THE IRREVERSIBILITY OF THE HEUSLER ALLOYS.

BY BRUCE V. HILL.

A LITTLE over a year ago the author, while making some experiments upon specimens of Heusler's magnetic bronzes (of manganese, copper and aluminium) found that these alloys are magnetically irreversible, but in a sense opposite to the irreversibility of the nickel-steels. A brief account of these experiments was published in this journal.[1] If an alloy of iron and nickel, containing up to twenty-seven per cent. of nickel, in the magnetic condition, be heated to from 700° to 750°C. it becomes unmagnetic. On being cooled it does not regain its susceptibility until the temperature has fallen to a point considerably below, in some cases several hundred degrees below, that at which the transformation took place during the heating. When heated a second time the metal remains magnetic until the original temperature of transformation is reached. If, starting at a high temperature with the steel in the unmagnetic condition, we descend to a moderate temperature, reheat to the starting point, cool again to a temperature lower than that reached in the first cooling, reheat and so on, we find that the susceptibility of the metal will, in general, be greater at room temperature, the lower the point to which it has been cooled.

With the magnetic bronzes, on the other hand, cooling to low temperatures, as in liquid air, has very little effect. The susceptibility depends rather upon the temperature from which the specimen has been cooled before the test is made. If heated to a temperature slightly above that of transformation, the susceptibility at room temperature is found to be smaller than when the alloy was first cast. On cooling successively from still higher temperatures the susceptibility at ordinary temperatures increases as the temperature from which the metal has been cooled rises.

It was further pointed out that, inasmuch as certain compounds

[1] PHYSICAL REVIEW, 21, p. 335, 1905.

of manganese possess a higher susceptibility than the corresponding ones of iron, we might expect to obtain the former in a strongly magnetic condition if a suitable alloy could be obtained. The magnetic bronzes are such alloys.

According to the allotropic theory, which seems to be pretty well established, the ferromagnetic metals can exist in several allotropic forms depending upon the temperature. Only one of these forms is magnetic. In the case of many of the mechanical and physical properties, it is impracticable to study a substance at temperatures much above the ordinary. An indirect method must then be employed. When a metal is heated to a high temperature and suddenly cooled, passing in so doing a point of allotropic transformation, this transformation which would have occurred during a slow cooling, is hindered or perhaps almost wholly prevented by the rapid cooling. This is made use of in studying the conditions existing in alloys at high temperatures. In the case of the steels, for example, when the metal has been heated to a high temperature and suddenly cooled, we consider that we have it before us in the condition in which it was at the high temperature. If this were strictly true in all cases we might expect to find the magnetic bronzes magnetic at high temperatures since their susceptibility is greater the more elevated the temperature from which they have just come. Without experiment it is impossible to say whether this will be true or not. We naturally think of these metals as being always unmagnetic above the temperature of transformation. Dr. D. K. Morris found, however, that iron becomes unmagnetic at a temperature between 700° and 750°C, but regains its susceptibility to a slight extent between 800° and 1,000°C. I thought it worth while therefore to test some further specimens of the Heusler alloy in order to determine the magnetic condition of the metal between the points of transformation and fusion, that is between 250° and 1,000°C.

Four alloys were prepared. They were made by melting a copper-manganese alloy with aluminium. An ordinary gasoline furnace was used and the melts made under borax glass as a flux. The copper-manganese was obtained from one of the best firms in the country and was supposed to contain thirty per cent. of manganese. The upper portion of the pig of metal contained a considerable amount of slag. The analyses showed the per cent. of aluminium to be so nearly that of the mixture before melting, while that of the manganese was so much below what was expected, that it is fair to assume that the manganese was burned out in the manufacture of the original copper-manganese alloy. The analyses, for which I am indebted to Professor McFarland of the Department of Chemistry of the University of Kansas, are as follows :

Alloy Number.	Copper.	Aluminium.	Manganese.
1	74	22	4
2	65	34	1
3	65	32.5	2.5
4	63	34.3	2.7

The discoverer of these bronzes, F. Heusler[1] has recently published an account of experiments upon certain alloys rich in copper which are, nevertheless, very highly magnetic and which are malleable.

