is in between. The magnetization of these alloys does not completely saturate in fields of 12 000 oersteds, an effect which can be characterized as a parasitic paramagnetism. This effect becomes increasingly apparent with increasing Al content. Among these alloys, the alloys from 28 to 31 atomic percent show strange magnetic behavior. They show a decrease in spontaneous magnetization with decreasing temperature at low temperatures. These alloys fall in what we term the transition composition range.

3. Finally, the alloys above 34 atomic percent are antiferromagnetic at low temperatures in the ordered state. These alloys display the striking change from weak paramagnetism to strong ferromagnetism on crushing or filing at room temperature.

DISCUSSION OF RESULTS

Since the discussion of these results is predicated upon the assumption of the existence of antiferromagnetic interactions in iron-aluminum alloys, it is well to discuss the data on the four samples with higher aluminum content first. The inverse susceptibility *versus* temperature plots for these alloys are shown in Fig. 5. In all cases, if the data between 50°K and 300°K are fitted by a straight line, the intercept on the temperature axis is negative. If the use of a Curie-Weiss law is justifiable, this implies the presence of antiferromagnetic interactions. The susceptibility of three of the samples shows a clear maximum at approximately 20°K, strongly suggesting normal antiferromagnetic behavior with a Néel point at this temperature. The 42 atomic percent sample shows a leveling off of the susceptibility but not a true minimum.

All samples showed hysteresis effects at liquid helium temperatures. This hysteresis is similar to that observed by one of us in α-Mn [10] and in Hadfield steel. In α-Mn, it has been shown that the hysteresis appears as the result of internal stresses[11] and this has been interpreted as due to antiferromagnetic domain walls made "sticky" by the internal stresses.[12] This hysteresis observed in Fe-Al disappears before reaching the Néel temperature as it does in α-Mn. The remanence which appears after taking the samples to a field of 10 000 oersteds is readily destroyed by an increase in temperature, say from 4°K to 10°K, but can then be reinduced at 10°K by cycling again to high fields. The intrinsic coercive forces in all these samples are of the order of 10^3 oersteds. The magnetization curves at 4.2°K are shown in Figs. 6 and 7. An investigation of the suggested antiferromagnetism in these alloys by neutron diffraction by R. Nathans of the Brookhaven National Laboratory is in progress.

The room-temperature behavior of the alloys with aluminum content less than 32 atomic percent has been the subject of many investigations. The analysis of Yamamoto and Taniguchi has been particularly thorough.[3] Comparison of their magnetic data with previous data as well as with ours suggests that their chemical analysis above 25 atomic percent underestimates the amount of aluminum in their samples. For example, their "17.0 wt %" sample is probably closer to 18.0 wt %. That past observers have not noted the unusual effect we report here is partly because of failure to examine the temperature dependence of the magnetization. Actually the quenched "17.0 wt %" sample of Yamamoto and Taniguchi shows a lack of spontaneous magnetization at room temperature. This they take as evidence that the Curie temperature is below room temperature. However, it appears more likely that this sample shows the same effects as observed on our 17.5 wt % specimen, only in the case of Yamamoto's sample

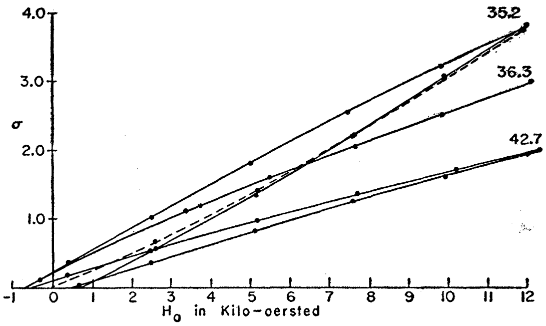

FIG. 6. Magnetization as a function of applied field at 4.2°K. The curve for 36.3 atomic percent aluminum is the demagnetizing portion of a hysteresis loop. The whole loop is shown for the other compositions. The dotted curve is the initial magnetization curve after cooling to 4.2°K in zero field.

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[10] Arrott, Coles, and Goldman, Phys. Rev. **98**, 1864 (1955).

[11] Cooper, Arrott, and Paxton, Bull. Am. Phys. Soc. Ser. II, **2**, 117 (1957); and to be published.

[12] Y. Y. Li, Phys. Rev. **101**, 1450 (1956).

the effects have been moved up in temperature due to the higher aluminum content of his specimen, namely about 31 atomic percent Al.

