MAGNETIC ORDERING IN DILUTE GOLD-IRON ALLOYS

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Borg, Booth, and Violet[1] have recently described an investigation using the Mössbauer effect of magnetic ordering in gold-rich AuFe alloys having a face-centered cubic structure, and Henry[2] has observed magnetic remanence and has measured the approach to saturation in strong magnetic fields in an alloy $Au_{0.95}Fe_{0.05}$. In both papers the authors conclude that long-range magnetic coupling effects dominate in such dilute alloys. The purpose of this note is to point out objections to this conclusion, and to present previously unpublished data which support an alternative view that the dominant effects are of short range.

Curie temperatures, obtained by Borg et al. from measurements of the Mössbauer effect and by ourselves from measurements of the bulk magnetization of solid specimens, are plotted against the iron concentration in Fig. 1. Our magnetizations (σ) were measured in a range of applied field strengths (H) up to 17 kOe, at different temperatures. The Curie temperatures were determined by plotting graphs of (H/σ) against σ^2 for various constant temperatures, and finding by linear interpolation the Curie temperature θ at which the linear (H/σ) against σ^2 graph passes through the origin. This method was first applied to homogeneous ferromagnetic materials by Belov and Goryaga[3] and by Arrott,[4] and in the present case the error in the Curie temperatures so defined is less than 2°K.

In addition, we have measured the magnetization, at temperatures down to 4.2°K and fields up to 20 kOe, of other alloys containing 11.1, 7.0, 6.1, and 5.1 atomic percent of iron (by analysis). The value for the 5.1% alloy at 4.2°K and 20 kOe agrees with that given for 5% by Henry; and otherwise the general shape of the (σ, H) curves obtained for these alloys is qualitatively similar to that found by Henry,[2] within the limits of our smaller range of field strengths. The other outstanding features of our measurements are as follows:

(1) The graph of H/σ against σ^2 for the 11.1% alloy at 4.2°K is linear, and its intercept on the (H/σ) axis is positive and relatively large, showing that the magnetization isotherm at 4.2°K is characteristic of a temperature well above the bulk Curie temperature. And linear extrapolation of the corresponding data obtained between 4.2°K and 11°K suggests that no finite Curie temperature exists for this alloy. The data obtained for the other alloys with lower iron contents at 4.2°K were indicative of a state well above any Curie temperature that might exist. We therefore conclude that alloys containing no more than 11.1 atomic percent of iron exhibit no long-range ferromagnetism.

(2) Extrapolation of the magnetization of the 11.1% alloy at 4.2°K to infinite field (i.e., to $1/H = 0$) shows that the saturation magnetization σ_s at this temperature is equivalent to 1.15 magnetons per iron atom.

(3) For the 11.1% alloy at 4.2°K the dependence of magnetization on externally applied field calculated from

$$\sigma/\sigma_s = (\coth a - 1/a), \qquad (1)$$

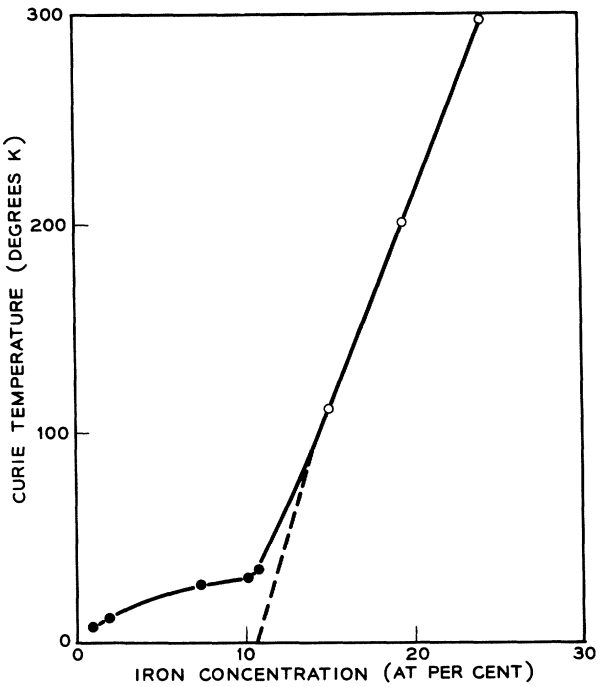

FIG. 1. Curie temperatures of gold-rich solid solutions of iron in gold as a function of composition. Open circles are from direct magnetization measurements, and closed circles are from the Mössbauer measurements.[1] The dashed line is an extrapolation of the magnetization data, drawn so as to be consistent with the bulk Curie point of the 11% alloy.

where $a = MH/kT$, agrees well with the measurements, after fitting at a single point.

