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Intermetallic Compounds of Iron and Cobalt with Gadolinium

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Thermomagnetic and X-ray measurements on the system gadolinium-cobalt indicate that antiferromagnetic interactions exist in agreement with the work of Nesbitt[1] *et al.*

Only two intermetallic compounds, $GdFe_2$ and $GdFe_5$ were found in the gadolinium-iron system instead of the seven reported by Vickery[2] *et al.*

Magnetic measurements on the compounds $GdCo_5$, $GdFe_5$ and $Gd(Co_xFe_{1-x})_5$ indicate that the spin arrangements change from Gd↑ 5Co↓ to Gd↑ 2Fe↓ 3Fe↓ as x varies from 1 to 0.

Introduction

A search for new materials having both high saturation magnetizations and Curie temperatures has lead to a study of the alloys of gadolinium with the transition metals. It was hoped that the gadolinium atom could be coupled ferromagnetically with the moment of a transition element to provide a saturation magnetization greater than that of iron or to raise the Curie point of gadolinium without seriously degrading the large saturation magnetization.

Earlier work on the system gadolinium-cobalt[1,3,4,5] indicated that antiferromagnetic interactions were operable between the gadolinium atoms and the cobalt atoms but there was lack of good agreement between the experimentally determined values of the magnetic moment and those calculated assuming complete antiferromagnetic interactions between the gadolinium and cobalt atoms as shown by Table I.

In the case of the system gadolinium-iron the agreement between the calculated and experimental moments as determined by various investigators[1),2)] appeared to be quite satisfactory. The agreement between investigators as to the number of phases present in this system however was much less satisfactory. Nesbitt[1] *et al.* reported two, or possibly three intermetallic compounds in this system with antiferromagnetic exchange couplings between the iron and gadolinium. Vickery[2] *et al.* reported the existence of seven intermetallic compounds of iron and gadolinium and also indicated a very sharp decrease in magnetization with the addition of small percentages of iron to gadolinium.

Table I

Compound	μ_0(expt'l.)	μ_0(calc.)
$GdCo_5$	0.89[1]	1.45
	2.00[3]*	
	1.90[4]	
$GdFe_5$	4.06[1,2]	4.00

Numbers refer to references.

* Extrapolated from 4°K; all others extrapolated from 77°K.

It was the purpose of this work to examine these systems in more detail and to see if a better understanding of the spin arrangements in the compounds could be developed.

Experimental

All samples were prepared by arc melting elemental metals having purities of 99.8% or better. Each specimen in the form of a button was melted at least twice under a reduced pressure of helium to insure homogeneity. X-ray and magnetic measurements were made from different portions of a button to insure homogeneity in the samples prepared by this procedure. The buttons were filed with a tungsten-carbide file in a dry box under an atmosphere of dried argon to produce 80 mesh powder for X-ray and magnetic measurements. Several complete series were prepared and measured. Chemical analyses are not yet available on these samples, but previous experience with the gadolinium-cobalt samples prepared, in the same manner, indicated that there was no appreciable loss of material.

The magnetic measurements were made on a vibrating sample magnetometer, after the design of Foner[6], that had been modified for

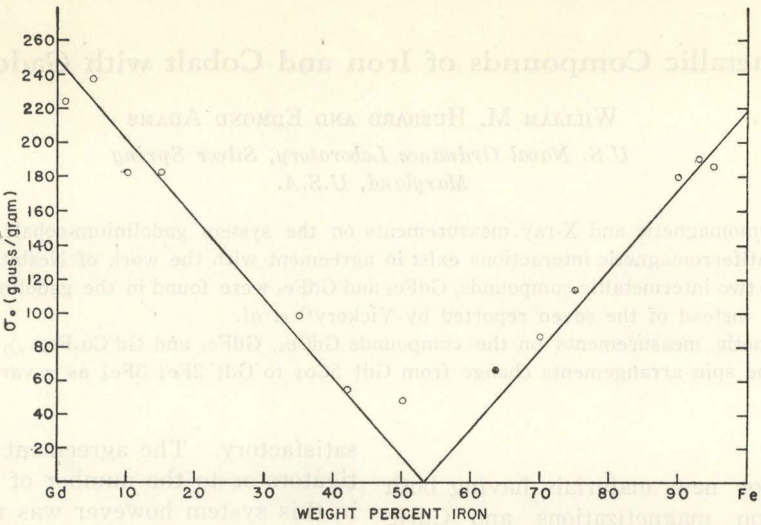

Fig. 1. Saturation magnetization per gram of iron-gadolinium alloys at 0°K as a function of composition.