The nature of our effect is shown most dramatically by the quenched 17.5 wt % (30.4 atomic %) sample, though it is apparent also for the annealed sample. At room temperature, the samples have a sizeable spontaneous magnetization (about that of nickel), a low coercive force, and probably little anisotropy.[13] The only thing particularly unusual at room temperature is the large parasitic paramagnetism evident above saturation. However, when observations are made at liquid nitrogen temperatures, no evidence for a spontaneous magnetization is to be found. Although the measured magnetization in high fields is almost as great as the spontaneous magnetization at room temperature, the low-field data strongly suggest the absence of a spontaneous magnetization. Observations of the temperature dependence of these effects shows a continual decrease in spontaneous magnetization with decreasing temperature below room temperature until the spontaneous magnetization disappears below 180°K for the quenched specimen. (This temperature was not determined for the annealed specimen.) This is illustrated in Fig. 8. It is noted that the magnetization in high fields shows a maximum around 200°K in fields above 5000 oersteds. The behavior below 180°K is strongly paramagnetic as far as its field dependence is concerned, but its temperature dependence is the opposite of any normal paramagnetic material. One might suspect that this sluggish magnetization may be due to the increase of the crystal anisotropy. However, that this is not due to the crystal anisotropy can be inferred from the field dependence of the magnetization. Also a direct observation of the anisotropy by Hall shows little anisotropy in this temperature range.[13]

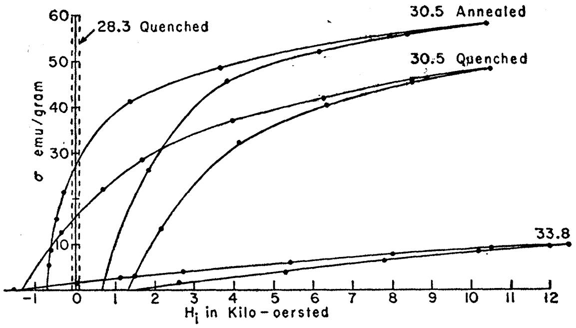

FIG. 7. Magnetization as a function of internal field at 4.2°K The dotted line for 28.3 quenched shows the portion of the magnetization curve which gives the coercive force for that alloy.

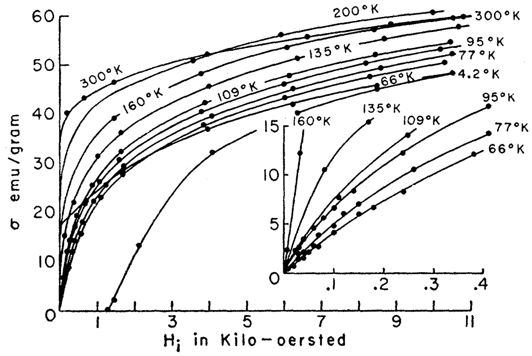

FIG. 8. Magnetization as a function of internal field for several temperatures between 300°K and 4°K for the quenched 30.5 atomic percent specimen. Note the crossing of the 200°K and 300°K isotherms. The 4.2°K isotherm is the same as in Fig. 7.

The disappearance of ferromagnetism is shown in Fig. 8 by the lack of spontaneous magnetization below 200°K; that is, below 200°K, the internal field must be finite to produce a finite magnetization and there is no hysteresis, which would accompany domain wall motion. Using the criterion for ferromagnetism proposed by one of us[14] and by Kouvel,[15] it is seen in Fig. 9 that data at finite fields extrapolate to a finite susceptibility in the limit of zero field for the data below 200°K. The figure of 180°K, for the temperature of disappearance of ferromagnetism, is obtained by the extrapolation of H/σ obtained from Fig. 9 as a function of temperature. This is a normal procedure for ferromagnetic materials above their Curie points, but its use here is somewhat arbitrary.