Similarly, if we plot (H/σ) against σ^2 for values taken from Fig. 1 of Henry's paper[2] we find that his 5% alloy is also well above its bulk Curie temperature at 4.2°K. However, since in this case the next higher temperature for which there are measurements is as high as 77.4°K, it is not possible to be certain that a finite Curie temperature does not exist. Henry's saturation magnetization at 4.2°K appears to be about 0.75 magnetons per iron atom. Also, his curve for 4.2°K fits well with one calculated from Eq. (1).

If we follow the suggestion of Borg et al.[1] and assume to a first approximation that the hyperfine field at the Fe nucleus as measured in their experiments is proportional to the spontaneous magnetization associated with the iron atoms, we may use their data to estimate the spontaneous magnetizations of their alloys. These are compared in Fig. 2 with our direct magnetic measurements for less dilute alloys, and with values obtained for the 11.1% and 5% alloys by extrapolating the magnetization at 4.2°K to $1/H = 0$.

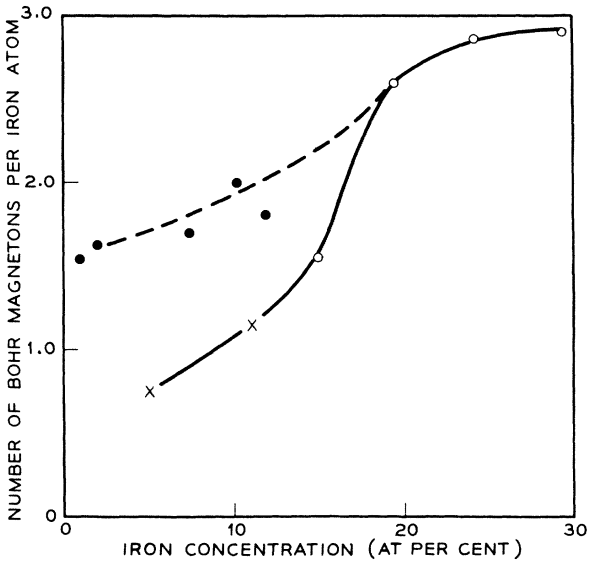

FIG. 2. Magneton numbers per iron atom plotted against alloy composition. The open circles are from extrapolation of the spontaneous magnetization to $T = 0$; the crosses are from extrapolation to $1/H = 0$ of magnetizations measured at a temperature well below the short-range Curie point given in Fig. 1 (lowest point from Henry[2]); and the closed circles are from Mössbauer measurements,[1] assuming universal proportionality of hyperfine field acting on iron atoms and magnetic moment.

We believe that the experimental observations are consistent with the following basic model.

As the iron content of the solid solution is reduced, long-range magnetic order which characterizes a normally ferromagnetic metal ceases at a little over 10% of iron. The dominant magnetic effects at lower iron contents are those of a superparamagnetic array[5] of short-range ferromagnetic regions possibly associated with statistical fluctuations of the iron concentration in the alloy. These regions interact only weakly or not at all with each other.

Since the observations using the Mössbauer effect are primarily concerned with short-range magnetic effects in the vicinity of the absorbing iron nucleus, and not with long-range order, the Curie temperatures given by Borg et al. might be expected to be the intrinsic (short-range) Curie points within the ferromagnetic regions. They are therefore still finite at compositions below which the bulk Curie temperatures of the whole specimens have become zero. In the less dilute alloys, where there is long-range magnetic order, the "Mössbauer" and the "bulk" Curie temperatures would be expected to coincide, as they do in pure iron.[6] It would thus be useful if the Mössbauer observations could be extended to higher iron concentrations. The break in Fig. 1 near 10% clearly indicates differences in the mechanism of magnetic ordering between the more dilute alloys and the others. The elements in a superparamagnetic array would orient themselves continuously with respect to the applied magnetic field and not in discrete steps, and would therefore obey the form of Eq. (1), as is indeed observed experimentally here.