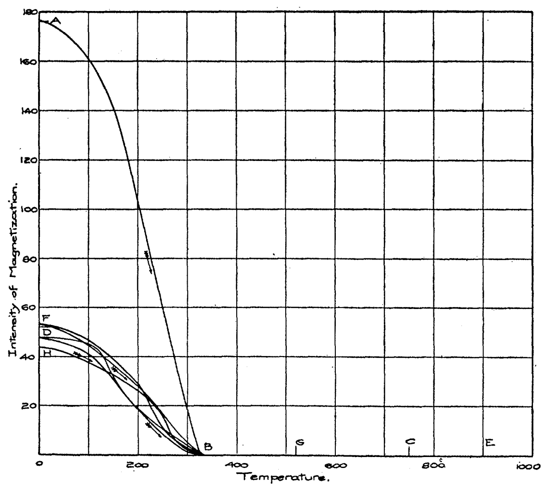
The apparatus used in the testing of my alloys was that described in the previous note upon this subject. The temperatures were measured with a platinum — platinum 10 per cent. rhodium couple. The curves showing the relation between the intensity of magnetization and the temperature for alloy No. 1 during three successive cycles are given in the figure. The field throughout the tests was 38 C.G.S. units. The curves of decreasing intensity, that is of increasing temperature, are indicated by the arrows immediately below them.

Starting at room temperature the intensity, I, is 175. It falls rapidly as the temperature rises, becoming zero at 320°C. The heating was continued up to 750°, the point C in the figure. On cooling the curve returns to the point D where $I = 47$. The tem-

[1] F. Heusler, Beiblaetter, 29, 1238, 1905, or Schriften d. Marburger Gesell. J. A. Fleming and R. A. Hadfield have published tests upon some of these alloys but nothing distinctly new is given.

Professor A. Gray, of Glasgow (Proc. Roy. Soc., Sect. A., 77, p. 256, March 6, 1906), has also tested two specimens of these bronzes finding them like those of the other experimenters.

perature was now raised to 890°C., the point E. This time the return was to F, $I = 53$. Being heated the third time to 520°C. and cooled to room temperature the rod showed an intensity $I = 44$.



Alloy No. 2 was unmagnetic as cast. Pounding or jarring produced no noticeable effect. The rod was then heated successively to 325°, 490° and 810° C., but it remained unmagnetic throughout. When broken in a mortar the fragments were magnetic. It melted at 975° C.

Alloy No. 3 showed a course similar to that of No. 1. When cast the intensity, I, was 123. Transforming at 300° it was heated to 320° and allowed to cool. I attained a value of 128. The second heating was to 500°, after which the intensity at room temperature was 48. The third heating was to 730°, after which the value of I was 41. The fourth heating was to 890°, the intensity becoming $I = 44$ at room temperature.

Alloy No. 4 was tested at room temperature as cast and after being heated to the same series of temperatures as was No. 2. The results were: As cast $I = 41$, after heating to 325° $I = 41$, after heating to 490° $I = 14$ and after heating to 810° $I = 63$.

The data for these four alloys, as well as for the one described in the previous paper, which will be called No. 5, are brought together in the table. $H = 38$ for the first four and 85 for the fifth.

Alloy No. 1	As cast	520°	750°	890° C.	
	$I=175$	44	47	53	
Alloy No. 2	As cast	325°	490°	810° C.	
	$I=0$	0	0	0	
Alloy No. 3	As cast	320°	500°	730°	890° C.
	$I=123$	128	48	41	44
Alloy No. 4	As cast	325°	490°	810° C.	
	$I=41$	41	14	63	
Alloy No. 5	As cast	368°	500°	650°	850° C.
	$I=311$	267	27	90	155

From these results we see that, in general, the susceptibility of the alloy at room temperature decreases as the temperature to which it has been heated rises, up to a certain point, beyond which the intensity increases again. Nos. 4 and 5 show these characteristics more pronouncedly than the others. All the alloys, with the possible exception of No 3 in the first cycle, show them however, though in the first four, those of low manganese content, they are not so marked.

We see also from the figure that the alloys do not regain their magnetic properties at the higher temperatures. If we assume that the molecular state of the metal is in great measure that naturally stable at the higher temperature due to the retardation or non-occurrence of certain allotropic changes, then we must suppose that there are different molecular arrangements of the metals constituting the alloy at different temperatures. The relations existing at high temperatures allow the transformation of the manganese from the *beta* to the *alpha* state to take place more completely than in those molecular groupings existing at lower temperatures. The temperature-intensity cycles are irreversible, but cannot be repeated as with the irreversible nickel-steels. This is probably due, in part at least, to the fact that the time during which the metal is held at a certain temperature plays a great part in determining the nature of its subsequent properties, as shown originally by Messrs. Haupt and Stark[1] and by E. Gumlich[2]. By casting a number of rods from the same crucible, and thus avoiding the repeated heating of the same rod, some additional information might be gained.

[1] Marburger Schriften, Bd. 13, 1904.
[2] Ann. d. Phys., 16, p. 535, 1905.

While the experiments upon these bronzes may have brought us very little nearer to an understanding of the phenomenon of magnetism, they seem to make it evident that we have here to do with a molecular and not an atomic property.

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