That all this is associated with antiferromagnetism is brought out by the measurements at 4.2°K. The large

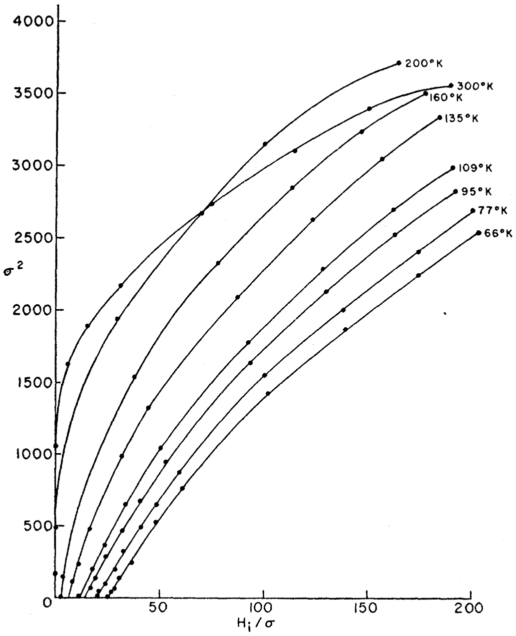

FIG. 9. Analysis of the curves in Fig. 8 in terms of σ^2 and the inverse susceptibility H_i/σ.

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[13] R. C. Hall, J. Appl. Phys. 28, 707 (1957).

[14] A. Arrott, Phys. Rev. 108, 1394 (1957).

[15] J. S. Kouvel, (private communication); see for instance, Kouvel, Graham, and Becker, J. Appl. Phys. 29, 518 (1958).

hysteresis that appears at 4.2°K is shown in Fig. 8. This hysteresis loop is compared with the results for the antiferromagnetic 33 atomic percent sample at 4.2°K in Fig. 7. The isotherms for the other antiferromagnetic specimens are shown in Fig. 6. Note that the coercive force of the 30.5 atomic percent sample is very similar in magnitude to that of the antiferromagnetic specimens. The results for the annealed 30.5 atomic percent sample at 4.2°K are also shown in Fig. 7. The difference between the annealed and quenched samples are clearly quantitative rather than qualitative. These coercive forces are considerably higher than those observed in homogeneous bulk specimens of ferromagnetic materials. To conclude, on the basis of these results, that the 30.5 atomic percent sample changes from ferromagnetic to antiferromagnetic on decrease in temperature may not be entirely justified until confirmed by such additional evidence as neutron diffraction. The fact that a simple model using antiferromagnetic interactions explains the various and diverse phenomena encountered in this system does lend support to this view. The theoretical model is discussed in the accompanying paper.

The effects of the antiferromagnetic interactions are somewhat evident in the behavior of the 28.3 atomic percent alloys. There is a slight decrease in spontaneous magnetization between 77°K and 4.2°K for both annealed and quenched specimens. In addition, there is the appearance of hysteresis at 4.2°K with coercive forces of the order of 10^2 oersteds which is at least 10^2 times the value at higher temperatures. Whether this is intrinsic to the 28.3 atomic percent composition or arises from concentration fluctuations including regions approaching 30 atomic percent is not known. If it is intrinsic, then the hysteresis would have to be associated with ferrimagnetic domain walls rather than antiferromagnetic (compensated ferrimagnetic) domain walls as was the case in the specimen of greater aluminum concentration.

CONCLUSIONS

From these measurements, the following conclusions appear valid and it would seem reasonable to expect a satisfactory model of the magnetic interactions in the iron aluminum system to explain them:

1. Increasing Al content in iron-aluminum alloys leads to a transition from ferromagnetism to antiferromagnetism.
2. A transition range exists in which the stable magnetic state is ferromagnetic at high temperatures and antiferromagnetic at low temperatures.
3. For disordered alloys, the magnetic moment decreases with increasing aluminum content much as would be expected on simple magnetic dilution.
4. A comparison of magnetic moment at any given composition gives for a comparison of the disordered state and the two states of order,

$$\sigma[\text{disordered}] > \sigma[\text{Fe}_3\text{Al}] > \sigma[\text{FeAl}].$$

Note that the moment is not a monotonic function of order.

5. There is a marked increase in parasitic paramagnetism with increasing Al content as the compositions approach the transition range.
6. In general, the striking differences are between the disordered behavior and that of either of the two types of order. The differences between the two types of order are quantitative rather than qualitative.

The anomalies reported here are associated with the ordering of the aluminum and iron atoms. However, it is felt that a detailed mechanistic explanation of these effects will extend considerably the understanding of the general nature of magnetic interactions in metals. In particular, antiferromagnetic interactions in metals are probably more general than previously presumed. A phenomenological interpretation of these results is presented in the following paper.

ACKNOWLEDGMENTS

The authors wish to thank the Metallurgy Department of the Scientific Laboratory of the Ford Motor Company for providing the metallurgical services which were necessary for this work and, in particular, Dr. V. F. Zackay for his helpful cooperation and stimulating discussions.