The only apparent difficulty with this model seems to lie in the differences between the Mössbauer and the magnetic determinations of the saturation moment per iron atom, shown by the two lines in Fig. 2. A possible explanation is that it may be wrong to assume proportionality between hyperfine field and magnetic moment, and that the upper line of Fig. 2 should really lie much lower. Alternatively if in some cases up to half the iron present were to act freely, in such a way as not to cooperate in the short-range regions, a paramagnetic component in the Mössbauer spectrum would be expected; but this is absent.[1]

Henry's observation of remanence does not seem to be a serious obstacle to the applicability of this model. The remanent magnetization

is about 2.7% of saturation, and values of this magnitude are often observed just above the Curie point of a normal ferromagnetic material, where short-range magnetic order persists.

A fuller treatment of the magnetic properties of these alloys, and also of the other systems PdFe, PtFe, PtCo, and PdNi, where the effects of long-range interactions do certainly occur, is being prepared for publication.

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ANALYSIS OF THE EXPERIMENTAL EVIDENCE FOR THE EXISTENCE OF A ^{4}H STATE WITH $T=2$†

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In this note we shall be concerned with the interpretation of results recently obtained by Argan et al.[1] on the process

$$\gamma + {}^4\mathrm{He} \rightarrow \pi^+ + \mathrm{T} + n \qquad (1)$$

whose characteristics have previously been interpreted in terms of a low-lying resonant state in the n-T system, possibly a $T=2$ state which decays electromagnetically.[2] We shall conclude that the outstanding features observed for this process can be understood without requiring the hypothesis of a resonant state, or even of any n-T final-state interaction. This analysis may be of use in clarifying the background of the current searches for a tetraneutron and other manifestations of a four-nucleon $T=2$ state.

We first review the evidence for states other than the α particle in the four-nucleon system:

^{4}He: No bound excited state of ^{4}He has been found in a recent search.[3] It has been suggested, however, in the interpretation of n-^{3}He and p-T interactions that a $^1S_0$ resonance exists between the p-T and n-^{3}He thresholds.[4]

^{4}H: No bound ^{4}H was found in recent searches.[5,6] However, Nefkens[7] has reported results interpretable in terms of the β decay of a stable ^{5}H and Gol'danski has made a shell-model–pairing argument that the existence of such a state of two neutrons bound to a triton would imply a bound or low-lying resonant n-T state as well.[8] Other experiments[9] appear to be in disagreement with that of Nefkens at the moment and the situation remains to be clarified.

Low-energy n-T scattering results peaking at 3-MeV c.m. energy[10] have been interpreted theoretically in terms of a model by Bransden and Robertson[11] which involves negative S-wave phase shifts and positive but nonresonant P-wave phase shifts. A resonance or bound state would also be apparent in a study of the proton spectrum from the reaction T(d,pn)T for which only the neutron spectrum has been published.[12]

No experimental evidence exists at present for other four-nucleon states.

In the experiment of Argan et al., the reaction (1) was observed for a bremsstrahlung beam from the Frascati 1-BeV electron synchrotron passing through a helium diffusion cloud chamber. The following distributions were measured: (a) $\theta(\vec{\mathrm{T}}, \vec{\mathrm{T}}+\vec{n})$, the angle between the T and the T$+n$ directions in the c.m. frame for an assumed 250-MeV incident γ ray [Fig. 1(a)]; (b) $\varphi(\vec{\mathrm{T}}, \vec{\pi})$, the projection of the supplement of this angle onto the plane perpendicular to the beam direction [Fig. 1(b)]; (c) the variable $A = m_T v(\mathrm{T}+n) \sin\theta(\vec{\mathrm{T}}, \vec{\mathrm{T}}+\vec{n})$ evaluated in the c.m. frame [Fig. 1(c)], where $v(\mathrm{T}+n)$ is the velocity of the center of mass of (T$+n$); (d) $\theta(\vec{\pi}, \vec{\gamma})$, the distribution of the π direction relative to the beam direction in the lab frame [Fig. 1(d)].[13]

No Q-value spectrum was reported for the $(n+\mathrm{T})$ system. The peaking at small angles in the actual and projected $\theta(\vec{\mathrm{T}}, \vec{\mathrm{T}}+\vec{n})$ was interpreted as suggesting that the neutron and the triton tend to go off in the same direction because of the existence of a low-lying state. By assuming the peaking in the spectrum to be substantially due to configurations with prependicular T and π momenta in the (T$+n$) c.m. system, a resonant energy of about 4 MeV was deduced.