high temperature measurements in an argon atmosphere with continuous recording of the magnetic moment versus temperature. X-ray measurements were made on a Norelco diffractometer using filtered molybdenum radiation and a Krypton-filled geiger counter.

Results and discussion

The results of the investigation of the gadolinium-iron system (shown in Fig. 1) are in essential agreement with Nesbitt[1] *et al.* in regards to the effect on the magnetic moment of adding iron to gadolinium. Only two intermetallic compounds were found in the system, *i.e.*, $GdFe_2$ and $GdFe_5$ whose parameters and structure types are listed in Table II. No anomalous decrease in magnetization at small iron concentrations was noted except for samples that were ground in air and had presumably reacted with the atmosphere. It was found that the phase $GdFe_5$ was difficult to prepare in pure form and that samples giving a diffraction pattern of a single phase consistently gave higher values (4.97 Bohr magnetons per unit cell) for saturation magnetization than could be accounted for by antiferromagnetic spin arrangements (4.00 Bohr magnetons per unit cell).

Table II

Compound	Structure Type	Lattice Parameters
$GdFe_2$	C15	$a=7.394$Å
$GdFe_5$	$D2_d$	$a=4.83$Å, $c=4.13$Å

Measurements were made on single crystal samples of $GdCo_5$ in an effort to resolve the discrepancy between predicted and measured values that was mentioned previously. The value obtained for the number of Bohr magnetons per unit cell at O°K (extrapolated from data at 77°K) was 1.43. This is in excellent agreement with the predicted value of 1.45 assuming 7.10 Bohr magnetons per atom of gadolinium, 1.71 Bohr magnetons per atom of cobalt and antiparallel ordering between gadolinium and cobalt spins.

The excellent agreement between the experimental values obtained in this study and the predicted values in the case of $GdCo_5$

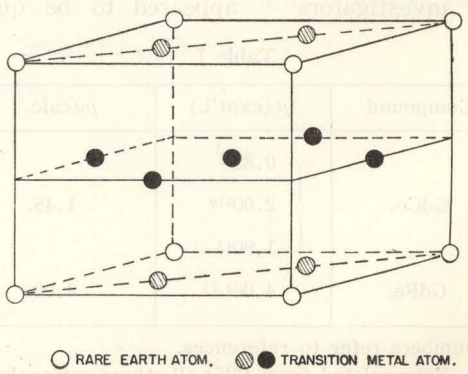

Fig. 2. Uuit cell $D2_d$ structure.

and the poor agreement obtained in this study in the case of $GdFe_5$ led to a reappraisal of the spin model as follows. The crystal structure of the GdX_5 compounds is depicted in Fig. 2. In this structure there are three different atomic sites available; those occupied by the rare earth atoms (hereafter called "*A*" sites), those occupied by transition metal atoms in the base plane (hereafter called "*B*" sites) and those occupied by transition metal atoms in the midplane (hereafter called "*C*" sites). These different sites can give rise to three magnetic sub-lattices with spin orientations dependent on the different interaction parameters. In such a system one might observe transitions in the slope of the curve showing moment versus temperature as the sub-lattice magnetizations vary. Three of these transitions are observable in the data of Nesbitt[1),3)] *et al.* for $GdCo_5$. One transition occurs at approximately 80°K, one at approximately 300°K and the third at approximately 1000°K (T_c).

For the case of three sub-lattices it is possible that the spin orientations in $GdFe_5$ might be different than in $GdCo_5$ because of different interaction parameters. A possibility consistent with the data for $GdFe_5$ in this work is the structure in which the transition metal atoms on the "*B*" sites have their moments directed parallel to those on the "*A*" sites, while those on the "*C*" sites have their moments directly antiparallel to those on the "*A*" sites. To test for the applicability of this spin model, a series of ternary compounds of iron, cobalt and gadolinium

Table III. Magnetic moments (Bohr magnetons / unit cell), Curie temperatures, and proposed spin configurations of $Gd(Co_xFe_{1-x})_5$ compounds.

X	μ_0 (expt'l.)	Proposed Spin Configurations			μ_0 (calc.)
		A Site	*B* Site	*C* Site	
0	4.97	Gd↑	2Fe↑	3Fe↓	4.88
0.2	5.46	Gd↑	2Fe↑	2Fe↓ Co↓	5.39
0.4	5.01	Gd↑	Fe↑ Co↑	2Fe↓ Co↓	4.88
0.5	3.44	Gd↑	Average ↕	Average ↕	3.68
0.6	3.13	Gd↑	2Co↓	2Fe↓ Co↓	2.47
0.8	2.21	Gd↑	2Co↓	Fe↓ 2Co↓	1.96
1.0	1.43	Gd↑	2Co↓	3Co↓	1.45

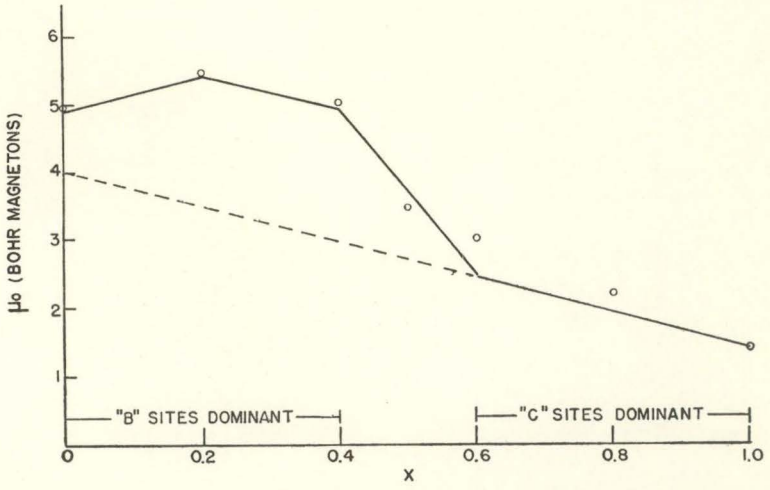

Fig. 3. Magnetic moments per unit cell (extrapolated from 77°K) for $Gd(Co_xFe_{1-x})_5$ compounds (H=12,500 oersted).
○ Experimental values.
—— Calculated for proposed spin arrangements.
- - - Calculated for antiparallel spin arrangements.

were prepared, corresponding to the general formula $Gd(Co_xFe_{1-x})_5$.

The proposed spin arrangements were arrived at by considering a unit cell and imagining that one were going to substitute an atom at a time for the transition atoms in either the "*A*" or "*B*" sites. If the substitution of an iron atom for a cobalt atom had no effect on the spin arrangements, one would expect the moment per unit cell of the assembladge to increase linearly with a slope of 0.5 Bohr magnetons per unit cell as X varied from 1 to 0 (dashed line in Fig. 3). If the substitution of an iron atom for a cobalt atom alters the spin arrangements however, then there are many possibilities from which to choose. The arrangements shown in Table III are those for which the calculated moment per unit cell agreed most closely with the experimental data (solid line in Fig. 3). It should be recognized that the solid curve of Fig. 3 is but a representation of these assumed arrangements; whereas in actuality, the curve representing an exact calculation would probably be without breaks but would involve even further assumptions as to the stability of the various spin arrangements and some form of statistical averaging of these arrangements. The purpose of the proposed scheme is simply to indicate that one must assume a more complicated arrangement than that in which all the spins of the transition atoms are aligned antiparallel to the spins of the rare earth atoms in order to explain the experimental data for these systems.

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